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Review

Recent advances in electrocatalytic membrane for the removal of micropollutants from water and wastewater

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SUMMARY

The increasing occurrence of micropollutants in water and wastewater threatens human health and ecological security. Electrocatalytic membrane (EM), a new hybrid water treatment platform that integrates membrane separation with electrochemical technologies, has attracted extensive attention in the removal of micropollutants from water and wastewater in the past decade. Here, we systematically review the recent advances of EM for micropollutant removal from water and wastewater. The mechanisms of the EM for micropollutant removal are first introduced. Afterwards, the related membrane materials and operating conditions of the EM are summarized and analyzed. Lastly, the challenges and future prospects of the EM in research and applications are also discussed, aiming at a more efficient removal of micropollutants from water and wastewater.

INTRODUCTION

Micropollutants such as pharmaceutical, personal care products, endocrine disruptors, biocides, and polyfluoroalkyl substances are a group of emerging pollutants that pose a severe threat to the environment (Alsbaiee et al., 2016; Carpenter and Helbling, 2018). It has been reported that a long-term exposure to trace concentrations of micropollutants can impose adverse effects on wildlife and human health (Barbosa et al., 2016). However, the traditional wastewater treatment plants (WWTP) designed to remove conventional pollutants (such as COD and nutrients) is less effective for the removal of emerging micropollutants (Stamm et al., 2015; Cai and Hu, 2018), leading to micropollutants frequently detected in waters with the concentrations ranging from ng/L to μg/L (Kumar et al., 2019; Li et al., 2020a; Yu et al., 2020a). Therefore, it is urgent to develop more effective technologies for an efficient removal of micropollutant.

Microfiltration (MF) and ultrafiltration (UF) technologies have been widely used in the water and wastewater treatments due to the advantages of high effluent quality, small footprint, and ease of automation control (Owen et al., 1995; Park et al., 2017). However, both technologies have poor rejection of micropollutants due to the limitation of membrane pore size (Peters, 2010). Moreover, micropollutants may accumulate on the surface or in the pores of microfiltration and ultrafiltration membranes, which eventually leads to membrane fouling and permeability loss (Wang et al., 2014; Miller et al., 2017). Although the accumulated micropollutants could be removed from the membrane surface or pores through physical and/or chemical cleaning processes, the operation might cause a "secondary pollution" (Wang et al., 2021). Furthermore, the cleaning processes may cause irreversible damage to the membrane materials and thereby shorten the membrane lifetime.

To overcome these challenges, MF or UF has been combined with the electrochemical advanced oxidation process (EAOP), as EAOP is considered as an efficient and environmentally friendly technology for the removal of micropollutants in water and wastewater (Chen et al., 2021; Ren et al., 2021). This hybrid technology is also termed electrocatalytic membrane (EM) technology, in which the direct electron transfer and the generated strong oxidizing species are two main mechanisms for the removal of micropollutants. In this system, the mass transfer from the bulk solution to the reactive surface (i.e., membrane surface) is also enhanced through the filtration process (Trellu et al., 2018a). Compared with the traditional MF and UF processes, the advantages of the EM include production of high-quality effluent, mitigation of membrane fouling, and elimination of secondary pollution.

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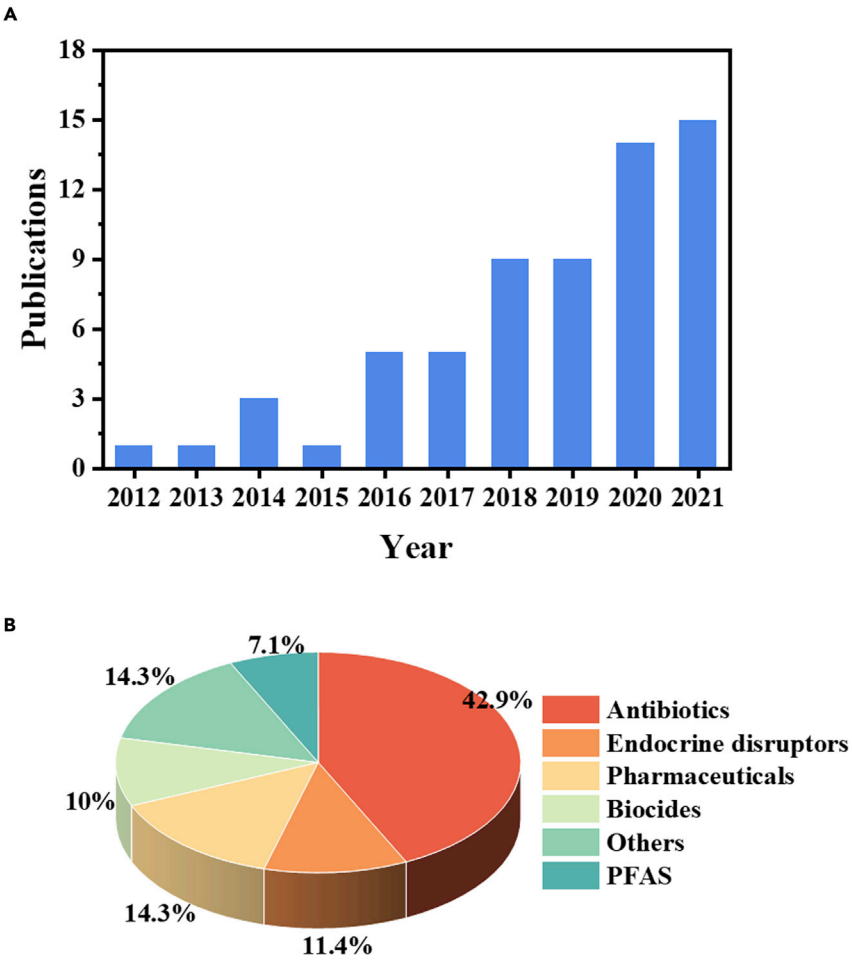

Figure 1. Publications regarding EM for micropollutant removal

(A) The number of publications involved in EM for micropollutant removal since 2012; (B) the existing applications of EM in micropollutant removal. The data were collected from the database of Web of Science updated to November 2021.

Despite that the concept of solid/liquid separation process of electro-filtration had been proposed in 1977 (Henry et al., 1977), the EM has only attracted increasing attention in the past decade but a rapid progress has been made in the use of EM to remove micropollutants (Liu et al., 2015; Zhao et al., 2020). Based on the database of Web of Science (Figure 1), the number of annual publications on EM for micropollutant removal increases rapidly. These EMs are widely used in micropollutant removal, such as antibiotics, endocrine disruptors, pharmaceuticals, biocides, and polyfluoroalkyl substances. Those studies have laid the groundwork for the application of EM in micropollutant removal. However, a comprehensive review on the recent advances in EM for micropollutant removal is still lacking. Therefore, we aim to consolidate the recent advances in EM for micropollutant removal from the standpoints of mechanisms, performance, and key operating conditions. The review is organized as follows: the main mechanisms of EM to remove micropollutants are firstly introduced. Performance and operating conditions of EM are systematically analyzed based on the literature database. Finally, the technical challenges and future development prospects of EM toward a more efficient removal of micropollutants are discussed.

MECHANISMS OF ELECTROCATALYTIC MEMBRANE FOR MICROPOLLUTANT REMOVAL

Electrosorption

Generally, the main mechanisms of EM for the micropollutant removal include (1) electrosorption, (2) electrochemical oxidation, and (3) membrane filtration. Electrosorption is a physicochemical sorption process (Figure 2A) that does not involve electron transfer reaction. Diffusion, electromigration, and convection can also occur in the EM, and electromigration can promote the selective migration of charged micropollutants

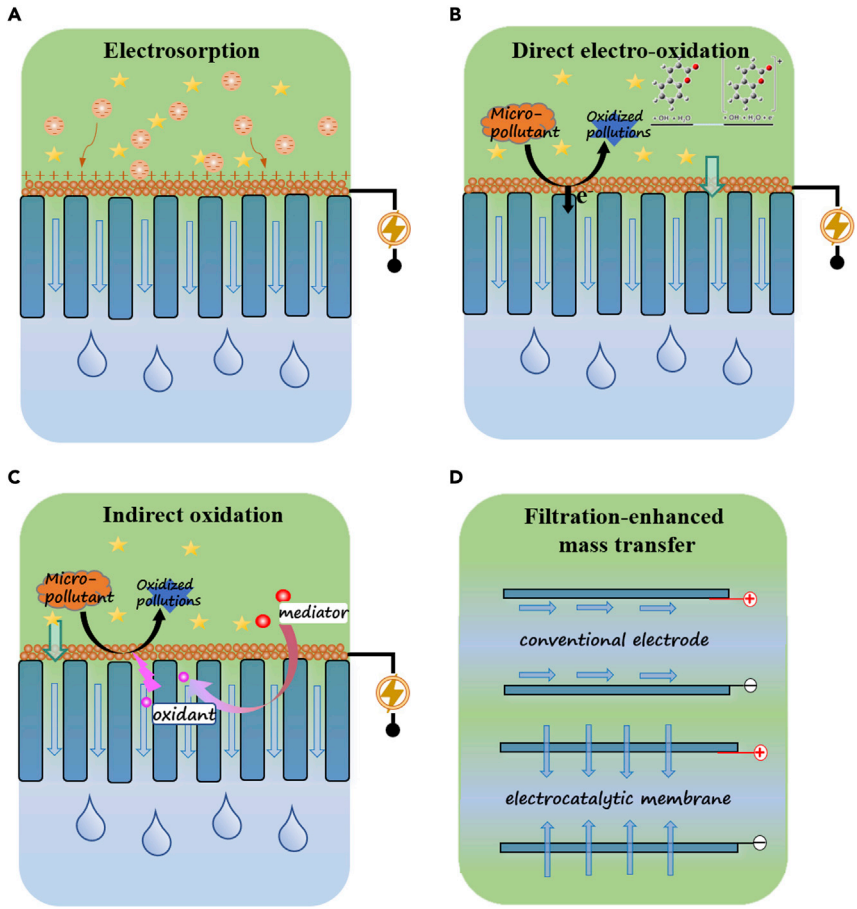

Figure 2. Schematic of micropollutant removal mechanisms in EM

(A) Electrosorption, (B) direct electro-oxidation, (C) indirect oxidation, and (D) filtration-enhanced mass transfer process.

to the oppositely charged electrode for electrosorption (Lissaneddine et al., 2022). The electrostatic interaction between micropollutants and the surfaces of porous electrode materials is one of the key factors for the adsorption and removal of micropollutants, whereas the removal efficiency depends largely on the characteristics of electrode materials (e.g., specific surface area, pore size, surface electronic structure, etc.) and the ionization characteristics of micropollutants (Foo and Hameed, 2009). It is reported that the electrosorption contributes to the removal of negatively charged micropollutants such as PFASs, antibiotics, and drugs containing acidic groups in the anodic electrocatalytic membrane processes (Zhou et al., 2017; Radjenovic et al., 2020).

Electrochemical oxidation

Electrochemical oxidation is the dominant mechanism for the micropollutant removal in the electrocatalytic membrane-based processes. Electrochemical oxidation processes can be further classified into (1) direct electro-oxidation and (2) indirect oxidation processes (Figures 2B and 2C) (Wang et al., 2021). It is worth noting that the electrochemical oxidation efficiency of micropollutants in EM is higher than that in the conventional flat plate electrode, which is mainly attributed to the filtration-enhanced mass transfer in EM (Figure 2D) (Le et al., 2019).

Direct electro-oxidation

Electrons are directly transferred from the micropollutants to the electrocatalytic membrane surface in the direct electro-oxidation. In this process, the micropollutants are directly oxidized to intermediates and ultimately to CO_2 and H_2O. Direct electro-oxidation is theoretically possible at low potentials. For instance, Zhou et al. showed that pyrrole was removed mainly by direct oxidation in graphite-RuO_2-MWCNTs filter at

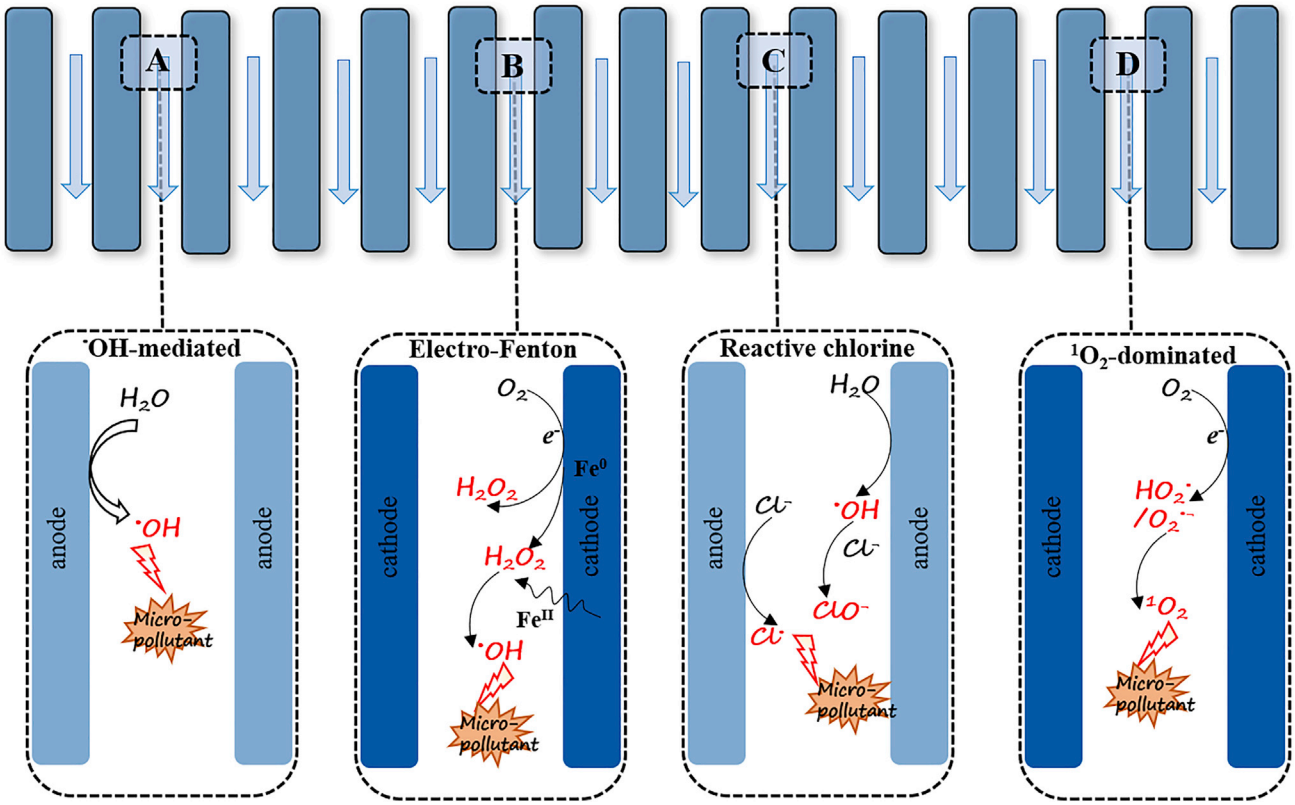

Figure 3. Schematic illustration of indirect electrooxidation processes

(A) ·OH-mediated oxidation, (B) electro-Fenton process, (C) reactive chlorine-mediated oxidation, and (D) $^1O_2$-mediated oxidation.

0.3–1.2 V (lower than the oxygen evolution potential of 1.42 V), whereas the contribution of indirect oxidation became significant when the anodic potential was higher than 1.2 V (Zhou et al., 2017). However, the removal of micropollutants by direct oxidation is usually a slow process because the reaction rate is low at low potentials (Panizza and Cerisola, 2009; Wang et al., 2021). Recent study, however, reported that using noble metals such as Pd could achieve a direct anodic oxidation with high electrochemical reaction rate. For example, Huang et al. prepared a Ti_4O_7 electrode loaded with amorphous Pd, and it was found that the presence of Pd-O bonds enhanced the electron transfer rate and thus achieved the direct oxidation of perfluorooctanoic acid (PFOA) (Huang et al., 2020).

Indirect oxidation

Indirect oxidation of micropollutants relies on strong oxidizing species (e.g., ·OH, Cl·, and $^1O_2$) generated in the electrocatalytic membrane system (Figure 3), whereas hydroxyl radical (·OH) is considered to be a dominant reactive oxygen species to remove micropollutants in electrocatalytic membrane processes. Feng and Johnson reported that adsorbed ·OH was generated by water electrolysis reactions, which could oxidize micropollutants through an O-transfer reaction at high anodic potential (Feng and Johnson, 1990). The removal efficiency of micropollutants by hydroxyl radical-mediated oxidation depends on the membrane material and electrode potential. According to the literature, when the electrochemical membrane is used as an anode, the membrane materials can be classified into "active" and "nonactive" ones (Panizza and Cerisola, 2009). "Active" membrane materials, such as carbon, graphite, IrO_2, and RuO_2, have a low oxygen evolution overpotential, and the generated ·OH can further react with the active anode, leading to a partial oxidation of micropollutants. "Nonactive" anodes, such as antimony-doped tin oxide and Ti_4O_7, typically have a high oxygen evolution overpotential, which is capable of directly mineralizing micropollutants to carbon dioxide. The oxygen evolution potential (OEP) of "nonactive" anode (e.g., Ti_4O_7, 2.2–2.7 V versus SHE) is commonly higher than that of the "active" anode (e.g., carbon, 1.7 V) (Martínez-Huitle et al., 2015).

When the electrochemical membrane is used as a cathode, $^{\cdot}OH$ can be generated indirectly via electro-Fenton reaction (Jiang et al., 2018). In the electro-Fenton process, dissolved oxygen is reduced to H_2O_2 through two-electron oxygen reduction, and the produced H_2O_2 further reacts with Fe(II) to form $^{\cdot}OH$. The yield of H_2O_2 and the dosage of Fe(II) are crucial for the generation of $^{\cdot}OH$. The production of H_2O_2 can be enhanced by using a 3D flow carbon-based cathode (Zhang et al., 2013; Ren et al., 2016; Ma et al., 2016). Compared with the traditional Fenton process, electro-Fenton process requires a lower Fe(II) dosage because a portion of Fe(II) can be generated by electro-reduction of Fe(III) on the membrane (Jiang et al., 2018; Trellu et al., 2020). If the stainless steel wire mesh is used for the cathode, Fe(II) can be generated *in situ* through a Fe(III)/Fe(II) cycle on the surface of the membrane and then reacts with H_2O_2 to realize the efficient production of $^{\cdot}OH$ (Zheng et al., 2017).

When chloride ions exist in wastewater, the reactive chlorine species (RCS), such as chlorine radicals ($Cl^{\cdot}$, $Cl_2^{\cdot-}$) and free chlorine (Cl_2, HClO, ClO^-), can be generated by anodic oxidation of chloride ions on the anode (Cho et al., 2014). Due to the favorable electrocatalytic properties for chlorine evolution, the platinum or metal oxides (e.g., RuO_2, IrO_2) is commonly used for the preparation of *in situ* active chlorine electrode (Martínez-Huitle and Panizza, 2018). It is reported that active chlorine-mediated oxidation can be used to remove a variety of micropollutants, including sulfamethoxazole (Chi et al., 2022), sulfamethazine (Song et al., 2022), paracetamol (Zhang et al., 2022), etc. However, toxic chlorine-organic derivatives (such as chloroform) and chlorine-oxygen by-products (such as ClO_2^-, ClO_3^-) are also simultaneously generated in some cases (Martínez-Huitle and Brillas, 2009).

Singlet oxygen ($^1O_2$), a nonradical derivative of oxygen, is one of the strongest reactive oxygen species. $^1O_2$ is mainly generated from electrically exciting precursors (e.g., $O_2^{\cdot-}$ and H_2O_2) via superoxide-mediated chain reaction (Zhao et al., 2020) and cathodic activation of persulfate (Cheng et al., 2017). Recent studies on EM have shown that singlet oxygen ($^1O_2$) can effectively remove micropollutants including sulfamethoxazole, carbamazepine, nitrobenzene, diclofenac, and tetracycline due to its high reactivity and excellent selectivity (Liu et al., 2019; Ossola et al., 2021).

Filtration-enhanced mass transfer

The conventional electrode oxidation process is generally operated in batch or flow-by mode, resulting in a thick diffusion boundary layer (~100 μm) (Chaplin, 2014). In this process, the removal efficiency of micropollutants is limited by the diffusion rate of pollutants to the electrode surface. According to the steady-state equilibrium between the convection and diffusion, the thickness of the diffusion boundary layer is theoretically smaller than the membrane pore radius, and convection enhances mass transport of micropollutants from the bulk solution to the electrode surface in the flow-through mode, leading to an enhanced mass transfer rate (Liu and Vecitis, 2012; Trellu et al., 2018a). For instance, Chen at al. showed that removal efficiency of 2-methyl-4-iso-thiazolin-3-one in the flow-through mode was 10.3 times higher than that of the flow-by mode using an electrocatalytic membrane, and such fast kinetics was primarily attributed to the enhanced mass transfer by the convection through the membrane in the flow-through mode (Chen et al., 2020b).

APPLICATIONS OF ELECTROCATALYTIC MEMBRANES FOR MICROPOLLUTANT REMOVAL

Membrane materials

Micropollutant removal efficiency and electrochemical oxidation efficiency largely depend on the properties of electrocatalytic membrane materials. Currently, electrode membrane materials mainly include carbon-based membrane, porous-Ti-based membrane, magnéli-phase-based membrane (mainly Ti_4O_7), electrochemical ceramic membrane, and polymer composite membrane, most of which are microfiltration membranes. In this section, the application and operating conditions of different electrode membrane materials for micropollutant removal are described.

Carbon-based electrocatalytic membranes

Carbon-based electrocatalytic membranes have been widely used for the degradation of micropollutants due to the ease of synthesis, high specific surface area, high conductivity, and excellent mechanical strength (Cui et al., 2021; Patil et al., 2021). The micropollutant removal performance of the carbon-based electrocatalytic membranes is summarized in Table 1. It can be seen that most of the carbon-based

Table 1. Micropollutant removal by carbon-based electrocatalytic membranes under different operation conditions

MPs		Influent concentration (mg/L)	Conductive material	Removal efficiency (%)	Operating voltage or current	Electrolyte	Membrane flux (L/(m^2·h))	References
Carbon								
EDCs	Bisphenol A	1–100	boron-doped MWNTs	>95	3 V	10 mM NaCl	0.202	(Bakr and Rahaman, 2017)
	Bisphenol A	1	MWNTs	100	3 V	10 mM NaCl	12	(Bakr and Rahaman, 2019)
	Bisphenol A	50	coal-based CM	97	2 V	100 mM Na_2SO_4	43.67	(Pan et al., 2019)
	Bisphenol A	30	$BiSnO_2$/CNTM	76.8	3 V	10 mM Na_2SO_4	48	(Zhao et al., 2021)
	Bisphenol A	50	ECM	97.73	2 V	100 mM Na_2SO_4	393.17	(Pan et al., 2020)
	Bisphenol A	0.5	RuO_2/TiO_2 NRs/CNF	98	1.0 mA/cm^2	10 mM Na_2SO_4	360	(Li et al., 2020b)
Pharmaceuticals	Ibuprofen	20	MWNTs-COOH	100	3 V	10 mM NaCl	8.89E-04	(Bakr and Rahaman, 2016)
	Ibuprofen	1	MWNTs	100	3 V	10 mM NaCl	12	(Bakr and Rahaman, 2019)
	Ibuprofen	2	CC	64.87		10 mM NaCl	140	(Yang et al., 2021)
	Paracetamol	15.12	CM	100		50 mM Na_2SO_4	79,575	(Olvera-Vargas et al., 2018)
Antibiotics	Tetracycline	50	ECM	99.98%	2 V	100 mM Na_2SO_4	393.17	(Pan et al., 2020)
	Tetracycline	88.89	CNT	>99	1.5 V	10 mM Na_2SO_4	127.48	(Liu et al., 2015)
	Tetracycline	44.45	CNT	88	0.8 V	10 mM Na_2SO_4	127.48	(Liu et al., 2014)
	Tetracycline	50	Sb-SnO_2/CM	96.5–99.3	3.5 V	100 mM Na_2SO_4	150	(Liu et al., 2017a, 2017b)
	Tetracycline	50	nano-TiO_2/CM	100	1.0 mA/cm^2	105.6 mM Na_2SO_4	/	(Liu et al., 2016b)
	Tetracycline	50	Sb-SnO_2/CA	99	25 mA	70.4 mM Na_2SO_4	150	(Liu et al., 2016a)
	Sulfadiazine	0.5	RuO_2/TiO_2 NRs/CNF	98	1.0 mA/cm^2	10 mM Na_2SO_4	360	(Li et al., 2020c)
	Sulfamethoxazole	5–25	MWCNT	51–90	3 V	20 mM NaCl	127.48	(Tan et al., 2020)
	Ciprofloxacin	5–25	MWCNT	16–99				
	Amoxicillin	5–25	MWCNT	43–75				
	Tetracycline、4-epianhydrochlortetracycline、chlortetracycline	4.45	CNT	98.3	2.5 V	10 mM Na_2SO_4	/	(Yang et al., 2020a)
Others	Pyrrole	300	Graphite-RuO_2-MWCNTs	97.7	3 mA/cm^2	21/35/50/63 mM Na_2SO_4	/	(Zhou et al., 2017)
	Phenol	18.8–94.11	CNT/C-CNT/B-CNT		2 V	100 mM Na_2SO_4	/	(Gao and Vecitis, 2012)

electrocatalytic membranes have decent removal efficiencies for antibiotics, endocrine disrupters, pharmaceuticals, etc. Also, carbon nanotubes (CNTs) are the most commonly used carbon materials in these electrocatalytic membrane processes.

Carbon-based electrocatalytic membranes are capable of removing various antibiotics such as tetracycline (Liu et al., 2014, 2016a, 2016b), sulfadiazine (Li et al., 2020b), sulfamethoxazole, ciprofloxacin, ampicillin (Tan et al., 2020), and 4-epianhydrochlortetracycline (Yang et al., 2020a). For example, more than 99% of tetracycline was removed by a carbon nanotube (CNT) electrochemical filter (Liu et al., 2015) when the cell potential was 2.5 V and the flow rate was 1.5 mL/min (HRT <2 s). In another study, the removal efficiency of tetracycline reached up to 96.5% after a 6-h operation by a novel carbon membrane coated with nano antimony-doped tin dioxide (Sb-SnO_2) (Liu et al., 2017a, 2017b). CNT electrocatalytic filter also showed high removal efficiency (96.8%) for 4-epianhydrochlortetracycline (a hydrolysis product of tetracycline) (Yang et al., 2020b). Tan et al. prepared an electrochemical membrane with multi-walled carbon nanotube (MWCNT) for the removal of various antibiotics, and the removal efficiencies were 90% for sulfamethoxazole, 99% for ciprofloxacin, and 75% for amoxicillin, respectively (Tan et al., 2020).

Bisphenol A (BPA), a typical endocrine disrupting compound, is widely used as an important monomer for synthesizing adhesives and plastics (Bakr and Rahaman, 2017). Even at the level of a few ng/L, BPA can cause severe damage to the endocrine system (Kumari et al., 2021). Recent studies have evidenced that the carbon-based electrocatalytic membranes are very effective to remove BPA (Bakr and Rahaman, 2017, 2019; Li et al., 2020c; Zhao et al., 2021). For example, Pan et al. reported the preparation and application of a new electrochemical microfiltration membrane made of coal-based carbon materials (Pan et al., 2019). When treating the wastewater containing 50 mg/L BPA, the BPA and COD removal efficiencies were up to nearly 97 and 90%, respectively.

In addition, carbon-based electrocatalytic membranes were also used to remove ibuprofen, paracetamol, and other pharmaceuticals (Olvera-Vargas et al., 2018; Yang et al., 2021). The membrane prepared with carboxylated multi-walled carbon nanotubes (MWNTs-COOH) achieved an almost complete removal of ibuprofen at a low applied potential (2 V) (Bakr and Rahaman, 2016). Olvera-Vargas et al. developed a dynamic cross-flow electro-Fenton (DCF-EF) system with a carbonaceous membrane as the cathode, which achieved a complete degradation of paracetamol and 44% of mineralization (Olvera-Vargas et al., 2018).

Porous Ti-based electrocatalytic membranes

Porous Ti is known for its good conductivity, excellent corrosion resistance, high porosity, and outstanding biocompatibility (Zhao et al., 2015). By coating catalysts such as IrO_2, RuO_2, PbO_2, and doped SnO_2 on the surface of porous Ti, the composite membrane could exhibit high electrocatalytic activity (Niu et al., 2012; Xu et al., 2016; Wang et al., 2020; Li et al., 2020a), which has been widely used for micropollutant removal (Table 2), such as endocrine disrupters, pharmaceuticals, antibiotics, and biocides.

Ti-based electrocatalytic membranes achieved a high removal of antibiotics (>90%) such as tetracycline, sulfamethoxazole, and norfloxacin. For example, Yang et al. developed a new type of ultrahigh-throughput tubular filter coated with β-PbO_2, which could remove norfloxacin and sulfamethoxazole of trace concentrations in surface water and wastewater effluent with a residence time of 2.0–5.4 s (Yang et al., 2020a). A novel reactive electrochemical film on a palladium-loaded porous Ti anode was prepared by Ren et al., in which singlet oxygen was directly generated on the anode by Pd-O_2 interaction, resulting in an ultrafast and efficient anodic oxidation of trace antibiotic sulfamethoxazole (Ren et al., 2022).

Ti-based electrocatalytic membranes have also been used for removing biocides. The electrochemical membrane based on Ti and 3D ordered macroporous PbO_2 was fabricated by Liu et al., which achieved an effective removal of flutriafol (75%) (Liu et al., 2017b). The porous-Ti-based electrochemical membrane loaded with RuO_2 was used to remove tricyclazole. Under the optimal conditions (3 mA/cm^2), the removal efficiency of tricyclazole was about 78.4% (Zhang et al., 2016). In another work, Chen et al. developed a novel electrocatalytic membrane for the removal of 2-methyl-4-isothiazolin-3-one by growing TiO_2 nanotube array on the macroporous Ti substrate, followed by a coating of SnO_2-Sb_2O_3 (Chen et al., 2020c). The removal efficiency was approximately 80% (Chen et al., 2020b).

Table 2. Micropollutant removal by Ti-based electrocatalytic membranes under different operation conditions

MPs		Influent concentration (mg/L)	Conductive material	Removal efficiency (%)	Operating voltage or current	Electrolyte	Membrane flux (L/(m^2·h))	References
Ti								
EDCs	Triclosan	10	TiO_2 NTA	99.8	3 mA/cm^2	20 mM Na_2SO_4	3582.8	(Qian et al., 2021)
Pharmaceuticals	Stavudine	0.02	Ti/SnO_2-Sb	100	8 mA/cm^2	20 mM Na_2SO_4	/	(Zhou et al., 2019)
	Naproxen	0.02–0.2	Ti/SnO_2-Sb/Ce-Ti/PbO_2	96.6	10 mA/cm^2	10 mM Na_2SO_4	/	(Xu et al., 2019)
	Paracetamol	15.12	Ti grid		0.5 V/170 mA	50 mM Na_2SO_4	2833	(Huong Le et al., 2019)
Antibiotics	Tetracycline	250	Ti/SnO_2–Sb	99	20 mA/cm^2		40–80	(Xu et al., 2014)
	Tetracycline	17.78	Ti/CNT		0.8 V	10 mM Na_2SO_4	/	(Li et al., 2020a)
	Norfloxacin sulfamethoxazole	0.127–0.152	β-PbO_2-TRF/Ti		250 mA	surface water	/	(Yang et al., 2020b)
	Sulfamethoxazole	0.1	Pd/Ti	>95	0.5 mA/cm^2	50 mM Na_2SO_4	1000	(Ren et al., 2022)
Biocides	Flutriafol	258	3DEM-PbO_2/Ti		5 mA/cm^2	50 mM Na_2SO_4	18339.28	(Liu et al., 2017a, 2017b)
	Flutriafol	100	RuO_2/SnO_2-Sb/Ti	79.6	1.3 mA/cm^2	50 mM Na_2SO_4	82.9	(Xu et al., 2017)
	Tricyclazole	100	Ti-RuO_2	100	3 mA/cm^2	35 mM Na_2SO_4	4172.7	(Zhang et al., 2016)
	2-Methyl-4-isothiazolin-3-one (MIT)	50	MP-Ti-ENTA/SnO_2-Sb_2O_3			100 mM $NaClO_4$		(Chen et al., 2020b)

Likewise, pharmaceuticals can be removed by porous-Ti-based electrocatalytic membrane. The Ti-based membrane loaded with Sb-doped SnO_2 has been used for the removal of typical antiretroviral drug (e.g., stavudine) due to its cost-effectiveness, easy preparation, and high catalytic activity (Zhou et al., 2019). Similarly, Xu et al. prepared a porous Ti/SnO_2-Sb/Ce-PbO_2 membrane for the degradation of naproxen in aqueous solution, with an almost 100% removal at a current density of 10 mA/cm^2 (Xu et al., 2019).

Magnéli-phase-based electrocatalytic membranes

Magnéli phase titanium (Ti_4O_7) has been intensively studied in the field of EM in recent years due to its excellent chemical stability, high conductivity, and low production cost (Gayen et al., 2018b; Liang et al., 2018; Qaseem et al., 2020). Several studies have shown that the Ti_4O_7 electrode can be used as an active electrode for the direct oxidation of pollutants or inactive electrode for the indirect oxidation of pollutants by $^{\cdot}OH$ radicals. The removal performance of micropollutants using magnéli-phase-based electrocatalytic membrane is summarized in Table 3. The magnéli-phase-based electrocatalytic membrane can achieve more than 90% removal of most micropollutants such as pharmaceuticals, antibiotics, biocides, PFAS, and *p*-substituted phenol.

Magnéli-phase-based electrocatalytic membranes have been successfully used for pharmaceutical removal. High carbamazepine degradation (>98%) and mineralization efficiency (70%) can be achieved when treating the secondary effluent of a wastewater treatment plant containing 100 μg/L of carbamazepine (Ganzenko et al., 2021). Trellu et al. used paracetamol as a model contaminant to evaluate the performance of a novel TiO_x membrane synthesized by carbothermal reduction of TiO_2. Under the optimal current density (6 mA/cm^2), the removal efficiency of paracetamol was above 99.9% (Trellu et al., 2018b). Because the intermediate product of paracetamol (i.e., 1,4-benzoquinone) has a strong resistance to direct electron transfer, the mineralization became rather difficult. To enhance the mineralization, Trellu et al. combined TiO_x membrane anode with cathodic electro-Fenton process to synergistically degrade paracetamol into intermediate products (e.g., carboxylic acids), and the mineralization efficiency was increased to 77% (Trellu et al., 2020).

Antibiotics can also be removed by the porous magnéli phase Ti_4O_7 anode. The removal efficiencies were 95% for tetracycline and 95.7% for sulfamethoxazole (Liang et al., 2018; Misal et al., 2020). In these studies, hydroxyl radicals dominated the degradation of antibiotics, mainly by attacking the double bonds as well as phenolic and amine groups of the antibiotics (Wang et al., 2018). Ti_4O_7 electrochemical membrane was also coupled with ozonation, which could completely remove tetracycline within 20 min. Under the optimal conditions (at an ozone dose of 2 mg/min and current density of 2 mA/cm^2), the mineralization efficiency reached ~77% (Zhi et al., 2020).

For the treatment of pesticides, the magnéli-phase-based membrane deposited by bismuth-doped tin oxide (BDTO) catalysts can enhance the mineralization efficiency of pesticides due to the high yield of $^{\cdot}OH$. Atrazine (ATZ) and clothianidin (CDN) have been chosen as the representative pollutants that are commonly found in drinking water sources affected by agriculture activities to evaluate the removal efficiency of magnéli phase electrocatalytic membrane. At 3.5 V versus SHE, the magnéli phase electrocatalytic membrane achieved a complete mineralization of ATZ and CDN within 3.6 s (flux 600 L m^{-2} h^{-1}) (Gayen et al., 2018a).

Magnéli-phase-based electrocatalytic membranes have also been used for the removal of PFAs due to its wide electrochemical window (Le et al., 2019). The porous Ti_4O_7 electrocatalytic membrane successfully removed perfluorooctanoic acid (PFOA) (>99.9% of removal) and perfluorooctane sulfonate (PFOS) (93.1% of removal) at an anode potential of 3.7–3.9 V versus SHE (Lin et al., 2018). It has also been reported that the reaction can be initiated by a direct electron transfer on the anode to generate PFOS free radicals ($PFOS^{\cdot}$) in the degradation of PFOS and then driven by $^{\cdot}OH$ generated via water oxidation (Shi et al., 2019).

p-substituted phenol can also be effectively removed using the porous Ti_4O_7 electrocatalytic membrane. At a current density of 1.0 mA/cm^2, the removal efficiency was 99.9% for *p*-methoxyphenol and 98.2% for *p*-nitrophenol (Zaky and Chaplin, 2013, 2014). It was found that electrochemical adsorption and $^{\cdot}OH$ oxidation were mainly responsible for the removal of *p*-nitrophenol and *p*-methoxyphenol.

Table 3. Micropollutant removal by magnéli-phase-based electrocatalytic membranes under different operation conditions

MPs		Influent concentration (mg/L)	Conductive material	Removal efficiency (%)	Operating voltage or current	Electrolyte	Membrane flux (L/(m^2·h))	References
Ti_4O_7								
Pharmaceuticals	Carbamazepine	0.1	Ti_4O_7 REM	95	7.3/14.5/29.0 mA/cm^2	Secondary effluent from wastewater	3300	(Ganzenko et al., 2021)
	Paracetamol	15.12–347.67	TiOx REM		15 mA/cm^2	50 mM Na_2SO_4	561	(Trellu et al., 2018b)
	Paracetamol	55	TiOx REM		15 mA/cm^2	5 mM Na_2SO_4	95	(Trellu et al., 2020)
Antibiotics	Tetracycline	10–50	Ti/Ti_4O_7	93.9	0.5–3 mA/cm^2	100 mM Na_2SO_4	150	(Liang et al., 2018)
	Tetracycline	5	Ti/Ti_4O_7		15 mA/cm^2	30 mM Na_2SO_4	/	(Wang et al., 2018)
	Tetracycline	5			2 mA/cm^2	30 mM Na_2SO_4	40	(Zhi et al., 2020)
	Florfenicol	5	TiO_2/GF	99.8	0.542 V	50 mM Na_2SO_4	110	(Jiang et al., 2020)
	Sulfamethoxazole	25.32	Ti_4O_7 REM	95.7	2.03 V vs. SHE	100 mM NaClO4	300	(Misal et al., 2020)
Biocides	Atrazine and clothianidin	2.16/2.50	Ti_4O_7		2.1/2.6/3.0/3.5 V	100 mM KH_2PO_4	600	(Gayen et al., 2018a)
PFAS	PFOA、PFOS	10 μM	Ti_4O_7	99.99	3.3/3.6	100 mM KH_2PO_4	240	(Le et al., 2019)
	PFOS	2.0 μM	Ti_4O_7	98.3	0.5–4 mA/cm^2	100 mM Na_2SO_4	272.96	(Shi et al., 2019)
	PFOA PFOS	0.5 mM 0.1 mM		>95	5 mA/cm^2	20 mM NaClO4		(Lin et al., 2018)
Others	Tetrabromobisphenol A	3.50	Fluorinated titanium suboxides (TiSO)	99.7	3.75 V	100 mM $NaClO_4$	651	(Pei et al., 2021)
	1,4-Dioxane	44.06	GO/Fe_3O_4 Ti_4O_7		20 mA/cm^2	20 mM Na_2SO_4		(Li et al., 2022)
	p-Methoxyphenol	127.57	Ti_4O_7			10 mM $NaClO_4$	9–102	(Zaky and Chaplin, 2013)
	p-Nitrophenol *p*-Methoxyphenol *p*-Benzoquinone	139.11 127.57 108.10	Ti_4O_7			10 mM $NaClO_4$	9–102	(Zaky and Chaplin, 2014)

Table 4. Micropollutant removal by electrochemical ceramic microfiltration membranes under different operation conditions

MPs		Influent concentration (mg/L)	Conductive material	Removal efficiency (%)	Operating voltage or current	Electrolyte	Membrane flux (L/(m^2·h))	References
Ceramics								
EDCs	2,4-Dichlorophenoxyacetic acid	1	MI-TiO_2@SnO_2-Sb/ceramic	62.4	3 V	50 mM Na_2SO_4	17–278	(Chen et al., 2020c)
Antibiotics	Sulfamethoxazole	1.27	Pd-CM	82.9	1.6 V	100 mM Na_2SO_4	27,680	(Zhao et al., 2020)
Others	*p*-Chloroaniline	1.27	TiO_2@SnO_2–Sb/ceramic membrane	85.5	3 V	50 mM Na_2SO_4	11.6–138.9	(Zheng et al., 2018)
	p-Chloroaniline	6.38	Ti/RuO_2	91	2 V	50 mM Na_2SO_4	17	(Xu et al., 2018)
	p-Chloroaniline	76.5	ceramic MF	100	800 mA	50 mM Na_2SO_4	80	(Zheng et al., 2019)

Electrochemical ceramic membranes

Ceramic membranes have a high water permeability and robust mechanical strength. However, traditional ceramic membrane materials such as Al_2O_3 and ZrO_2 are nonconductive, and thus electrochemical ceramic membranes are generally prepared by combining electrode materials with ceramic membranes or loading electrocatalytic materials on the surface of ceramic membranes (Collins and Way, 1993; Fu et al., 2019; Zhao et al., 2020). Electrochemical ceramic membranes exhibit very high electrochemical corrosion resistance, which are widely used for the removal of micropollutants (Table 4). In general, the electrochemical ceramic membrane has a good removal efficiency for endocrine disruptors, antibiotics, and aniline compounds, for which the removal efficiencies range from 62% to 100% under the applied voltage of 1.6–3.0 V.

Electrochemical ceramic membrane has been used for removing *p*-chloroaniline (PCA), a chlorinated aromatic amine compound. Zheng et al. prepared a novel electrochemical ceramic microfiltration membrane using TiO_2@SnO_2-Sb anode. Under the optimal conditions (3 V of operating voltage and 17.4 L/(m^2·h) of membrane flux), the removal efficiency of PCA was 85.5%, and the degradation products were mainly nontoxic short chain carboxylic acids (e.g., formic acid, acetic acid, and oxalic acid) (Zheng et al., 2018). The electrocatalytic Ti/RuO_2 membrane prepared by Xu et al. also showed high PCA removal efficiency. The removal efficiency of PCA was 87.1% at 2.0 V of voltage (Xu et al., 2018). Cathodic electrochemical membrane process combined with electro-Fenton process was also used for PCA removal. When pH was 3 and 0.2 mM of Fe^{2+} was added, PCA was completely degraded, and the mineralization efficiency was 75.1% (Zheng et al., 2019). Furthermore, by combining molecularly imprinted TiO_2@SnO_2-Sb anode with ceramic membrane, a selective removal of 2,4-dichlorophenoxyacetic acid (2,4-D) could be achieved with a removal efficiency of 63.6% (Chen et al., 2020c). Zhao et al. developed a Pd-Pt-ceramic membrane using confocal co-sputtering, and the singlet oxygen generated in the electrocatalytic process can remove 82.9% of sulfamethoxazole (Zhao et al., 2020).

Polymer composite membranes

Conductive polymers polyaniline (PANI) and polypyrrole (PPy) are also used for the preparation of electrocatalytic membranes (Ahmed et al., 2016; Duan et al., 2016) due to their conjugated skeleton, hydrophilicity, and relatively high conductivity. For example, Liu et al. prepared conductive polymer composite membrane by blending sulfosalicylic acid dehydrated doped polyaniline (PANI) with polyimide (PI) (Liu et al., 2021). Stainless steel mesh or Ti mesh can be the support layer of conductive polymer composite membranes (Chen et al., 2019; Li et al., 2021). For example, Zheng et al. prepared a cathodic electrocatalytic membrane by embedding stainless steel mesh into the active layer of polyvinylidene fluoride (PVDF) microfiltration membrane and used the composite membrane for the removal of sulphanilic acid (>80% of removal efficiency). The $^{\cdot}OH$ produced at the membrane surface via Fenton reaction contributed to the degradation of sulphanilic acid (Zheng et al., 2017). Similar result was reported for the removal of sulfadiazine in surface water (79% of removal efficiency) (Sun et al., 2018). Jiang et al. prepared graphene-modified electro-Fenton catalytic membrane on the polytetrafluoroethylene (PTFE) membrane for the degradation

of antibiotics. The removal efficiencies of florfenicol and sulfadiazine were about 90% and 82%, respectively, whereas the other three antibiotics (i.e., amoxicillin, ofloxacin, and tetracycline) were completely degraded (Jiang et al., 2018).

According to the existing literature, the relationship between different electrode membrane materials and targeted micropollutants was analyzed (Figure 4). Due to the great electrical conductivity and adsorption capability, carbon-based electrocatalytic membrane is one of the main electrocatalytic membranes for the removal of micropollutants, which are mainly used for the removal of antibiotics, endocrine disruptors, and pharmaceuticals. However, owing to the low standard potential for carbon oxidation reaction (0.207 V versus SHE), carbon-based electrocatalytic membranes are prone to oxidation of carbon materials at high anodic potential. Therefore, carbon-based electrocatalytic membranes generally operate at lower applied voltages (<3 V). Due to the excellent mechanical strength and electrochemical stability, Ti-based electrocatalytic membrane and magnéli-phase-based electrocatalytic membrane are widely used for the removal of antibiotics, endocrine disruptors, pharmaceuticals, pesticides, PFAs, and other micropollutants. Among them, Ti_4O_7 has high oxygen evolution potential and can react with various micropollutants in a wide range of redox potentials. Due to the high hydrophilicity and porous structure, electrochemical ceramic composite membrane and polymer composite membrane are mainly used for the removal of antibiotics and aniline compounds.

Operating conditions

Operating voltage and current density

Operating voltage or current density (i.e., current normalized by per unit area of the electrode) plays an important role in electrochemical oxidation and dominates the generation of strongly oxidizing species on the electrocatalytic membrane surface (Panizza and Cerisola, 2009). In general, there are two major operation modes: constant voltage mode and constant current mode. Based on the available literature (Figures 5A and 5B), the cell voltage is usually about 1–3 V in the constant voltage mode, and the current density is in a range between 0.5 and 20 mA/cm^2 in the constant current mode. Operating voltage and current density are always closely related to micropollutant removal efficiency. Generally, the removal efficiency of micropollutant increases with an increase in the operating voltage or current density. However, a high applied voltage or current density can make the anode potential exceed the oxygen evolution potential, and the occurrence of side reactions will affect the electrocatalytic activity (Zaky and Chaplin, 2014; Hui et al., 2019). In addition, higher operating voltage or current density results in higher energy consumption, thereby increasing the operating cost. Therefore, the determination of the optimal operating voltage or current density needs to be well balanced between the removal efficiency and operation cost. Typically, the operating voltage of 2–3 V or the current density of 3–10 mA/cm^2 is widely used in EM to remove micropollutants (Figure 5C).

Membrane flux

Membrane flux is one of the most important parameters for the operation of EM. Statistical analysis (Figure 5D and Tables 1–5) shows the median fluxes for different electrocatalytic membranes: 138.74 $L/(m^2 \cdot h)$ for carbon-based membranes, 1916.5 $L/(m^2 \cdot h)$ for porous-Ti-based membranes, 195 $L/(m^2 \cdot h)$ for magnéli-phase-based membranes, 138.9 $L/(m^2 \cdot h)$ for electrochemical ceramic membranes, and 64 $L/(m^2 \cdot h)$ for polymer composite membranes. A high membrane flux can facilitate mass transfer and enhance the removal efficiency of micropollutants (Sun et al., 2021). However, if the membrane flux is over high, the contact time between the organic compounds and the strong oxidizing species will be too short to degrade the micropollutants, resulting in a decrease of the removal efficiency (Guo et al., 2016; Trellu et al., 2018b).

Electrolyte concentration/composition

The solution conductivity, electrolyte concentration, and composition strongly influence the performance and stability of the electrocatalytic membrane. In general, the removal efficiencies of micropollutants increase with an increase of the electrolyte concentration. For example, Zhou et al. investigated the effect of electrolyte concentration on the removal efficiency of pyrrole, and the results showed that the removal efficiency of pyrrole increased from 67.2% to 97.7% with an increase of Na_2SO_4 concentration from 3 to 7 g/L (Zhou et al., 2017). Most of the lab-scale studies use synthetic solutions with a 10–100 mM saline as the electrolyte solution to attain a high solution conductivity, although it is much higher than the

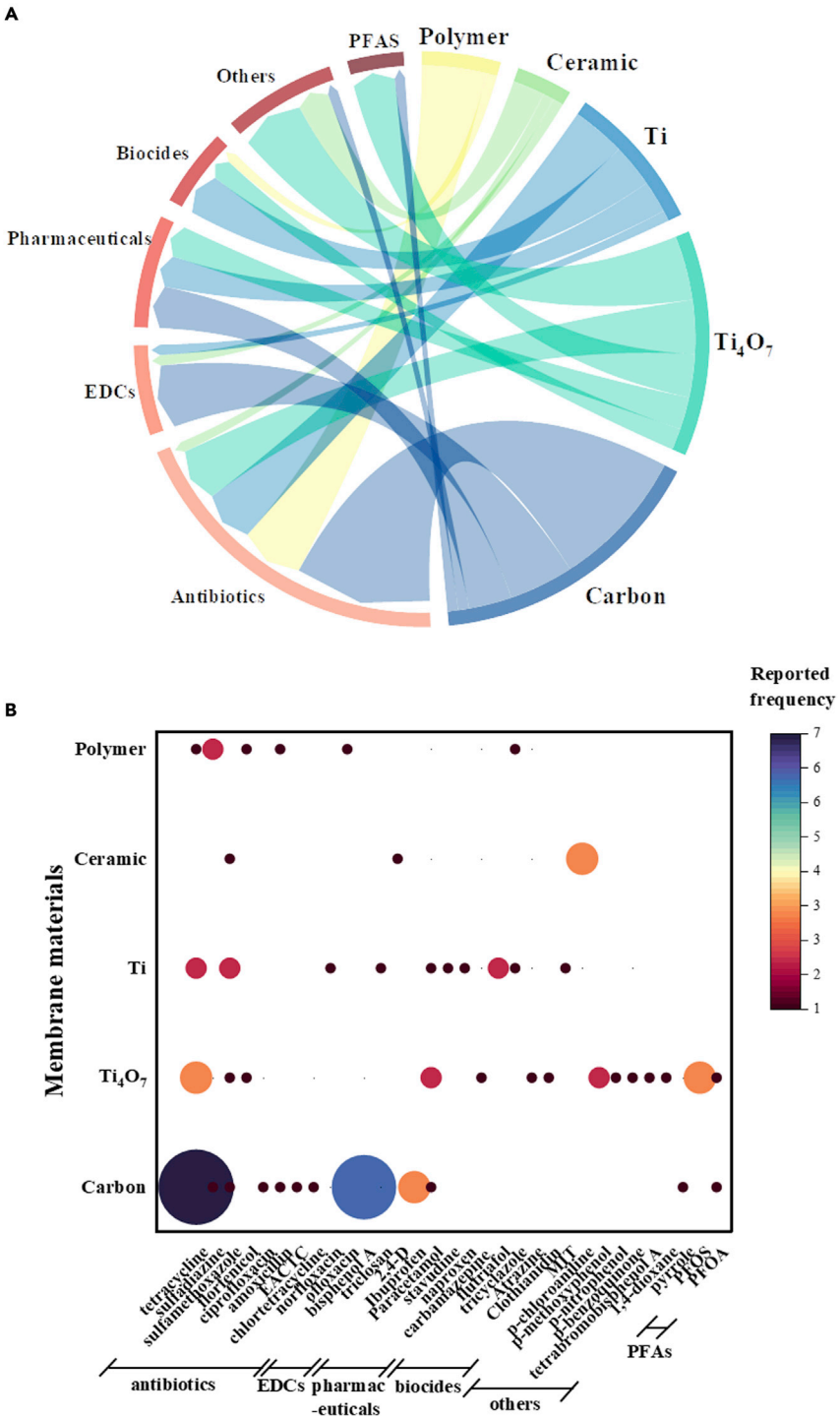

Figure 4. Relationship between electrocatalytic membrane materials and micropollutant removal

(A) Chord diagram and (B) bubble matrix diagram of different electrode membrane materials and targeted micropollutants.

concentration of realistic matrices (such as surface water and municipal wastewater). Nevertheless, it is reported that coal-based carbon membrane can achieve more than 90% of removal of 50 ppm BPA in tap water solution (with a low electrical conductivity of 255 μs/cm) (Pan et al., 2019).

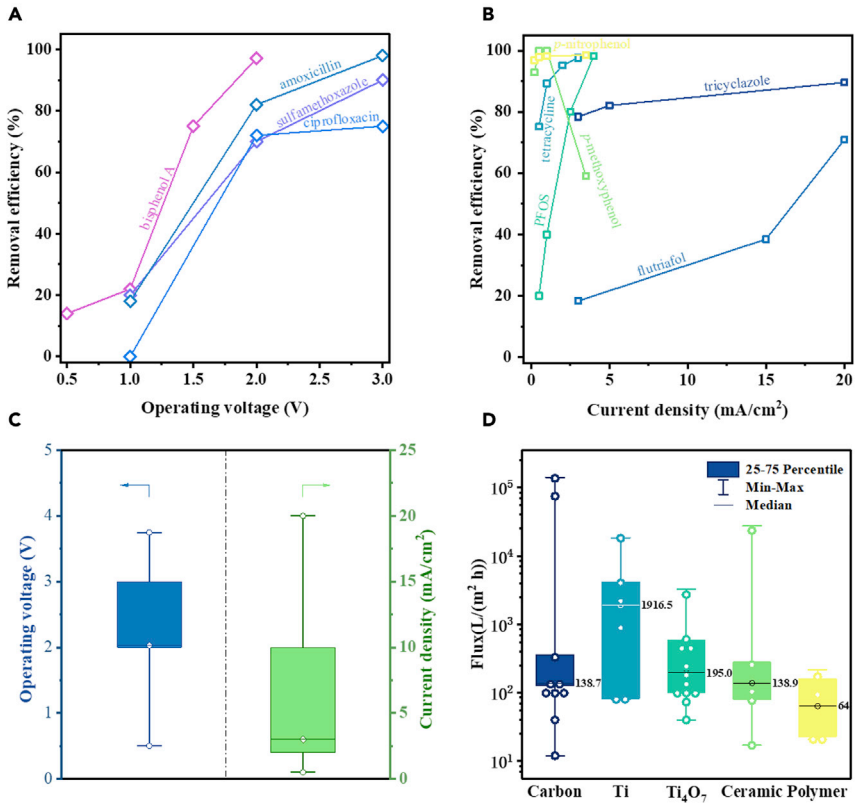

Figure 5. Key operating parameters of EM for micropollutant removal

Effects of (A) operating voltage and (B) current density on the removal efficiency of micropollutants in electrocatalytic membranes. Statistical analysis of (C) operating voltage and current density in electrocatalytic membranes, as well as (D) membrane fluxes of different membranes. Data were collected from the literature summarized in Tables 1, 2, 3, 4, and 5.

In most of laboratory-scale studies, Na_2SO_4 is usually used as the electrolyte (Martinez-Huitle and Brillas, 2008; Liu et al., 2019). When Cl^- is present, reactive chlorine species can be formed, which affects the removal of micropollutants. It should be noted that various anions and natural organic matters (NOM) exist in water and wastewater (Liu et al., 2019). The competitive reaction between these substances (e.g., NOM and CO_3^{2-}) and $^{\cdot}OH$ will affect the electrochemical oxidation reaction (Westerhoff et al., 2007; Khan et al., 2017), thus affecting the removal efficiency of micropollutants.

pH and temperature

The pH and temperature of the solution also affect the removal efficiency of micropollutants in electrocatalytic-membrane-based processes. In most cases, there is an optimal pH value to achieve the highest removal efficiency of micropollutant. One reason is that the pH can affect the ionization of micropollutants in the aqueous solution (Ma et al., 2018). For $^{\cdot}OH$-dominated oxidation, $^{\cdot}OH$ was an electrophile that attacks negatively charged molecules. Therefore, micropollutants in alkaline conditions (i.e., the deprotonated form) are more easily oxidized than those in acidic conditions (i.e., the protonated form) (Martínez-Huitle et al., 2015; Tan et al., 2020). However, the removal efficiency of micropollutants under acidic conditions can be higher than that under alkaline conditions (Zhou et al., 2019). The reason is that the abundant H^+ ions on the surface of the electrocatalytic membrane react with O_2 to form H_2O_2 under acidic conditions, which is conducive to the generation of $^{\cdot}OH$. In addition, for electro-Fenton process, the acidic pH (pH = 3) can promote H_2O_2 to generate $^{\cdot}OH$.

Increasing temperature within a certain range is favorable to micropollutant removal in electrocatalytic-membrane-based processes. For example, the removal efficiencies of sulfamethoxazole (SMZ) at 15°C, 25°C, and 35°C by multi-walled carbon nanotube filter were 77%, 90%, and 96%, respectively (Tan et al., 2020). That is mainly because increasing temperature can reduce the viscosity of the solution, thereby increasing the mass transfer rate of the pollutants to the membrane surface.

Table 5. Micropollutant removal by polymer composite membranes under different operation conditions

MPs		Influent concentration (mg/L)	Conductive material	Removal efficiency (%)	Operating voltage or current	Electrolyte	Membrane flux (L/(m^2·h))	References
Polymer								
Biocides	Tricyclazole	85	CB-PTFE	79	1 mA/cm^2	50 mM Na_2SO_4	103	(Xu et al., 2016)
Antibiotics	Sulfadiazine	0.1–10	PVDF stainless steel mesh microfiltration membrane	98	2.5 V	50 mM Na_2SO_4	25	(Sun et al., 2018)
	Florfenicol Sulfadiazine Amoxicillin Ofloxacin tetracycline	1	e-Fenton catalytic membrane			100 mM Na_2SO_4	20.8	(Jiang et al., 2018)
	Sulfamethoxazole	5–15	G/SnO_2/CFs membrane		1 V	10 mM NaCl	72.9–216.4	(Yu et al., 2020b)

CHALLENGES AND PERSPECTIVES

Due to electrocatalytic oxidation and enhanced mass transfer, EM has shown great potential in the removal of micropollutants in water and wastewater. Although a number of studies have been published in this field in recent years, micropollutant removal by electrocatalytic membranes is still in the early stage for practical application. To promote the application of electrocatalytic membranes in the removal of micropollutants, several key issues need to be addressed.

Mineralization of micropollutants

Although micropollutants are oxidized through direct oxidation or strong oxidants (e.g., $^{\cdot}OH$, $^{1}O_2$) produced by indirect oxidation, some intermediates may be as toxic as or more toxic than their parent compounds (Tan et al., 2020). Besides, toxic by-products may be formed during electrooxidation, such as perchlorate or halogenated organic compounds. Therefore, the efficacy of electrocatalytic membranes should be evaluated from the perspective of the formation of intermediate products and toxicity of by-products. More important, it is of great significance to further improve the mineralization performance of electrocatalytic membranes.

Low-cost, long-lifetime, and environmentally friendly electrocatalytic membrane materials

Membrane materials are of great importance to the efficiency and cost of electrocatalytic-membrane-based processes. Of commonly used membrane materials, CNT and Ti-based membrane materials are still not affordable for practical applications (Patil et al., 2021). Also, the leaching of loaded catalysts such as Pb and Sb into the solution during the filtration process is also a challenging issue (Gao et al., 2014; Liu et al., 2018). Therefore, robust, low-cost, and stable membrane materials suitable for EM need to be further explored.

Simple method for preparation of electrocatalytic membrane

Currently, the preparation methods of electrocatalytic membrane are relatively complex and still in the laboratory stage. For instance, the synthesis conditions of magnéli phase titanium oxides are relatively harsh, requiring a long reduction time under certain pressure (Guo et al., 2016). Therefore, developing a simple preparation method with large-scale production potential is of great significance for the application of electrocatalytic membrane in real wastewater treatment.

Innovative electrocatalytic membrane module and process

As mentioned in Membrane materials, compared with the electrochemical microfiltration/ultrafiltration process, electrochemical nanofiltration and reverse osmosis processes are quite scarce. The coupling of nanofiltration and reverse osmosis membranes with electrochemical advanced oxidation technology deserves further exploration. In addition, for the removal of micropollutants, the integration of biological processes with electrocatalytic membrane filtration processes provides new opportunities for the enhanced removal of micropollutants (Chen et al., 2020a).

Practical application

Although lab-scale studies provide important information about (1) the mechanism of EM for micropollutant removal and (2) the optimization of operating conditions to improve the removal efficiency, there is a gap between the laboratory studies and practical applications. For the real wastewater conditions, the matrix in water is complex. The performance and stability of EM under real wastewater conditions needs further investigation. There is quite limited pilot-scale/full-scale studies on the removal of micropollutants by electrocatalytic membranes. The viability of the technology in a large scale requires further verification.

Limitations of the study

A detailed comparison of strengths and limitations of different membrane materials can be further conducted. In addition, the full-scale application of electrocatalytic membrane technology can be explored.

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AUTHOR CONTRIBUTIONS

L.R.: Investigation, Writing—Original Draft; J.M.: Writing—Original Draft; M.C.: Conceptualization; Y.Q.: Investigation; R.D.: Conceptualization; X.L.: Writing—Review & Editing; Z.W.: Supervision, Writing—Review & Editing, Funding acquisition.

DECLARATION OF INTERESTS

The authors declare no competing interests.

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