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Wei Sun[1], Qibin Xu[1], Shuaishuai Yang[1], Suo Liu[1], Murtaza Sayed[2,3], Emmanuel Mousset[4] and Chun Zhao[1]

1 Key Laboratory of the Three Gorges Reservoir Region's Eco- Environment, Ministry of Education, Chongqing University, Chongqing 400045, PR China

2 Environmental Engineering and Science Program, Department of Chemical and Environmental Engineering, University of Cincinnati, 705 Engineering Research Center, Cincinnati, OH 45221-0012, United States

3 Radiation and Environmental Chemistry Laboratory, National Centre of Excellence in Physical Chemistry, University of Peshawar, Peshawar 25120, Pakistan

4 Nantes Université, ONIRIS, CNRS, GEPEA, UMR 6144, F-85000 La Roche-sur-Yon, France

Corresponding author: Zhao, Chun (pureson@163.com, pureson@cqu.edu.cn)

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Abstract

This paper reviews the latest research advancements in cathodic membrane (CM)–based electrochemical redox processes (CMERs) for water treatment. The water purification mechanisms by CMERs, including CMER reduction, CMER Fenton, and CMER coupling other oxidant processes (CMEOs), are explained. Especially, the pathways of formation of reactive species (e.g. •OH, 1O2, and O2•e) are presented in detail. Besides, the effects of different CMs and operating conditions are considered. The applications extending to refractory pollutants removal, disinfection, membrane fouling alleviation, and resource recovery are well presented and analyzed. CMER reactors are also discussed for their potentials of scale up for water treatment. Finally, the trends in the field encompassing current knowledge gaps are highlighted, and the recommendations for future research are proposed.

Introduction

With the economic growth, a large quantity of emerging organic pollutants are ubiquitously discharged into the aquatic environment [1]. The regulations on drinking water supplies and common monitoring of the environment are on implementing in practice, driven by the re-cognition that emerging contaminants pose high health risks to the human health [2]. However, it has been documented that traditional water treatment processes are ineffective in removing recalcitrant pollutants, thereby posing substantial challenges to the water safety [3,4]. Thus, there is an urgent need to develop efficacious technologies for addressing emerging water quality issues. Electrocatalysis can remove contaminants by *in situ* generating active species [5]. In conventional electrochemical reactions, the stagnant boundary layer on the 2D flat electrodes severely hinders mass transfer and thus reduce current efficiency [6]. Therefore, conductive membranes are employed in electrochemical processes to solve this issue. Numerous studies have shown that the anodic membrane–based electrochemical processes (AMERs) exhibit high mass transfer efficiency, and the produced hydroxyl radicals (•OH) can effectively de-grade pollutants. However, anode corrosion, along with irreversible membrane fouling resulting from electro-sorption of pollutants, hinders its further applications [7]. Compared with anodic membranes, cathodic membrane (CM)–based electrochemical redox processes (CMERs) produce strong oxidizing species for decontaminations through electro-Fenton or electro-activate persulfate (PS)-like reactions [8]. Also, the CM can remove some pollutants, such as heavy metal ions through electro-chemical reduction. Besides, CMs exhibit superior dur-ability because of the absence of electrochemical corrosion and reducing irreversible membrane fouling by electrostatic repulsion [9,10]. In summary, CMERs ef-fectively overcome drawbacks associated with AMERs, enhance efficiency and cut costs in water treatment. To date, many reviews concentrate on AMERs, whereas only a few reviews are dedicated to CMERs. This re-view concisely serves as a supplementary to the ex-tensive literature with a specific emphasis on the production mechanisms of active species in the CMERs (CMER reduction, CMER Fenton and CMER coupling other oxidants process [CMEOs]). Focus is also paid on the CMs used and the influence of various operating parameters. Additionally, the performance of water purification, membrane fouling mitigation and recovery resources by CMERs was presented and analyzed. Moreover, CMER reactors are also discussed for their potential of scale-up water treatment. Finally, the future research needs for the improvement of CMERs are also discussed in detail.

Mechanisms of cathodic membrane–based electrochemical redox process water treatment

CMERs possess the capability to degrade and transform contaminants into nontoxic substances through the generation of reactive species and electron exchange [11]. Based on the decontamination mechanisms, CMERs can be divided into three types: (1) CMER reduction, that is, electrons transferring from the CM to the pollutant; (2) CMER Fenton involving the *in situ* generation of •OH; and (3) CMEOs, which refers to the generation of reactive species by activating externally added oxidants [12]. In this part, the mechanisms of CMER reduction, CMER Fenton and CMEOs are summarized (Table 1) and reviewed systematically. Cathodic membrane–based electrochemical redox process reduction CMER reduction has been utilized for selectively de-grading organics that

readily accept electrons, such as heavy metal ions. As shown in Figure 1, CMER reduc-tion can be divided into direct and indirect reduction. Under a negative potential, a CM with moderate active sites such as functional groups (NO, NH) [13], defective carbon [14], oxygen vacancies [15] and transition metals (Ni, Re) [16] can effectively mediate electron transfer to degrade reducible pollutants (Equation 1; active sites on CM surface are represented by CM_{act}, pollutants are represented by R, and R_T represents transformation products of the parent pollutants). In the other way, the filtration mode accelerates the water reduction to pro-duce atomic hydrogen (H*) on the CM surface, thereby reducing the contaminants indirectly (Equations 2 and 3) [17]. It is noteworthy that the mass transfer is sig-nificantly enhanced due to the hydraulic flow in filtration mode and constraining effect of CM pores. As a result, pollutants are more easily transported to the CM surface, facilitating contact with active sites and thus being re-duced by CMER direct or indirect reduction pro-cesses [54,55].

$$CM_{act} + R \rightarrow CM^{*} + R_T \quad (1)$$

$$H_2O + e^{-} \rightarrow H^{*} + OH^{-} \quad (2)$$

$$H^{*} + R \rightarrow H^{+} + R_T \quad (3)$$

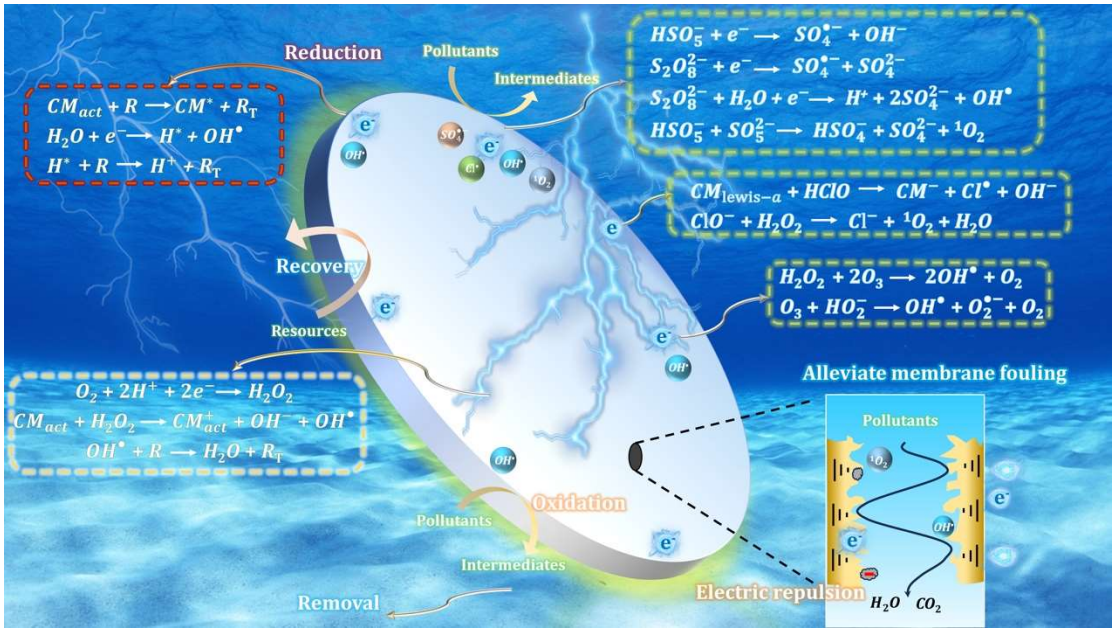

Figure 1. Mechanisms of CMER water treatment and membrane fouling mitigation.

Cathodic membrane–based electrochemical redox process Fenton

CMER Fenton has exhibited great potential in the field of electrochemical wastewater treatment due to its cap-ability to produce hydrogen peroxide (H_2O_2) and •OH. Typically, CMER Fenton operates as a heterogeneous process using CMs with catalytic sites. In a CMER Fenton process, convection enhances the transfer of dissolved oxygen (O_2) towards the CM surface, thereby increasing the yield of H_2O_2 through a two-electron O_2 reduction reactions (Equation 4) [19]. Subsequently, H_2O_2 undergoes Fenton-like reactions on the Fenton catalytic sites of the CM to produce •OH for deconta-mination (Equations 5 and 6), where CM_{act} represents the transition metals (e.g. CuO, Co_3O_4) [20], atomic H* [21], surface N-active species and oxygen-containing functional groups [22]. Meanwhile, the limitations posed by CM enable effective regeneration of the Fenton catalytic sites through electroreduction (Equation 7). In this way, CMER Fenton can persistently oxidize the pollutants that are concentrated within the reactive oxygen species (ROS) diffusion layer of the CMs.

$$O_2 + 2H^{+} + 2e^{-} \rightarrow H_2O_2 \quad (4)$$

$$CM_{act} + H_2O_2 \rightarrow CM^{+}_{act} + OH^{-} + OH^{\bullet} \quad (5)$$

$$OH^{\bullet} + R \rightarrow H_2O + R_T \quad (6)$$

$$CM^{+}_{act} + e^{-} \rightarrow CM_{act} \quad (7)$$

Cathodic membrane–based electrochemical redox process coupling other oxidants

To further improve the performance of CMER, com-bining CMs with other oxidant processes has recently become a vital research topic. The chemical oxidants (e.g. persulfate, O_3 and hypochlorite [ClO_e]) can be active by CM to produce increased active species (e.g. •OH, sulfate radicals [SO_4•] and singlet oxygen [$^1O_2$]) for pollutant removal. *Cathodic membrane–based electrochemical redox process coupling persulfate* Persulfate, including peroxymonosulfate (PMS) and peroxydisulfate (PDS), have been extensively studied for coupling with CM. In the persulfate-assisted CMERs (CM/PS), the CM serves as electron donors, directly activating PMS or PDS to SO_4•e and •OH (Equations 8–11) [23]. Additionally, $^1O_2$, which exhibits high se-lectivity towards electron-rich pollutants, is also involved through subsequent chain reactions (Equations 12–15) [24–26].

Cathodic membrane–based electrochemical redox process coupling hypochlorite

Researchers have also found that the common disin-fectant, ClO_e, can be activated by CMs to generate ROS and reactive chlorine species (RCS) both with and without the involvement of H_2O_2 [27]. Specifically, ClO_e can be directly activated by CM to produce chlorine radicals (Cl•), •OH, or hypochlorite radicals (ClO•; Equations 16–18) [28]. Alternatively, H_2O_2 and •OH generated via Fenton-like reactions can react with ClO_e to form ClO• and $^1O_2$ (Equations 19 and 20) [29].

Cathodic membrane–based electrochemical redox process coupling ozone

Ozone, as an inexpensive oxidant, has also been studied in combination with CMER. Typically, when a negative potential is applied, the CM initially produces H_2O_2 through oxygen reduction reactions. Subsequently, H_2O_2 tends to be selectively converted into HO_{2e} due to the reduction of activation energy barrier by CM (Equation 22). As the result, the yield of •OH is sig-nificantly increased, as O_3 reacts faster with HO_{2e} (= 5.5 × 10_6 M_{e1} s_{e1}; Equation 23) than H_2O_2 (< 0.07 $M_{e1}s_{e1}$; Equation 21) [30,31].

Resulting from the generation of reactive species men-tioned above and the enhanced mass transfer, the pol-lutants in water and those trapped in the membrane pores during CMEOs are degraded immediately (Figure 1).

$$HSO_5^{-} + e^{-} \rightarrow SO_4^{\bullet -} + OH^{-} \quad (8)$$

$$S_2O_8^{2-} + e^{-} \rightarrow SO_4^{\bullet -} + SO_4^{2-} \quad (9)$$

$$HSO_5^{-} + e^{-} \rightarrow SO_4^{2-} + OH^{\bullet} \quad (10)$$

$$S_2O_8^{2-} + H_2O + e^{-} \rightarrow H^{+} + 2SO_4^{2-} + OH^{\bullet} \quad (11)$$

$$OH^{\bullet} + SO_4^{\bullet -} \rightarrow HSO_5^{-} \rightarrow H^{+} + SO_5^{2-} \quad (12)$$

$$HSO_5^{-} + SO_5^{2-} \rightarrow HSO_4^{-} + SO_4^{2-} + {}^1O_2 \quad (13)$$

$$S_2O_8^{2-} + H_2O \rightarrow HSO_5^{-} + HSO_4^{-} \quad (14)$$

$$HSO_5^{-} + 2OH^{\bullet} + 2e^{-} \rightarrow 2SO_4^{2-} + 2H_2O + {}^1O_2 \quad (15)$$

$$CM_{lewis-a} + HClO \rightarrow CM^{-} + Cl^{\bullet} + OH^{-} \quad (16)$$

$$CM_{lewis-a} + HClO \rightarrow CM^{-} + Cl^{-} + OH^{\bullet} \quad (17)$$

$$ClO^{-} + Cl^{\bullet} \rightarrow ClO^{\bullet} + Cl^{-} \quad (18)$$

$$ClO^{-} + H_2O_2 \rightarrow Cl^{-} + {}^1O_2 + H_2O \quad (19)$$

$$ClO^{-} + OH^{\bullet} \rightarrow ClO^{\bullet} + OH^{-} \quad (20)$$

$$H_2O_2 + 2O_3 \rightarrow 2OH^{\bullet} + 2O_2 \quad (21)$$

$$H_2O_2 \rightarrow H^{+} + 2HO_2^{-} \quad (22)$$

$$O_3 + HO_2^{-} \rightarrow OH^{\bullet} + O_2^{\bullet -} + O_2 \quad (23)$$

Table 1. Some recent reports on CMERs including CMER reduction, CMER Fenton and CMEOs.

CMER reduction or oxidation processes	Cathodic membrane	Membrane pore size	Contaminants	Experimental conditions	Removal efficiency	Reference
Reduction	Microchannel charcoal cathode	Average radius of about 10 μm	Nitrate ($NaNO_3$)	0.3 A, pH 8-11, C_0 = 500 mg L^{-1}, 50 mM Na_2SO_4 electrolyte	70.8% in 2 min	[14]
Reduction	Co-CuO_x filter	20-40 μm	Nitrate (KNO_3)	-1.1 V, pH 8.2, C_0 = 20-N-mg L^{-1}, 250 mg L^{-1} $NaHCO_3$ 5 00 mg L^{-1} Na_2SO_4 100 mg L^{-1} NaCl electrolyte, 25 °C	95.95% in 60 min	[15]
Reduction	stainless steel cathode (SS)	mesh number 80	Hexahydro-1,3,5-tri- nitro-1,3,5-triazine (RDX)	100 mA, C_0 = 25 mg L^{-1}, 5 mM Na_2SO_4 electrolyte	100% in 240 min	[17]
Fenton-like reaction	TiO_2@CN membrane cathode	1-10 μm	Propranolol (PRO)	-3 V, C_0 = 20 mg L^{-1}, 10 mM Na_2SO_4 electrolyte	73% in 390 min	[19]
Fenton-like reaction	Fiber felt 316L cathode	25 μm	Benzoic acid (BA)	-3 V, pH 3.0, C_0 = 30 mg L^{-1}, 30 mM H_2O_2, 10,000 μS cm^{-1}, Na_2SO_4 electrolyte	A k_{obs} of 1.93 h^{-1} of mineralization to BA, 100% TOC in 200 min	[21]
Fenton-like reaction	polyaniline-entangled oxidized CNTs cathode (PANI@O-CNTs)	50-100 nm	methyl orange (MO), methylene blue (MB)	-2 V, C_0 = 50 mg L^{-1}, 50 mM Na_2SO_4 electrolyte	89.7% for MO and 93.4% for MB in 50 min	[22]
Combined PDS process	Iron decorated carbon membrane cathode (FeCM)	10-100 μm	Ciprofloxacin (CIP)	15 mA cm^{-2}, pH 6.8, C_0 = 20 mg L^{-1}, 15 mM PDS, 20 mM Na_2SO_4 electrolyte	100% in 60 min	[23]
Combined PMS process	$MnFe_2O_4$-reduced graphene oxide nanocomposite catalytic membrane cathode ($MnFe_2O_4$-rGO)	1-10 μm	Oxytetracycline (OTC)	3 V, pH 7, C_0 = 10 mg L^{-1}, 300 mg L^{-1} PMS, 100 mg L^{-1} K_2SO_4 electrolyte	88.3% at a flow rate of 8 mL min^{-1}	[24]
Combined PDS process	Carbon nanotube cross-linked polypyrrole composite ultrafiltration membrane cathode (CNT-PPy/PVDF)	~100-200μm	Small molecule organic pollutants (i.e., carbamazepine, sulfamethoxazole, phenol, diclofenac)	2.3 mA cm^{-2}, pH 3.2, C_0 = 2 mg L^{-1}, 50 mM PDS, 293 K	Over 95% in 30 min for all small molecule organic pollutants	[25]
Combined HClO process	Electrically conductive carbon nanotube (CNT) membrane cathode	0.03 μm	Benzoic acid (BA)	C_0 = 0.1 mM, 100 mg L^{-1} chlorine as Cl_2, 22 ± 2 °C	50% in 120 min	[29]
Combined O_3 Process	Graphite felt cathode	5 μm	Chloramphenicol (CAP)	5 mA cm^{-2}, pH 3, C_0 = 10 mg L^{-1}, Q_{O_3} = 0.4 L min^{-1}, 50 mM Na_2SO_4 electrolyte, 298 ± 2 K	100% in 40 min	[31]

Factors affecting the cathodic membrane–based electrochemical redox process

Advances on cathodic membranes

The characteristics of CMs are crucial for CMER water treatment. As presented in Table 2, the reported CMs are mainly prepared from polymeric composite and car-bonaceous membranes. Polymeric composite membrane typically possesses excellent separation capability but exhibit low electroconductivity. Therefore, an additional conductive layer is necessary to endow electrocatalytic activity to it. In addition, multiple studies have shown that delocalized π–electron pairs, structural defects and surface functional groups in carbonaceous cathodes serve as electrochemical active sites, enabling the production of strongly oxidative species such as H_2O_2, $SO_4^{\bullet e}$ and •OH [32,33]. However, carbonaceous materials are sus-ceptible to electrochemical oxidation, leading to a de-cline in cathode catalytic activity and integrity. Therefore, many researchers attempted to improve or modify the electroactivity and durability of CM. In this regard, the carbon nanotube (CNT) [30], reduced gra-phene oxide (r-GO) [34] and metallic catalysts [18] are incorporated into the CM to enhance the degradation of pollutants. In addition, Xu et al. suggested that by ap-plying the moderate voltage, electrons can be con-tinuously supplied to the CM to safeguard active sites on it from depletion [35]. Besides, an electroactive single- atom copper anchored MXene nanohybrid CM was uti-lized to boost the removal of sulfamethoxazole (SMX) [36]. Compared with conventional modified methods, single atom–modified CMs exhibit greater performance in pollutant removal because of their superior atomic efficiency and catalytic selectivity.

Influence of operating parameters

In addition to CMs, several operating parameters related to the performance of CMERs are also reviewed (Table 2). The pH plays a crucial role as it can influence the ionization and surface charge of pollutants, making them more easily bonded with the CM and degraded by electrophilic free radicals [29,37]. Furthermore, the pH alters the types and yield of active species. For example, Lee et al. applied active sodium NaClO using a CNT- modified CM to remove benzoic acid (BA) [29]. As the pH increased, the degradation of BA was inhibited

of RCS. In case of cell voltages, the ranges of 1–2 V are suitable for electron exchange between pollutants and CM, while 3–5 V are conducive to generation of active species. Nevertheless, excessive voltages will exacerbate hydrogen evolution side reactions and lead to the gen-eration of disinfection by-products [37]. Moreover, the inorganic ions (e.g. NO_3^{e}, Cl_e, HCO_3^{e}) will generally occupy active sites of CM or quench active species to produce weakly or strongly oxidants. For instance, Zhao et al. observed that Cl_e severely inhibited the degrada-tion of roxarsone in the CM/PMS process due to the quenching of $SO_4^{\bullet e}$ by Cl_e [26], while Yang's study reported that an elevated Cl_e content slightly facilitated the generation of RCS for more rapidly degrading ben-zotriazole [38]. These contradictory trends imply that choosing an appropriate CMER according to water quality and optimizing operating conditions is crucial for improving water purification performance.

Table 2. Some recent reports on the influence of CMs and operating conditions on the performance of CMERs.

CMER reduction or oxidation processes	Cathodic membrane	Membrane pore size	Contaminants	Experimental conditions	Removal efficiency	Reference
Combined PDS process	CNT-PPy/PVDF	5 μm	Carbamazepine, sulfamethoxazole, phenol, diclofenac (DCF)	2.3 mA cm^{-2}, pH 3.2, C_0 = 2 mg L^{-1}, 50 mg L^{-1} PDS	100% in 30 min	[35]
Combined O_3 process	CNT/PTFE membrane cathode	0.09-0.1 μm	Ibuprofen	-2.5 V, pH 7, C_0 = 2 mg L^{-1}, Q_{O_3} = 23.39 mg L^{-1}, 10 mM NaCl, 1mM $NaHCO_3$ electrolyte	100% in 20 min	[30]
Fenton-like reaction	CNM/support/rGO membrane	0.6 nm	Polyethylene glycol (PEG) 1000	0.9 mA cm^{-2}, C_0 = 50 ppm, 1000 ppm Na_2SO_4 electrolyte	100% in 15 min	[34]
Reduction	Ni/AC-PVDF	~ 1-20 μm	Ammonia (NH_3)	17.1 A m^{-2}, pH 7, C_0 = 1 g NH_3-N L^{-1}, 50 mM Na_2HPO_4, NaH_2PO_4, 2.13 g in 1 L of DI water electrolyte	50.3 ± 0.4 g NH_3-N L^{-2} d^{-1}	[18]
Combined PMS process	Cu-SA/Ti_3C_2Tx membrane cathode	0.47 μm	Sulfamethoxazole (SMX)	-1 V, pH 6.2, C_0 = 0.04 mmol L^{-1}, 1.5 mmol L^{-1} PMS	A k_{obs} of 1.93 h^{-1} of mineralization to BA, 100% TOC in 200 min	[36]
Combined NaClO process	CNT membrane cathode	0.03μm	BA	1.9 A m^{-2}, C_0 = 0.1 mM, 10 ppm chlorine as Cl_2, 100 mM phosphate buffer electrolyte	89.7% for MO and 93.4% for MB in 50 min	[29]
Combined PMS process	Fe_3O_4 functionalized CNT (CNT-Fe_3O_4) nanohybrid filter	10-200 nm	Roxarsone (ROX)	-1 V, pH 6.8, C_0 = 1.5 mg L^{-1}, 1.5 mM PMS, 20 mM Na_2SO_4 electrolyte	91.8% in 120 min	[26]
Combined PMS process	PDDA–CNT filter	0.2-1 μm	Congo red (CR)	-1 V, pH 7, C_0 = 10 mg L^{-1},1.5 mM PMS, 100 mg L^{-1} K_2SO_4 electrolyte	100% in 40 min at a flow rate of 5 mL min^{-1}	[37]
Combined NaClO process	CeO_2@CNT membrane	~ 0.5 μm	DCF	0.5 mA cm^{-2}, pH 10.58, C_0= 7.89 mg L^{-1}, 7.5 mg L^{-1} NaClO	92% in 240 min	[27]

Applications of cathodic membrane–based electrochemical redox process in water treatment

Degradation of small organic molecules and disinfection

CMERs have shown exceptional performance that ex-ceed those of traditional electrochemical process in re-moving pollutants. Li et al. presented the first report of a flow-through mode electrochemical test using a GO/Fe_3O_4-NPs@Ti_4O_7 CM, achieving a 1,4-dioxane decay efficiency that is 7.1 times higher than that obtained in the flow-by mode [39]. Similar results were obtained by Ma et al. in degrading tetracycline and bisphenol A [37]. The CMERs have also been reported to be effective in water sterilization [40]. A branched CuO-Co_3O_4 nano-wires coated with carbon on Cu foam CM exhibited a high sterilization efficiency against *Escherichia coli* and *Staphylococcus aureus* [41]. The external electric field al-ters the energy transition between vibrational levels, thereby generating more •OH and superoxide free ra-dicals ($O_{2\bullet e}$) for inactivating bacteria [42]. Moreover, membrane filtration and electrocatalysis, respectively, are generally considered invalid for removing molecular pollutants smaller than the CM pore size, that is, large molecular pollutants, such as polysaccharides as well as small molecules (e.g. SMX, carbamazepine). However, Li et al. constructed a C/PVDF ultrafiltration CM to effectively retain and inactivate antibiotic resistance bacteria (ARB) and antibiotic resistance genes (ARG) while displaying excellent SMX oxidation simulta-neously [42]. This indicates the CMER's promising potential in addressing intricate water containing both small and large molecule pollutants.

Membrane pollution mitigation

Membrane fouling severely limits the performance of traditional filtration processes. As shown in Figure 2, since 1999, researchers have been continuously working on applying a negative voltage to the membrane to mitigate fouling. The electrostatic repulsion plays a pivotal role in controlling membrane fouling. Sun et al. reported that a voltage of e1 V mitigate 19% of the CM fouling by bo-vine serum albumin (BSA) through electrostatic repulsion [43]. Similar findings were also proved by Jiang et al. [22]. They pointed out that the negatively charged pollutants, benefiting from the increased electrostatic repulsion, are less prone to CM fouling. Moreover, the reactive species play a crucial role in alleviating membrane fouling as well. In a study focused on the removal of methyl orange (MO) dye, the electrochemical filtration mode reduced mem-brane flux decline by approximately 15%, whereas the electrochemical filtration mode involving H_2O_2 partici-pation demonstrated a reduction of 38% [22]. The *in situ* generated H_2O_2 oxidizes the foulants occupying the inner pores, resulting in the reduction of membrane fouling. The CM/PMS and CM/PDS processes have also shown remarkable effectiveness in membrane pollution mitiga-tion (Table 3). In summary, the enhanced generation of active species in CMEOs distinguishes it for mitigating membrane fouling.

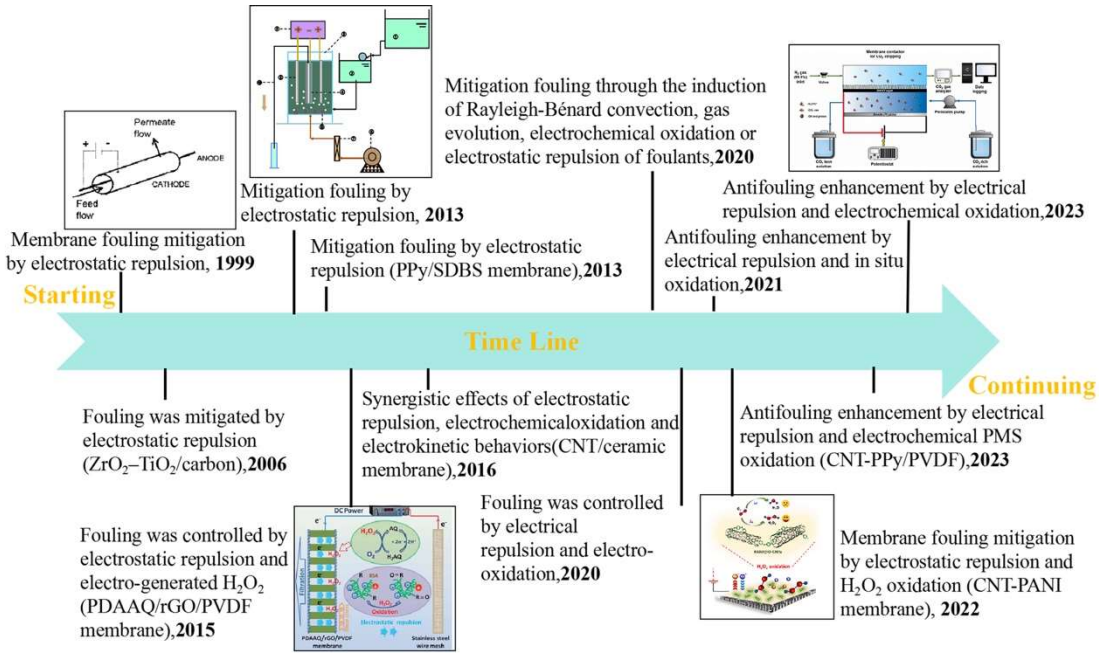

Figure 2. Related studies of CMs used for membrane fouling mitigation.

Recovery of resources

The wastewater contains a significant quantity of nu-trients and metal elements, demonstrating significant potentials for resource recovery [44]. In the past studies on resource recovery using electrochemical filtration, separate cathodes and membranes were predominantly used, inducing elevated cathodic potential losses [45]. Thus, integrated CMs are being explored to solve this issue. As shown in Table 3, a recent report indicated that an activated carbon CM functionalized with nickel achieved a high NH_3-N recovery rate with a low energy consumption owing to the improvement of phase se-paration and faradaic process [18]. Also, the CMERs can improve the selectivity of element conversion for re-source recovery. For example, Kekre et al. constructed a CNT-modified CM to selectively convert 65% of N and P into struvite precipitates for utilization rather than their ionic forms [46]. Therefore, CMER resource re-covery shows promise in adding economic value to wastewater treatment in the future.

Table 3 Some recent reports on the application of CMERs in water treatment

CMER reduction or oxidation processes	Cathodic membrane	Membrane pore size	Contaminants	Experimental conditions	Performance	Reference
Combined PMS process	GO/Fe_3O_4-NPs@Ti_4O_7 REM cathode	0.1-0.3 μm	1,4-dioxane	2.3 mA cm^{-2}, C_0 = 0.5 mM, 45 Mm PMS, 20 mM Na_2SO_4 electrolyte	85.4% in 180 min	[39]
Combined PMS process	Co-r-TNTs membrane cathode	10-100 μm	N,N-dimethyl-p-nitrosoaniline (RNO)	-3 V, pH 8.2, C_0 = 0.1 mM, 0.1 mM Na_2SO_4	76% in 250 min	[47]
Fenton-like reaction	MnOx-in-CNT nanohybrid filter	0.1-0.5 μm	bisphenol A (BPA)	100 mA, pH 6.5, C_0 = 0.022 mM, 10 mM Na_2SO_4 electrolyte	93.3% in 240 min at a flow rate of 1.5 mL min^{-1} ,	[48]
Fenton-like reaction	Co-r-TNTs	30-60 nm	*E. coli*	-3 V, C_0 = 10^6 CFU mL^{-1}, 0.1 M Na_2SO_4 electrolyte	99.9% in 390 min in 4 h at a flow rate of 1.5 mL min^{-1}	[41]
Fenton-like reaction	CuO-Co_3O_4@C NWs filter	10-100 μm	*E. coli* and *S. aureus*	10 V, pH 3.0, C_0 = 10^4 CFU mL^{-1}	99.5% for *E. coli* and *S. aureus* at a flow rate of 100 mL min^{-1}	[29]
Fenton-like reaction	C/PVDF ultrafiltration membrane	30-80 nm	SMX, ARB, ARG	-2 V, C_0 = 2 mg L^{-1}, 10^6 CFU mL^{-1}, 10^9 copies mL^{-1}, 50 mM Na_2SO_4 electrolyte, 1.0 bar pressure, 30.0 ± 0.5 °C	100% for SMX, 4-5log removal for ARB and 95% - 100% for ARG in 12 h	[42]
Combined PMS process	Conductive polypyrrole (PPy)-modified membrane (PMM)	0.1-0.5 μm	Bovine serum albumin (BSA)	-1 V, pH 6.8, C_0 = 100 mg L^{-1}, 100 mg L^{-1} PMS, 0.03 MPa	18.82% in membrane fouling mitigation compared to filtration	[43]
Fenton-like reaction	PANI@O-CNTs membranes	50-100 nm	Methylene blue (MB)	-2 V, pH 6.8, C_0 = 50 mg L^{-1}, 50 mM Na_2SO_4 electrolyte, D_o = 19.21 mg L^{-1}	32.6 % in membrane fouling mitigation improvement at 50 min compared to filtration	[22]
Fenton-like reaction	PPY-SSM	8 μm	sodium alginate (SA), bovine serum albumin (BSA), humic acid (HA)	-2 V, pH 6.8, DOC_0 = 6.5 mg L^{-1}, 0.05 mol L^{-1} NaCl electrolyte	15%, 18%, 27% and 5% permeability recovery for SA, BSA, HA removal in fist cycle compared to filtration	[49]
Reduction	CNT/PES membrane	10-100 nm	N, P	-3.5 V, pH 6, C_0 = 1000 mg L^{-1}, 0.9 bar	65% for N, P removal in 30 min	[45]
Reduction	Composite conductive membrane cathode	0.1-1 μm	Cu-EDTA	-3V, C_0 = 0.5 mM, 50 mM Na_2SO_4 electrolyte	81.5% decomplexation of the Cu-EDTA and 72.4% recovery of Cu in 2.5 h	[44]

CMER reactors and scale-up application

In the previous study, the CM generally serves as the working electrode, while a steady-state anode is placed on the feed side. The mostly used dead-end filtration mode in CMERs, where feed flows across the CM surface with the electric field perpendicular to the flow, is susceptible to the formation of membrane a filter cake [50]. Compared with the cross-flow devices, the flow-through ap-paratus reduces the thickness of the diffusion boundary layer to a scale comparable to the CM pore radius, thereby enhancing mass transfer and controlling the membrane fouling [51]. Generally, the reported flat CMs employed in the cross-flow and flow-through reactors are of laboratory scale (surface areas less than 90 cm_2). How-ever, when handling municipal waters on the order of several hundreds of millions of litres per day, large CM units become necessary. When the CM area exceeds that of the anode, it leads to an uneven distribution of current density and a sharply decrease in current density as the CM extends longitudinally, thereby reducing the elec-trocatalytic efficiency [14]. However, the high cost of large steady-state anodes makes widespread application impractical. The tubular reactor equipped with coiled electrodes may be a viable option to address these issues. First, industrial tubular modules typically occupy 30–200 m_2 m_{e3} of plant space, which is 17–45 times less than that of flow-by reactors [52]. Meanwhile, the tubular reactor has been shown to preserve the benefits of filtra-tion-enhanced mass transfer and effective exposure of active sites [51,53]. More importantly, tubular devices can accommodate large cathode membranes arranged con-centrically with an anode, effectively preventing the loss of current efficiency caused by CM longitudinal exten-sion. This implies that tubular membrane reactors can maintain high current efficiency and daily water treat-ment capacity even with a small steady-state anode. Therefore, the tubular reactor seems to be a feasible option for implementing the CMERs in scale-up waste-water treatment in practical.

Conclusion and future perspectives

The CMERs have obtained increasing interest in the field of water treatment. Compared with the traditional electrochemical and AMERs, CMERs offer the ad-vantages of the high yield of free radicals, electrostatic repulsion to mitigate membrane fouling and absence of electrochemical corrosion. Researchers have developed CMs that simultaneously possess excellent conductivity and electrocatalytic capability. The single atom-mod-ified method seems to be more closely aligned with the pursuit. In addition, several operating parameters should be optimized according to the specific water quality in practice. Moreover, it is worth noting that both CMER Fenton and CMEOs show exceptional performance in pollutant degradation. Especially, the latter appears outstanding in alleviating membrane fouling, thanks to the production of large numbers of active species. In terms of resource recovery, the CMER Fenton process seems to emerge as the sole choice. Furthermore, the tubular reactors accommodating large conductive mem-branes are the most probable choice for the large-scale water treatment. Nevertheless, the CMERs still require testing and engineering to come closer to relevant ap-plications in real water treatment. First, various free radicals have been confirmed as the predominant contributors to pollutant degradation in CMERs. However, their generation and reaction me-chanisms have not been fully elucidated, causing con-fusion when applying CMERs in real water treatment. For example, whether the CM pore size affects the types of generated free radicals, whether they exist only in the electroactive layer or tend to accumulate inside the CM pores and whether the degradation mechanism of pol-lutants changes with the longitudinal depth of the CM. Besides, real water matrices are more complex, and pollutant concentrations are much lower than in labora-tory settings. Thus, the viability of CMERs in actual water treatment requires further verification. In addition, it is noteworthy that the CMEOs possess excellent performance in degrading pollutants and alleviating membrane fouling. However, the current mainstream CM materials are sensitive to oxidants (e.g. PS, H_2O_2 and NaClO), which leads to membrane failure easily. Therefore, controlling the dosage of oxidants to obtain the trade-off between dose of oxidants and CM stability is necessary for the application of CMEOs. Moreover, tubular electrochemical reactors provide the potential for large-scale water treatment using CMERs. Nevertheless, a mature method for preparing CMs with large surface areas, great flexibility and electrocatalytic activity has not been established. Overall, the CMERs deserve more effort to be dedicated to their practical application in regard to widespread water treatment.

Data Availability

Data will be made available on request.

Declaration of Competing Interest

The authors have no competing interests to declare.

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