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Next-generation graphene oxide additives composite membranes for emerging organic micropollutants removal: Separation, adsorption and degradation

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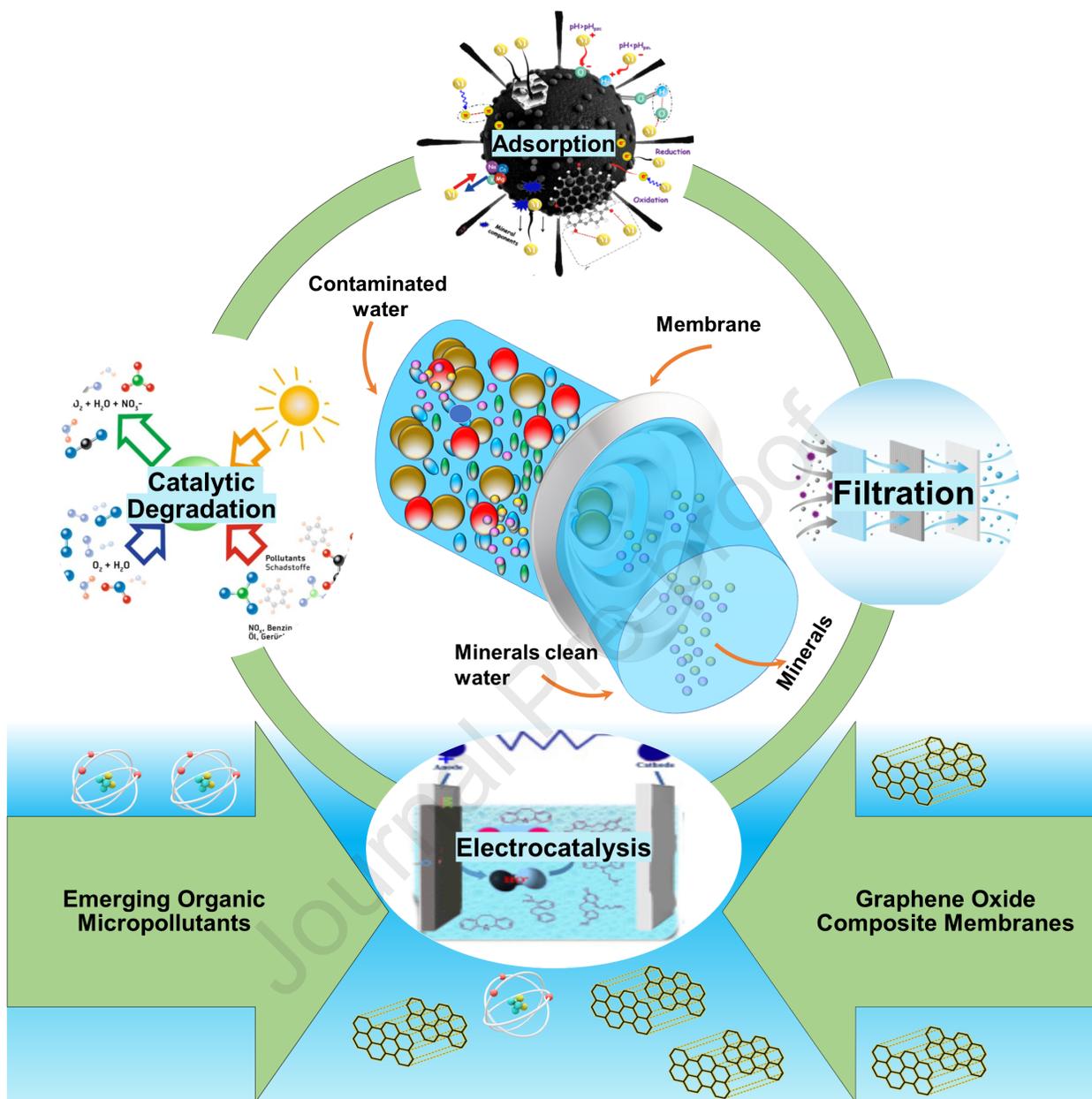
Next-Generation graphene oxide additives composite membranes for emerging organic micropollutants removal: Separation, adsorption and advanced oxidation process.

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1 **Next-Generation graphene oxide additives composite membranes for**  
2 **emerging organic micropollutants removal: Separation, adsorption**  
3 **and degradation**

4

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23

24

25 **Abstract**

26 In the past two decades, membrane technology has attracted considerable interest as a viable and  
27 promising method for water purification. Emerging organic micropollutants (EOMPs) in wastewater

28 have trace, persistent, highly variable quantities and types, develop hazardous intermediates and are  
29 diffusible. These primary issues affect EOMPs polluted wastewater on an industrial scale differently than  
30 in a lab, challenging membranes-based EOMP removal. Graphene oxide (GO) promises state-of-the-art  
31 membrane synthesis technologies and use in EOMPs removal systems due to its superior  
32 physicochemical, mechanical, and electrical qualities and high oxygen content. This critical review  
33 highlights the recent advancements in the synthesis of next-generation GO membranes with diverse  
34 membrane substrates such as ceramic, polyethersulfone (PES), and polyvinylidene fluoride (PVDF). The  
35 EOMPs removal efficiencies of GO membranes in filtration, adsorption (incorporated with metal,  
36 nanomaterial in biodegradable polymer and biomimetic membranes), and degradation (in catalytic,  
37 photo-Fenton, photocatalytic and electrocatalytic membranes) and corresponding removal mechanisms  
38 of different EOMPs are also depicted. GO-assisted water treatment strategies were further assessed by  
39 various influencing factors, including applied water flow mode and membrane properties (e.g.,  
40 permeability, hydrophily, mechanical stability, and fouling). GO additive membranes showed better  
41 permeability, hydrophilicity, high water flux, and fouling resistance than pristine membranes. Likewise,  
42 degradation combined with filtration is two times more effective than alone, while crossflow mode  
43 improves the photocatalytic degradation performance of the system. GO integration in polymer  
44 membranes enhances their stability, facilitates photocatalytic processes, and gravity-driven GO  
45 membranes enable filtration of pollutants at low pressure, making membrane filtration more inexpensive.  
46 However, simultaneous removal of multiple contaminants with contrasting characteristics and variable  
47 efficiencies in different systems demands further optimization in GO-mediated membranes. This review  
48 concludes with identifying future critical research directions to promote research for determining the  
49 GO-assisted OMPs removal membrane technology nexus and maximizing this technique for industrial  
50 application.

51 **Keywords:**

52 **Composite membranes, Graphene oxide, Membrane technology, Micropollutants, Wastewater**

53

## 54 **1. Introduction**

55 Organic micropollutants (OMPs) in water have gained significant attention due to their implication for  
56 water security, human health, and environmental safety (Silvestro et al., 2021). These pollutants include

57 a wide range of chemicals: Pharmaceuticals and personal care products (PPCPs), Pharmaceutically active  
58 compounds (PhACs), hormones/ endocrine-disrupting chemicals (EDCs), pesticides and other industrial  
59 chemicals. The small molecular weights but long persistence and high diffusivity trace quantities of  
60 OMPs from industrial effluents and daily wastewater remain untreated due to low concentrations. The  
61 concentration of OMPs may be varied from  $\text{ng L}^{-1}$  to  $\text{mg L}^{-1}$  (Zheng et al., 2022), and if present below  
62 parts per million (ppm), they are known as emerging organic pollutants (EOMPs) (Lapworth et al., 2012).

63 Although EOMPs are always present in trace concentrations, they significantly impact human health  
64 (Han et al., 2018). Only endocrine-disrupting chemicals (EDCs) cause considerable financial costs, such  
65 as 217 billion (1.28% of gross domestic product, GDP) in Europe (Trasande et al., 2015) and 340 billion  
66 (2.33% of GDP) in the United States (Attina et al., 2016). On the other hand, human-usable water  
67 resources are becoming increasingly scarce over time, necessitating the effective treatment of OMPs  
68 contaminated water. Since wastewater treatment plants (WWTPs) units are not logged to treat OMPs and  
69 treatment standards and guidelines for OMPs are yet to be defined, these contaminants remain unchecked  
70 in wastewater treatment processes and are frequently discharged into the environment (Zheng et al.,  
71 2022).

72 Incessantly, the scientific community seeks satisfactory enhancements to conventional cleanup  
73 procedures or novel alternatives. However, understanding OMPs, including EOMPs and related efficient  
74 treatment technologies, is still in infancy (Kanaujiya et al., 2019). Integration of membrane filtration  
75 technology with conventional WWTPs has proven to improve EOMPs removal performance in the  
76 system. To date, considerable research has been done on membrane separation performance for gas  
77 (Mubashir et al., 2015), dyes (Dadvar et al., 2017), and other environmental remediations (Ali et al.,  
78 2022).

79 Membrane systems, including reverse osmosis (RO), nanofiltration (NF), ultrafiltration (UF), and  
80 microfiltration (MF) are pollutants specific and have specific operating requirements, pollutants removal  
81 efficiencies, advantages, and limitations. For instance, due to high transmembrane pressures requirement,  
82 fouling, and membrane concentrate management, NF and RO spectacularly increase operational  
83 expenses (Luo et al., 2014b). Therefore, these membranes require additional assistance from different  
84 supporting technologies. On the other hand, advanced membrane bioreactors are well established in  
85 treating refractory organic pollutants, nutrients, and organic matter (e.g., humic acid (HA)) but are  
86 ineffective for nonbiodegradable EOMPs and often face severe membrane biofouling, need modern

87 water treatment strategies (Ding et al., 2021). Advanced oxidation processes (AOPs) disintegrate  
88 persistent MOPs into simpler minerals. In paired cleaning procedures, these AOPs reckon on various  
89 oxidants that can totally mineralize EOMPs or increase their biodegradability for later bioremediation  
90 (Li et al., 2022a). Fenton oxidation is a common and eminent AOPs method. Fenton oxidation and  
91 ozone/UV oxidation process has excellent removal efficiency, rapid functions, and broad use range (Yu  
92 et al., 2020).

93 Nonetheless, some organic substances are resistant to oxidation by ozone or UV-based processes,  
94 and partial mineralization of molecules can produce even more harmful byproducts. Besides, activated  
95 carbon is merely a separation procedure that requires additional post-filtration (Ma et al., 2022).  
96 Electrochemical advanced oxidation process (EAOP), a simple automated in situ executable system,  
97 compact and modular reactor architecture, and absence of chemical storage or handling, is efficiently  
98 used to treat aqueous OMP waste streams but is ineffective when used EOMPs contents are low.

99 To meet the challenges of these membrane coherent advance treatment processes, the development  
100 of robust, multi-functional, resilient, and economical composite membranes for treating domestic and  
101 industrial effluents containing EOMPs is progressing. However, some parameters that should be  
102 considered while considering advanced composite membrane are physical strength and stability against  
103 chemicals, selectivity, high flux, permeability, high rejection rate, type of pollutants, and removal  
104 mechanisms (Yuan et al., 2020; Wang et al., 2021). These features are directly related to the material  
105 characteristics of the composite membrane (Shen et al., 2017). Popular two-dimensional (2D) materials  
106 with outstanding selectivity and permeability, such as MXenes, graphene, 2D covalent-organic and  
107 metal-organic frameworks, and Molybdenum disulfide ( $\text{MoS}_2$ ), are increasingly used in membrane  
108 production for water purification. As evidenced by the vast amount of research conducted in this field,  
109 graphene oxides have been widely utilized in numerous membrane systems to remove various OMPs  
110 from contaminated water. GO membranes are emerging as additive materials that improve antifouling  
111 behavior, selectivity, and flux performance of membranes (Liu et al., 2020c; Wu et al., 2020) due to high  
112 mechanical strength, high thermal stability, huge surface area, hydrophilicity, and efficient electron  
113 transformation.

114 It has been proven various membrane substrates showed contrasting characteristics while in  
115 conjugation with GO. Once GO is incorporated, it alters the membrane physiochemical characteristics,  
116 impacting EOMPs removal pathways and corresponding efficiencies (Zhu et al., 2017b; Ye et al., 2021b).

117 Importantly membrane filtration studies denoting water purification have been applied in various systems  
118 such as UF, NF, MF, and RO (Table S1). These membrane systems are operated under various flux  
119 dynamics, e.g., cross-flow (CF), surface-flow (SF), and flow-through (FT) , as well as in different  
120 treatment systems (MBR, hybrid DMBR). Consequently, efficiencies and EOMPs removal mechanisms  
121 substantially varied with operating system changes. Likewise, treatment cost and environmental  
122 implications regarding membrane reusability, stability, and the desorption and leaching of supporting  
123 material are also changed (Nguyen et al., 2021; Ma et al., 2022). Only biofouling can increase 50% the  
124 cost of filtration operation (Fortunato, 2017). Such disparities limit their applications in multi-  
125 contaminated wastewater treatment. Although some lab-scale studies showed appreciable performance  
126 in this context, a field scale is still challenging (Li et al., 2021a; Wei et al., 2021; Ye et al., 2021b).

127 Based on the current literature, the following limitations can be concluded: (i) reviews discussed the  
128 single type of membranes based on removal mechanisms (Thakre et al., 2021; İlyasoglu et al., 2022), (ii)  
129 GO membranes removal performance partially reviewed or discussed along other 2D materials (Bodzek  
130 et al., 2020; İlyasoglu et al., 2022) (iii) Membrane performance for organic contaminated water treatment  
131 has been reviewed, but a detailed review on EOMPs removal by different GO membranes is still elusive  
132 (Khanzada et al., 2020; Jia et al., 2022), (iv) a detailed insight assessment of purely GO-based membranes  
133 and a critical comparison of GO functions in different removal mechanisms are not available, (v) and  
134 due to complex nature of EOMPs, limitations in conventional membrane filtration systems and increasing  
135 interest of researchers for GO membranes, a review on adsorption, degradation, and filtration of EOMPs  
136 by GO modified membranes is warranted.

137 To curb these gaps, the main objectives of this review are to (1) evaluate GO additive membranes  
138 in various types of filtration systems (e.g., microfiltration, ultrafiltration, nanofiltration, reverse osmosis)  
139 and delineate the synergism of adsorption, degradation, and electro-photocatalytic strategies in the  
140 removal of EOMPs, (2) probe the integration of GO with diverse membrane substrates (e.g., ceramic,  
141 polyethersulfone (PES), polyvinylidene fluoride (PVDF)) to remediate OMPs, (3) to compare the  
142 removal mechanisms of various EOMPs by GO-assisted membranes, and (4) elucidate the main factor  
143 affecting GO integration with different substrates and corresponding physicochemical changes in GO-  
144 modified membranes compared to the pristine membrane.

145 We are convinced that reviewing simple to most advanced GO additive membranes will advance  
146 our understanding of pollutants-specific, economic, and more stable membrane synthesis, which is  
147 essential for next-generation membranes development and advanced industrial applications.

## 148 **2. Literature search methodology**

149 For this study, the Scopus database (updated on July 1, 2022) was searched by entering the following  
150 keywords into the titles of publications: (("Graphene Oxide" or "GO"), ("membranes"), ("water  
151 treatment"), (emerging pollutants), and (organic micropollutants) yielded a total of 74 results. In a  
152 separate search, the keyword in the title: (("Graphene Oxide" or "GO"), ("membranes"), ("water  
153 treatment"), and (emerging pollutants) produced 333 articles. We selected all SCOPUS database indexes  
154 articles published between 2004 and 2022, with an emphasis on the last five years of study. This search  
155 approach is commonly employed in Scopus bibliometric studies (Palansooriya et al., 2020; Mehmood et  
156 al., 2021). Because this search resulted in many unrelated publications, the articles were initially filtered  
157 by reading the article titles based on GO's usage in removing EOMPs from water. The shortlisted articles  
158 were then read to decide which ones were relevant. All these investigations are summarized in Tables 1  
159 through 3.

## 160 **3. Emerging Organic Micropollutants in Environment**

161 Organic micropollutants are a broad chemical category that includes PAHs, endocrine disruptors,  
162 antibiotics (Silvestro et al., 2021), plasticizers, and flame-retardants. These pollutants are a global issue,  
163 and their trace amount ( $\text{ng L}^{-1}$  to  $\text{g L}^{-1}$ ) carries substantial adverse impacts on the environment and public  
164 health (Zheng et al., 2022). Other OMPs like personal care items, medications, insecticides, additives,  
165 and hormones in water and wastewater are found in fewer than parts per million (ppm), collectively  
166 called emerging contaminants (ECs), and if they are organic pollutants ascribed as EOMPs. These  
167 chemicals have adverse environmental implications but have untracked sources and remained formerly  
168 unregulated or unchecked in environmental samples but have suddenly become a public health issue  
169 (Yang et al., 2016).

170 EOMPs are classified as (i) PPCPs, (ii) EDCs, (iii) Per- and polyfluoroalkyl substances  
171 (PFASs)/industrial chemicals, and (iv) organic wastewater-related compounds based on their sources and  
172 properties (Rahman et al., 2009). EOMPs can be located in various settings, including industry,  
173 agriculture, healthcare, and everyday life. Wastewater treatment facilities (WWTPs) effluent, landfill

174 debris/leachate, septic/drainage tank leakage, and agricultural runoff comprise a major fraction of  
175 EOMPs in surface and groundwater. Likewise, household garbage, hospitals, and other industrial  
176 applications are the most common sources of EOMPs pollutants (Rahman et al., 2009; Zheng et al.,  
177 2022). A detailed description of EOMPs and an overview regarding the performance of membrane  
178 separation technology in the treatment of EOMPS contaminated water has been provided in the  
179 supplementary information.

#### 180 **4. Scope of GO for membrane separation**

181 Graphene is a potential adsorbent material for membrane separation due to its unusual p-electronic  
182 configuration and greater specific surface area. This material has recently been outlined for its perks in  
183 a spectrum of uses, particularly in photocatalysis (Chen et al., 2017; Zhu et al., 2017a; Heu et al., 2020),  
184 superlattices formation (Yu et al., 2014) and MST (Shen et al., 2017; Wu et al., 2021a). Of the best  
185 graphene derivatives is its oxidized form, known as "graphene oxide (GO; formerly known as graphitic  
186 oxide)," which has caught the interest of researchers worldwide. Brodie was the first to synthesize GO  
187 by oxidizing graphite with a potassium chlorate solution and fuming nitric acid in 1859. Previously, in  
188 1958, Hummers and Offeman described "The Hummers' Method," which included  $\text{KMnO}_4$ ,  $\text{NaNO}_3$ , and  
189  $\text{H}_2\text{SO}_4$  (Fig. 1 a-c). Various modified Hummer methods have been described in the literature  
190 (Khorrasmshokouh et al., 2022; Yin et al., 2022). GO is also synthesized by pyrolysis with the complete  
191 carbonization of carbon-enriched waste materials (e.g., sugarcane Bagasse) (Somanathan et al., 2015)  
192 and carboxyl-based compounds (e.g., citric acid) (Dong et al., 2012), as presented in **Fig. 1**.

#### 193 **Fig. 1 about here**

194 As shown in Fig. 1d, the oxidation of graphite and the exfoliation of graphite oxide are two main  
195 steps in preparing GO from graphene. 2D honeycomb-like crystalline lattice structure of GO contains  
196 numerous water channels. Analogous to graphene, sheets of GO are asymmetrically attributed to specific  
197 basal and edges holding oxygen-containing functional groups. On the other side, GO is a single atomic-  
198 layered thick nanosheet with a hexagonal carbon composition that, unlike graphene, has a number of  
199 unique properties, including being the stronger but lightest sheet, having exceptional thermal  
200 conductivity (approximately  $5000 \text{ W m}^{-1} \text{ K}^{-1}$ ), contains the meager electrical resistivity ( $10^6 \text{ U cm}$ ), and  
201 a massive theoretical specific surface area (SSA) ( $2630 \text{ m}^2 \text{ g}^{-1}$ ) when referred to similar materials. It also  
202 features a hexagonal structure with more active functional groups on the surface, such as hydroxyl ( $-\text{OH}$ ),  
203 alkoxy ( $\text{C}-\text{O}-\text{C}$ ), carboxyl ( $-\text{COOH}$ ), and other oxygen enriched functional groups (Wang et al.,

204 2021). Oxygen-rich functional groups enable the production of nanocomposite materials with prominent  
205 chemical permanency, excellent hydrophilicity, and remarkable antifouling properties by providing a  
206 varied variety of reaction sites for chemical reactions (Ye et al., 2021b; Ye et al., 2021c; Xu et al., 2022).

207 Due to its unique structural features, it has been increasingly applied to various MSTs as a novel  
208 type of additive in recent years. However, the GO bases membrane has many problems, which are  
209 exacerbated by several techniques. For example, such oxygen-bearing groups reduced the electrostatic  
210 stability of nanosheets in water, which is another limitation of GO (Nebol'sin et al., 2020). However,  
211 hydroxyl, epoxide, carbonyl, and carboxyl groups can be introduced to the basal planes of GO to facilitate  
212 dispersion in aqueous conditions in the absence of any stabilizer (Wang et al., 2021). The small layer  
213 spacing also impacts the water flux of GO nanosheets (Wu et al., 2014). Hence widening the layer  
214 spacing is a promising area of study. Water can travel through GO sheets if provided nanopores but other  
215 ionic substances cannot (Foller et al., 2022).

216 Membrane surface reconfiguration is a technique for adjusting membrane hydrophilicity, surface  
217 charge, porous structure, and roughness. Due to its particular qualities for separation efficiency, the GO  
218 has been commonly applied for surface modification. Several methods for assembling GO nanosheets  
219 into laminar membranes have been described in the literature (Fig. 2), like filtration (pressure-, vacuum-  
220 , and evaporation-assisted) (Baskoro et al., 2018), layer-by-layer (Nair et al., 2012b), dip-coating (Li et  
221 al., 2022b) and immersive method (Rashidian et al., 2021), spin coating (Nair et al., 2012b) and drop-  
222 casting (Rollings et al., 2016), and spray-coating (Guan et al., 2017). Hu et al. (Ayyaru and Ahn, 2017)  
223 used a layer-by-layer technique to segregate water from the dye mixture to build a coupled GO  
224 membrane. As an additive material in membrane production, GO offers many applications.

225 The most often used approaches include the incorporation of GO in polymer matrix by conventional  
226 blending (Ayyaru and Ahn, 2017) and/or coating via dip coating (Surana et al., 2015) and sol-gel  
227 methods (Luo et al., 2014a). However, due to the nanomaterials' restricted homogenous scattering in the  
228 polymer, these techniques lacked uniformity and repeatability for permeation and selective removal  
229 purposes. Essential characteristics for forthcoming surface alteration techniques are the uniform  
230 distribution and adjustable GO thickness on a membrane's active layer (Kong et al., 2021). Atomic layer  
231 deposition (ALD) and electrospinning are two novel methods for immobilizing functional GO and its  
232 uniform dispersion on membrane surfaces with a customizable thickness (Lee et al., 2020).

233

**Fig. 2 about here**

234 GO's smooth surface and hydrophilicity make it promising as a membrane additive as it  
235 demonstrates great changes, including anti-fouling/biofouling and selectivity behavior in membranes by  
236 altering pore size and surface charges while also amplifying the permeate and water fluxes of the  
237 membrane (Liu et al., 2020c; Wu et al., 2021a). These GO-based membranes can be classified into three  
238 categories: (i) porous GO layers, (ii) GO laminates, and (iii) GO composites (Shen et al., 2017; Wu et  
239 al., 2020). Nair et al. (2012a) showed that water could pass freely via helium-tight, submicrometer-thick  
240 membranes. Wei et al. (2021) developed a polyamide GO nanofiltration membrane (GO/PA) for  
241 purifying water with a high flux ( $44.2 \text{ L m}^{-2} \text{ h}^{-1}$ ). Organic dyes preserved a high percentage (>99  
242 percent), whereas ion salts retained a low rate (5%). About 20%–60% of total removal was attributed to  
243 GO due to its adsorption capacity for pollutants in water solution. For instance, Li et al. (2021a) used  
244 GO with a Europium-metal organic framework; the membrane showed 100% removal of organic  
245 phosphine and increased the flux from 0.76 to  $5.01 \text{ L m}^{-2} \text{ h}^{-1}$ .

246 Jin et al. (2015) demonstrated that magnetically reduced GO has an analogous mechanism for the  
247 adsorption of BPA and 4-n-nonylphenol. Aside from the fact that GO has been used in diverse ways to  
248 aid membrane pattern and surface changes, the following section relies on GO-based membranes and  
249 their suitability for micropollutant contaminated water treatment systems, owing to their widespread use  
250 and promising applications for improving membrane processes (Chu et al., 2017; Shen et al., 2017). In  
251 both pressure- and temperature-driven membrane processes, the introduction of GO materials has been  
252 widely exploited to increase permeability, selectivity, and fouling control (Ma et al., 2020; Li et al.,  
253 2021a; Zhang et al., 2022).

## 254 **5. GO-based separation/filtration membranes**

255 Numerous studies have demonstrated that incorporating GO into membranes either as a bulk component  
256 or as a surface graft improves the antifouling function of the membrane by improving hydrophilicity and  
257 lowering the surface roughness (Mahlangu et al., 2017; Zhang et al., 2018). Many GO additives  
258 membranes are eminent by underlying removal mechanism, proficiency, stability, and environmental  
259 implications. The next section provides a detailed overview of several GO-mediated next-generation  
260 approaches for EOMPs cleaning, emphasizing their efficiency, perks, and shortcomings (Section 4.1-4.4,  
261 Table 1).

262 **Table 1 about here**

### 263 **5.1. Mixing of GO with other 2D materials**

264 The oxygen-bearing groups on the basal boundary of GO nanosheets improve GO dispersion in water,  
265 which is beneficial for boosting interactions with transport species (water molecules and ions) during  
266 water purification (Li et al., 2019b; Wang et al., 2021). The repulsive forces between GO-linked  
267 functional groups and water molecules as well as within GO nanosheets are the major factor that causes  
268 GO dispersion in water, separation of EOMPs, and also cause instability and destroying its permeability  
269 (Nair et al., 2012a; Wang et al., 2021). Therefore, managing the balance between selectivity and  
270 permeability is challenging when simply employing GO membranes. However, utilization of ionic cross-  
271 linking ligands and functional materials and the decrease of oxygen-containing groups in GO are  
272 common approaches adopted to cope with this situation (Kang et al., 2017). Likewise, combining GO  
273 with other two-dimensional materials in membrane production showed encouraging elimination of  
274 EOMPs from water (Kang et al., 2017; Liu et al., 2020b).

275 MXene( $Ti_3C_2Tx$ ) is getting prominence due to its outstanding characteristics, which include high  
276 electrical conductivity, mechanical strength, extensibility, and wettability (Lu et al., 2022). MXene  
277 membranes with a lamellar configuration and high hydrophilicity, for instance, promoted quick and  
278 targeted water molecule transport with a desirable permeability flux in water filtration (Liu et al., 2020b).  
279 Assuming that the 2D lamellar heterogeneous form of the GO/MXene composite membrane, compared  
280 to individual GO or MXene membranes, may have offered synergistic benefits, leading to an optimal  
281 trade-off between selectivity and permeability.

282 Ding et al. (2017) demonstrated a considerably higher interlayer gap and flow than a GO membrane  
283 was by two-dimensional lamellar MXene membrane. They addressed the absence of oxygen-containing  
284 functional groups, and larger interlayer spacing are the main reasons for the GO/MXene composite  
285 membrane's improved flow, as discussed later. The lamellar construction with an uneven thickness of  
286 0.7 nm (GO) and 1.5 nm (MXene) promotes water flow transit by preventing the creation of lamellar  
287 nanosheets, allowing multistage micropores to be produced, and maximizing the production of multistage  
288 micropores the number of microchannels. Likewise, by carefully adjusting the surface functional group  
289 makeup of GO/MXene composite membranes, a high rejection frequency for EOMPs, such as common  
290 pigments and representative forms of natural organic matter found in raw waters, may be preserved (Liu  
291 et al., 2020b).

292 GO and MXene membranes laminated onto porous support successfully removed methyl red (68%),  
293 rose Bengal (93.5%), methylene blue (99.5%), and brilliant blue (100%) from water (Kang et al., 2017).  
294 At the same time, using GO/MXene intercalants membrane, Liu et al. (2020b) demonstrated 95%  
295 removal of organic dyes from raw water while rejecting 100% natural organic components (sodium  
296 alginate (SA), bovine serum albumin (BSA) and HA). Notably, the introduction of 2D material resulted  
297 in a potential improvement in the membrane separation process attributable to GO membranes. MXene  
298 comprises a vast family of 2D materials; however, only MXene has been coupled with GO and  
299 investigated in the EOMPs membrane removal method to date. This should be explored further in the  
300 context of forthcoming research.

### 301 **5.2. GO integrated ceramic membranes**

302 Ceramic membranes outperform polymeric membranes in harsh environments owing to corresponding  
303 chemical, mechanical, and thermal resistance. They are also easier to clean and have higher flux stability  
304 (Zielińska and Galik, 2017). However, the performance of ceramic membranes to remove low molecular  
305 ionic chemical retentions in nanofiltration is not much higher, which can be substantially improved by  
306 incorporating GO (Hu et al., 2016). Although the deformity of the GO layer in an aqueous environment  
307 widens interlayer gaps and resuspends the GO, which declines the membrane removal performance (Lou  
308 et al., 2020), but the interlayer space of GO can be modulated using layer-by-layer assembly (LbL)  
309 method, which produces more stable GO layer spaces (Nair et al., 2012b).

310 Shen et al. (2017) developed SG@GO composite membrane using pyrene derivative solvent green  
311 (SG) molecules and GO owing to  $\pi$ - $\pi$ -stacking interactions. As a result, they achieved a membrane with  
312 a desirable spacing arrangement (Fig. 3h). Previously, Liu et al. (2012) found the wide flat aromatic  
313 structure of SG molecules can bind with GO sheets' hydrophobic surface by  $\pi$ - $\pi$  – interactions. The  $\pi$ - $\pi$   
314 -stacking interactions is analogous to that of covalent bonding, which improves the stability of the GO  
315 composites in aqueous media. Additionally, the  $\pi$ - $\pi$  stacking alteration does not affect the GO sheet  
316 pairing, which preserves graphene's characteristics (Liu et al., 2012).

317 **Fig. 3 about here**

318 Shen et al. (2017) demonstrated that the incorporation of SG enhanced electronegativity, reduced  
319 contact angle, and improved hydrophilicity. A direct relationship between the interlayer spacing of GO  
320 laminate and SG concentration was attributable to a rise in sulfonate ( $-\text{SO}_3$ ) concentration, which  
321 enhances the electrostatic repulsive forces between the SG@GO sheets by increasing the negative surface

322 zeta potential (SZP) (Fig. 3g). Consequently, when the electrostatic repulsive forces between the GO  
323 layers increased, the nanochannels for water molecules grew larger. GO membrane thickness rose from  
324 2.2 to 2.7 nm in SG@GO sheets by increased static repulsion force, whilst the inter-layer gaps of GO  
325 sheets in pristine GO laminates (7.70Å) were increased to 8.29Å and 8.41Å in SG@GO laminates at SG  
326 concentrations of 1.2 and 1.6 g/L, respectively.

327 GO laminates by immersion method of depositing SG@GO nanocomposites onto PDA-amended  
328 hollow ceramic substrates demonstrated a significant increase in pores numbers and enhanced surface  
329 roughness (Fig. 3a-f) (Shen et al., 2017). Although GO improved removal but it hinders water flux too.  
330 In comparison, SG contains huge planar aromatic compounds that engage via  $\pi$ - $\pi$  stacking interactions  
331 with GO sheets, hence enhancing the mass transfer routes and electrostatic repulsive force, resulting in  
332 increased flows but decreased retention (Fig. 3i). However, a thicker and denser separation layer of  
333 SG@GO enhanced the rejection efficiency from 94.2 to 98.0% at moderately higher flux rate. The  
334 SG@GO hybrid membrane performed excellent electronegativity EBT molecules removal from water  
335 due to the Donnan effect and molecular filtering. However, advanced materials and fabrication  
336 procedures are necessary to improve ceramic membrane nanofiltration performance and more  
337 straightforward preparation.

338 Lee et al. (2015) addressed that an excellent cationic polyelectrolyte with a high positive charge  
339 density Polyethylenimine (PEI), can link GO to form electrostatic LbL arrangements. However, Yang et  
340 al. (2018) identified owing to wider pores of the substrate, only addition of PEI addition is void while  
341 co-deposition with GO could cover all pores, and they recommended a combination of metal-organic  
342 frameworks (MOFs) (Fig. 4a-e). The authors demonstrated 99% removal of organic solvent by co-  
343 depositing PEI and Zeolitic Imidazolate Framework (ZIF-8) nanoparticles additive GO on a tubular  
344 ceramic substrate via a vacuum-assisted process approach. Using GO sheets or ZIF-8 nanoparticles, they  
345 prevented PEI from entering the substrate's pores, resulting in a thin film composite membrane (Fig. 4g).  
346 Agglomeration occurred only when ZIF-8 particles were mixed with PEI, resulting in an extremely rough  
347 surface. ZIF-8@GO laminates aid in the construction of a regular separation layer structure while  
348 preventing the aggregation of ZIF-8 particles and the wrinkle formation of GO nanosheets in ZIF-8@GO  
349 composite nanosheets.

350 The improvement of ceramic membrane nanofiltration needs further assessment for other EOMPs  
351 (Yang et al., 2018). Cha et al. (2022) recently used LbL methods and incorporated PEI-GO assembly in

352 the ceramic membrane for semiconductor wastewater purification in NF. Three GO-PEI bilayers GO-  
353 ceramic eliminated 8.4 and 3.2 times as much ammonium as the pure ceramic NF substrate. However,  
354 the use of these strategies is still limited in literature, particularly for EMOPs removal studies.

355 **Fig. 4 about here**

### 356 **5.3. GO in PES membrane**

357 Although PES membranes are one of the most extensively used polymeric UF membranes, efficiency  
358 needs to be enhanced (Zhang et al., 2021). Metal oxides were found to be useful in increasing PES  
359 membranes' performance and stability when incorporated into GO. For example, during concurrent  
360 filtration and disinfection of a UF system, Jiang et al. (2016b) discovered that a GO-TiO<sub>2</sub> modified PES  
361 membrane under UV irradiation displayed excellent self-maintenance and increased antibacterial activity  
362 during extended operation intervals. Likewise, in a 72-hour long-term trace organics rejection  
363 experiment, Mahlangu et al. (2017) treat trace organic compounds (TOCs) and examine the influence  
364 of temporal solute adsorption on rejection using 5 g/L carbamazepine as the model solute. They  
365 discovered that although PES membranes without GO-ZnO had a significant drop in carbamazepine  
366 rejection (30%), those treated with nanohybrid GO-ZnO showed a less severe drop (less than 20%).

367 PES membranes impregnated with nanohybrid graphene oxide-zinc oxide (GO-ZnO) showed  
368 considerable improvements in hydrophilicity, pharmaceutical rejection (by lowering membrane-solute  
369 hydrophobic interactions), organic fouling resistance, and photodegradation. The specific alteration in  
370 GO could also improve its permeability, for instance, the use of HGO at 0.08 g/m<sup>2</sup> doubled the  
371 permeability of the PES membrane. A 0.04 g/m<sup>2</sup> level of HGO coated PES membranes showed improved  
372 (55% to 92%) rejection for NOM (e.g., BSA, SA, and HA). Increasing HGO loading also slowed the  
373 transition from pore blockage to the cake layer HA-based fouling and irreversible fouling of PES  
374 resistance lessened from 3.45 to 1.73 and from 62.86%–95.83% with HGO coating (Ding et al., 2021).  
375 HGO outperformed GO in terms of performance and NOM rejection, suggesting that membrane fouling  
376 in water supply, wastewater treatment, and reuse can be adjusted using modifying GO synthesis methods.  
377 It also notices that managing the HGO pore size, setting appropriate working conditions, and assuring  
378 the stability of the HGO composite membrane are all important considerations.

379 Lemos et al. (2021) demonstrated that GO-integrated membrane showed a 13% lower decline in flux  
380 than PES membrane. For both membranes, the permeate flux was less affected during the filtration of  
381 HA solution than the BSA one. The PES-GO membrane performed remarkably well regarding HA

382 solution, showing a minimal flux decay of only 2% after a permeate recovery rate of 80%. Irreversible  
383 fouling decreases significantly with GO due to the more negative surface charge flux recovery ratio  
384 (recoverable + irrecoverable) increased from 72% for the control PES membrane to 85% for the PES-  
385 GO high rejection and antifouling properties for HA and BSA in model solutions. After prolonged  
386 exposure to sodium hypochlorite solution, PES-GO exhibited loss of hydrophilicity and presented a  
387 significant indication of pitting-like damage on the active chlorine layer. However, GO strengthening  
388 with co-depositing PEI and Zeolitic Imidazolate Framework (ZIF-8) nanoparticles successfully removed  
389 up to 99% of organic pollutants.

### 390 5.3.1. GO-PES membranes antifouling functions in membrane MBRs

391 Ultrafiltration (UF) is a modern, cost-effective, and environmentally friendly technique used in  
392 membrane bioreactors (MBRs) (Zhang et al., 2018), and it is commonly supported by PES membrane,  
393 which is hydrophobic by nature (Zhang et al., 2021). However, over time the membrane could be  
394 ineffective for organic and microbial biomass deposition (Zhang et al., 2018).

395 Since GO is poisonous to bacteria, the GO composites membrane could prevent microorganism  
396 growth in MBRs. Unlike many other antimicrobial nanoparticles, GO nanosheets' antibacterial activity  
397 is triggered by direct contact with bacterial cells (Thambirajoo et al., 2021). Two major mechanisms by  
398 which GO disrupted the integrity of bacteria cells were (i) the penetration of GO's atomically sharp edges  
399 into the bacterial cell (Lu et al., 2017) and (ii) the oxidation reactivity with the GO nanosheets linked to  
400 cell membrane (Nanda et al., 2016; Zhang et al., 2021). Other organic fouling and early bacterial  
401 deposition inhibition methods include (i) antibacterial material coating, zwitterion polymers alteration  
402 (Zhang et al., 2018), and catalytic oxidation attributed elimination are all viable methods for decreasing  
403 (Zhang et al., 2021).

404 Zhang et al. (2018) developed a zwitterion polyampholyte hydrogel-imbedded PES membrane  
405 surface (p-PES membrane) for loading onto GO nanosheets (GO-p-PES composite membrane) (Fig. 5).  
406 BSA's adsorption potential was reduced by 70% due to the synergistic impacts of both. The composite  
407 membrane killed over 80% of *E. coli* colonies after 12 hours and showed a 25% reduction in flux  
408 compared to a 65% reduction in the pristine PES membrane. Furthermore, the biofilm layers on the GO-  
409 p-PES (9 m) and p-PES (8 m) membranes surfaces were lower than 19 m on the unmodified PES  
410 membrane. GO-p-PES and p-PES exhibited a similar organic fouling tendency in static biodegradable  
411 CA ultrafiltration membrane separation and dynamic filtration. Over 98% of the GO was still present on

412 the membrane surface after seven days, which shows very low leaching. Zwitterion polymers effectively  
413 lessen early bacterial adhesion and organic fouling, which in turn slows the development of biofilm.  
414 These results indicate that the GO-functionalized polyampholyte hydrogel membrane has effective anti-  
415 biofouling characteristics.

416 **Fig. 5 about here**

#### 417 **5.4. GO added PVDF membranes**

418 PVDF membranes are the most broadly used membranes in MST because of their inexpensive flexibility,  
419 mechanical strength, and chemical and thermal resistance. The improved membrane was predicted to  
420 show potential in practical applications based on its exceptional performance in water science. On the  
421 other hand, the high hydrophobicity of PVDF prevents their application in water purification. According  
422 to Wu et al. (2020), grafting polyvinylpyrrolidone additive GO (PVP-GO) onto PVDF membranes  
423 resulted in a negative charge and improved hydrophilicity, resulting in solid repulsive forces between  
424 biopolymer and membrane, boosting antifouling performance (Fig. 6c). Advanced treatment (tertiary  
425 treatment) may be required for organic residues resistant to ordinary treatment (Oller et al., 2011).

426 Since soluble microbial products and self-polymerized organic molecules have a high molecular  
427 weight, they can be separated from salts and water (Yang and Yen, 2013). Earlier studies indicated that  
428 specific preparation procedures could enable GO membranes to fix the challenges of concentrated waste  
429 containing organics, inorganic ions, and chemical or energy consumption (Han et al., 2018; Yang et al.,  
430 2019). However, just a few research looked into the production of GO membranes for enhanced treatment  
431 of refractory toxics from wastewater (Yu et al., 2020). Han et al. (2018) created a GO membrane that can  
432 separate organic and inorganic salts, reducing the problem of mixed concentrate waste and boosting  
433 resource recovery from inorganic concentrate waste generated during membrane filtration (Fig. 6a). The  
434 modified GO membrane rejects COD at 76% and salt at 14% in a typical secondary effluent of  
435 recalcitrant effluents. Additionally, a greater assembly pressure (~10 bar) helped develop a membrane  
436 with a better exclusion rate and lesser roughness. The results of this study established an innovative  
437 application for GO membranes in wastewater treatment enhancement (Fig. 6b). Removal of inorganic  
438 and organic salts and the fabrication and application of GO membranes are currently understudied.  
439 Likewise, the impact of the supporting membrane surface characteristics (pore size, roughness), synthesis  
440 pressure (Tsou et al., 2015), and GO layers levels (Lou et al., 2020) on different salts, dyes, and OMPs  
441 separation performance and other GO membrane parameters are still unknown.

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**Fig. 6 about here**

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## 5.4.1. GO-PVDF membranes antifouling performance in bioreactor (MBR)

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Recently, Wu et al.(2021a) demonstrated that grafting PVP-GO composites on the PVDF hollow fiber membrane surface and coating  $\text{Ag}_2\text{CO}_3$  significantly reduced the formation of bio-cake layers by (i) limiting microalgae growth on membranes, (ii) increasing hydrophilicity, (iii) decreasing protein binding, (iv) preventing the formation of bacterial aggregate and biopolymers, and (v) expelling large clusters from the membrane. The PVDF membrane has a maximum FDR of 83.1%. Following membrane-provided hydrophilic alteration, the A 83% FDR of PVDF (83%) was reduced and reached 67% (PVDF/GO) and 37.7% (PVDF/GO/ $\text{Ag}_2\text{CO}_3$ ), showing remarked anti-pollution properties of the modified membrane. This is because the high hydrophilicity of the membrane prevents the binding of hydrophobic BSA (Mansourpanah et al., 2015). In practical applications, the modified membrane is remarkably successful at enhancing the antifouling characteristics and permeability of PVDF UF membranes.

GO additives are commonly used in membrane bioreactors (MBR) for treating industrial and domestic wastewater due to their minor trail, good water quality, and ease of usage (Mukherjee et al., 2018; Qin et al., 2018). Nevertheless, significant membrane fouling is always seen as the main impediment to MBRs being used in industry (Wu et al., 2020; Zhang et al., 2021). The main foulants in MBRs, extracellular polymeric substances (EPS), cause membrane biofouling (Zhang et al., 2018; Zhang et al., 2021). Protein and polysaccharides are the two most important components, accounting for 70–80% of total EPS (Wu et al., 2020).

In PVP-GO modified membrane, the distinct hydrophilic ligands on PVP chains act as planar combs, inhibiting *Chlorella* bindings with the membrane surface. For example, Wu et al. (2020) developed PVP-GO/PVDF membranes in algae MPBR (solids retention times: 30 d; hydraulic retention times:24 h) that successfully removed 97.8% and 76.9% of influent COD and  $\text{NO}_3\text{-N}$ , respectively, which was higher than the pristine membranes' removal of 93.0% and 70.6%. Moreover, the concentration of  $\text{NH}_4\text{-N}^+$  in the influent declined below  $7 \text{ mg L}^{-1}$ , ranging from  $68.4$  to  $71.9 \text{ mg L}^{-1}$ . The results showed that farmed *Chlorella pyrenoidosa* biomass was more effective at removing ammonia nitrogen. To create microalgae *pyrenoidosa*, they obtained 96.8% removal in PVP-GO/PVDF composite membrane. Likewise, the protein content of loosely tied LB-EPS, EPS on the surface of PVP-GO/PVDF, and pristine PVDF composite membrane was ( $7.07 \text{ mg g}^{-1}$  versus  $55.24 \text{ mg g}^{-1}$ ), i.e., 1/8 of the latter. The protein percent of

472 tightly bound EPS and TB-EPS was ( $27.11 \text{ mg g}^{-1}$  versus  $72.55 \text{ mg g}^{-1}$ ), i.e., below half of the unchanged  
473 membrane surface (Fig. 6e). Overall, after 16 days, the biomass content was sustained at approximately  
474  $780 \text{ mg L}^{-1}$  and the mean biomass generation rate approached  $45 \text{ mg L}^{-1} \text{ d}$  (Wu et al., 2020).

#### 475 5.4.2. GO-PVDF additive molecularly imprinted polymers membranes

476 Tetracycline (TC) is a macrolide antibiotic with a wide range of applications in agriculture and human  
477 health (Liu et al., 2014; Wu et al., 2021b). Significant separation fluxes and sensitive molecular  
478 selectivity coexist in aquatic habitats, making TC removal problematic. Graphene oxide is a low-cost  
479 adsorbent for removing PhACs from wastewater, but its limited selectivity prevents it from being used  
480 for a wide range of pollutants (Yang et al., 2016; Zhang et al., 2018). The Molecular Imprinting  
481 Technology (MIT) based membrane showed excellent pollutants selectively water purification system  
482 (Ghasemi and Nematollahzadeh, 2018; Ma et al., 2022).

483 Ma et al. (2022) demonstrated  $\kappa$ -carrageenan-altered GO nanosheets-double-layer-imprinted mixed  
484 matrix membranes, TC-imprinted activated carbon (AC) and  $\text{SiO}_2$  GO@ $\text{SiO}_2$ /AC-based filter layers  
485 could be used with other PhACs or environmental contaminants than other MIMs. First and second-TC-  
486 imprinted layers created in DI-MMMs using PDA-based MIT and thermal-initiated radical imprinting  
487 polymerization resulted in improved-selectivity TC-imprinted spots with dual identifying function.  
488 Moreover, 50% of the porosity of composite membranes could be achieved for all prepared membranes  
489 in three operating cycles. Membrane showed excellent selectivity and removed 3.60, 3.33, and 3.51 for  
490 CEX/TC,  $\beta$ SMZ/TC, and  $\beta$ OTC/TC.

491 Wu et al. (2021b) presented the manufacture of dual-imprinted GO/ $\text{SiO}_2$  membranes (GS-DIMs)  
492 with excellent reabsorption capability for selective choice separation and elimination of TC via pressure  
493 filtering of TC-imprinted GO/ $\text{SiO}_2$  nanocomposite sheets by two-stage polydopamine (PDA) imprinting  
494 (Fig. 7a). These PDA-based MIPs were able to create stable complexation with identical sites that were  
495 complementary in size and shape to TC molecules, allowing them to form chelates with TC molecules.  
496 The membrane/GO/ $\text{SiO}_2$  contact would be more adherent with the PDA-modified supports. The amount  
497 of TC adsorption sites on GS-DIMs increased their ability to rebind considerably (more than 3.5). The  
498 TC-permeation efficacy ( $\beta$  value) of the GS-DIMs was 4.27, 3.88, and 4.11, indicating that the GS-DIMs  
499 were highly selective. One GS-DIM retained a good rebinding efficiency for the TC molecule after ten  
500 cycles of adsorption/desorption, losing just 8.9% of its maximum rebinding capability. They have  
501 recently designed dual-imprinted mixed matrix membranes loaded with GO/ $\text{TiO}_2$  (Wu et al., 2022). The

502 GT-DIMs' dual-imprinted approach enabled high permselectivity (5.0) of TC in complexation rejection  
503 systems and synthetic wastewater samples and better rebinding (70.63 mg/g) at a 30-minute adsorption  
504 equilibrium rate (Fig. 7b).

505 **Fig. 7 about here**

506 These findings suggested that porous dual-imprinted PDA membranes supported by GO and  
507 functional nanomaterials-loaded nanocomposite structures could be useful for a range of other selective  
508 separation applications, including practical wastewater samples and industrial-scale applications.

#### 509 5.4.3. GO-Biopolymer mediated membranes

510 Researchers have recently focused on developing a high-efficiency, low-cost biopolymer membrane for  
511 wastewater treatment. Bio-based polymers have the ability to link diverse molecules and are renewable,  
512 plentiful, and cost-effective (Tian et al., 2011; Silvestro et al., 2021). Some bio-based polymers that could  
513 be used in matrix membranes to improve their hydrophilic characteristics and permeability include  
514 cellulose acetate (CA), chitosan (CS), sodium alginate, and carboxymethyl cellulose (Mondal and  
515 Purkait, 2018; Lou et al., 2020).

516 According to Tian et al. (2011), CA (a bio-based polymer) is also used as a polymer matrix  
517 membrane because of its superior film forming ability, biocompatibility, environmental friendliness, and  
518 potential affinity for water penetration and metal ion rejection properties. Due to its poor mechanical and  
519 thermal qualities, CA, on the other hand, is inappropriate for NF (Sabir et al., 2015). CA is combined  
520 with GO to advance membranes' chemical and mechanical characteristics (Silvestro et al., 2021; Wang  
521 et al., 2021).

522 Wang et al. (2021) used an electrostatic self-assembly method to modify GO layers by injecting Co-  
523 Al LDH. They efficiently deposited on cellulose acetate (CA) membranes in various ratios (Fig. 8). They  
524 found that the Co-Al LDH contents are proportionate to the water affinity of GO/Co-Al LDH. The  
525 increase in flow was linked to the rise in Co-Al LDH, which widens the interlayer gap among GO,  
526 resulting in more outstanding water networks that allow easy movement of water molecules. The  
527 composite membrane has a high rejection rate (>90%) for n-hexane, Congo red (CR), rhodamine b  
528 (RhB), petroleum ether, methylene blue (MB), and mesitylene in vacuum filtration (VF). Other  
529 composite membranes have a rejection efficiency and flux of 87.16 percent, whereas 10 ppm MB has a  
530 rejection efficacy and flux of 97.19 percent. At a greater level of MB (80 ppm MB), it indicated 93.75%,  
531 which was still higher. Figure 8e shows that GO/Co-Al-2 has a high rejection efficiency (98%) for three

532 oils in water suspensions such as mesitylene suspension, n-hexane emulsion, and petroleum ether  
533 suspension, with corresponding membrane fluxes of 47.47, 83.28, and 57.35 Lm<sup>-2</sup> h<sup>-1</sup> bar<sup>-1</sup>., indicating  
534 about complete separation of these emulsions. Creating polymeric networks can generally prevent  
535 polymeric materials from breaking down and improve their mechanical properties.

536 **Fig. 8 about here**

537 Like CA, chitosan has attracted substantial attention as a sorbent material, but its hydrophilic nature  
538 may interfere with nonpolar or hydrophobic pesticide interactions (Sanchez-Martin et al., 2006).  
539 Although the strong hydrophilicity of amine groups made CS less stable in an acid medium, however,  
540 amine groups of CS are known to be excellent reactive sites for network development and are promising  
541 for polymeric networks to prevent dissolution and improve mechanical characteristics. Achieving  
542 physical linkages with anionic crosslinkers (genipin, epichlorohydrin, glutaraldehyde) or physical  
543 crosslinking with anionic crosslinkers (sulfosuccinic acid, sodium citrate, oxalic acid, and sodium  
544 triphosphate) has permitted the use of CS in a broader range of applications (Berger et al., 2004;  
545 Silvestro et al., 2021).

546 Silvestro (2021) discovered that a high GO content could successfully control the CS adsorption  
547 dynamics, favoring contacts with water repellent pollutants and decreasing exchanges with water-loving  
548 pollutants, using a glutaraldehyde crosslinker to assist chitosan in biopolymers-GO composite  
549 membranes. The pollutant recovery capacity (R percent) shows that the membrane water resistance  
550 influenced the yields. Pollutants that carry the octanol/water partition coefficient (log Kow) values (0.8  
551 to 2) showed a small R% when CS or CS-GO with a low GO concentration (1-5%) was used. This may  
552 be addressed by the composite membranes' more excellent hydrophilic character, which allowed for more  
553 robust interaction with contaminants yet impaired the extraction phase. The incorporation of both  
554 hydrophilic groups (COOH and OH groups in GO and NH<sub>2</sub> and OH groups in CS) and hydrophobic  
555 regions, on the other hand, allowed for a beneficial interface between the low log Kow pesticides and  
556 compound membranes. Similar results were obtained when log Kow 2 and 3 were used, but when log  
557 Kow 3 and 6 were utilized, the highest R percent was obtained. Increasing the matrix's hydrophobicity  
558 did not influence the reclamation of more water-repellent pollutants, which improved in lockstep with  
559 the rise in GO percentage (Fig. 8f).

560 The 20% GO chitosan/polyacrylamide/graphene oxide nanocomposites (CAGs) can achieve  
561 maximal adsorption of methylene blue. On the other hand, the magnetic CS-GO nanocomposites

562 removed rifampicin with a removal efficiency of over 95% (Silvestro et al., 2021). This behavior  
563 demonstrated that creating hydrophobic association ( $\pi$ -stacking) and hydrogen linkages may be regulated  
564 via changing the GO concentration in the CS integrated membranes, resulting in better pollutant  
565 extraction.

## 566 **6. GO assisted adsorptive membranes**

### 567 **6.1. GO nanoparticles as adsorption material in membranes**

568 Diverse kinds of membrane filtering apparatus have been implemented for water purification, such as  
569 MF, NF, UF and RO filtration (Fig. 9a). The exceptional purity of effluents from NF/RO treatment  
570 supports their use in post-treatment in wastewater/sewage treatment plants (Ma et al., 2022).  
571 Understanding the underlying mechanisms (size omission, hydrophobic bonding, and electrostatic  
572 interaction) is essential for NF/RO processes to remove EOMPs with varying physicochemical  
573 characteristics (size, hydrophobicity, hydrophobicity, solubility charge, and diffusivity) (Fig. 9b)  
574 (Ojajuni et al., 2015; Khanzada et al., 2020). Hydrophobicity has recently been found to significantly  
575 correlate with molecular weight (MW) by (Licona et al., 2018; Pollitt et al., 2019), indicating that size  
576 exclusion and adsorption are the most important removal processes.

#### 577 **Fig. 9 about here**

578 Bisphenol A can be removed effectively using RO and NF membranes. For example, Bolong et al.  
579 (2010) combined a negative charge polymer with polyethersulfone nanofiltration membranes and  
580 discovered that at a pH of 14-8.0, the retention rate of BPA can surpass 90%. Kim et al. (2008) reported  
581 removing more than 95% of BPA from polyamide NF membranes by surface grafting using  
582 ethylenediamine, methacrylic acid and succinic acid. Commercial polysulfone ultrafiltration membranes  
583 have a capacity of just 16.6 mg m<sup>2</sup> for BPA adsorption (Wu et al., 2010). Regardless of how effectively  
584 BPA is retained by RO and NF membranes which are limited in application by small permeability and  
585 more operating pressure (Schrotter and Bozkaya-Schrotter, 2010). While the permeability of the affinity  
586 membrane may be adequate, one of the most important measures is its adsorption capacity. Although  
587 Heo et al. (2012) used carbon nanotubes and a polyethersulfone UF membrane system to study BPA  
588 adsorption, the most effective rates and capabilities are likely to be larger than 90% and 60 mg m<sup>2</sup>,  
589 respectively.

590 For industrial applications, higher adsorption capacity is still desired. Chen et al. (2020) created  
591 graphene oxide (GO) nanosheets modified with -cyclodextrin ( $\beta$ -CD) to eliminate BPA (CDGO). The

592 grafted  $\beta$ -CD molecules provide adsorption spots on the surface of 2D nanochannel-containing CDGO  
593 membranes. Through host-guest recognition,  $\beta$ -CD, as a molecular chelator, may create serpentine  
594 complexes with BPA molecules (Alsaiee et al., 2016). While GO nanosheets were chosen, (i) GO-  
595 associated functional groups are easy to modify with  $\beta$ -CD molecules, and (ii) because they are single  
596 atomic layer materials with a lot of specific surface area for adsorption (Wang et al., 2014).

597 The researchers claimed that ethanol washing could renew the CDGO composite membranes, and  
598 100% restoration efficiency can be achieved after many cycles in the BPA exclusion process. Moreover,  
599 in comparison to standard GO, NF, or RO membranes, CDGO may have high flux and remarked  
600 adsorption performance for efficient BPA retention due to (i) plentiful adsorption positions on the surface  
601 of newly formed nanochannel and (ii) surface decorating of GO nanosheets with  $\beta$ -CD molecules, where  
602 the CDGO nanosheets may be crumpled with the fixing of  $\beta$ -CD molecules which compact the  
603 dimensions of 2D nanochannels in CDGO membranes (Chen et al., 2020).

604 **Table 2 about here**

605 To summarize, manufacturing high-permeability membranes with high adsorption capacity for  
606 efficient BPA removal remains challenging. Several studies have shown a number of advantages and  
607 limitations regarding GO-mediated adsorptive membranes (Table 2). During a four-hour treatment,  
608 Zambianchi et al. (2017) removed 90% of a variety of OMPs, including PhACs, pigments, PPCPs, and  
609 surfactants. Cho et al. (2019) demonstrated the use of GO nanoribbons (GONRs) to build composite  
610 membranes results in rapid water permeation and precise molecule separation >95 percent for OMPs  
611 with a larger hydration radius of 5 Å. Fryczkowska et al. (2020) created composite membranes that  
612 retained a significant protein (85 percent). When covalently coupled to GO, NF membranes maintain  
613 85.03% of rhodamine B, and 95.43 percent when covalently linked to GO/polyacrylamide (Cheng et al.,  
614 2019).

615 According to Nguyen et al. (2021), the most critical factor impacting adsorption efficacy appears to  
616 be outer surface adsorption. Owing to colloidal instability, the structure, surface charge, shape, and  
617 conformation of nanoparticles in an aqueous solution might affect the contact of surface area. They found  
618 removal processes comprise (i) adsorption by hydrophobic contacts and electrostatic linkages between  
619 charged species (a part of van der Waals) (Dugyala et al., 2016); start at long distance (up to 10 nm), (ii)  
620 adsorption via  $\pi/\pi$  stacking (Ghosh et al., 2019), XH /  $\pi$  (X = C, O) contacts (Yaya et al., 2019), van der

621 Waals attractions (Thierfelder et al., 2011), and O···H hydrogen bonding (Jin et al., 2015); at  
622 comparatively low ranges (0.2–0.5 nm) (Fig. 10a).

623 **Fig. 10 about here**

624 Nonetheless, developing a single plausible explanation for the observed molecule adsorption  
625 tendencies in all carbon-based materials is challenging. In contrast to aromatic interactions, GO-based  
626 desorption is controlled by linkages between basal plane groups such as epoxy, -COOH C=O, and -OH.  
627 This could be because there are fewer  $\pi$ -rings than functional groups. Furthermore, adsorption is a  
628 complicated process governed by quite a few adsorbate parameters, including size, branching extent, and  
629 solvent polarizability, which are hard to quantify (Bunmahotama et al., 2020). Nevertheless, carbon  
630 nanotubes and GO are generally interesting for composite membrane integration, shorter residence  
631 periods, and substantial adsorption (Nguyen et al., 2021).

## 632 **6.2. Metal/nanomaterial incorporated GO integrated biodegradable polymer membranes**

633 Membrane separation is suited for phenol removal because of its high surface area, microporous  
634 structure, superior adsorption capacity, purity, and simplicity of access. Nevertheless, low removal rates  
635 with various filter technologies and high operating pressure are frequently constraints (Nguyen et al.,  
636 2021). The NF and RO both demand higher pressures, which raises operating expenses (Luo et al.,  
637 2014b). However, UF is ineffective for specific contaminants like hormones (Li et al., 2021b). Li et al.  
638 (2017) hypothesized that a nano-augmented membrane (i.e., NPs incorporated membrane) could  
639 efficiently remove organic contaminants utilizing ultrafiltration.

640 Recently, Nguyen et al. (2021) looked into six different types of carbon-based nanoparticles,  
641 including graphene oxide, graphene in two grades, multi- and single-walled CNTs, and fullerene C60.  
642 Graphene and CNPs demonstrated substantial uptake capacity for hormones that couldn't be quantified  
643 in isotherm investigations (more than 10 mg g<sup>-1</sup>), and all NPs had swift kinetics. Moreover, based on the  
644 linear relationship between supernatant concentration and adsorbed mass, GO enhanced membrane had  
645 > 10 mg g<sup>-1</sup> adsorption efficiency, which was significantly more than that of GP-AO2 (6.59 mg g<sup>-1</sup>) and  
646 C60 (1.78 mg g<sup>-1</sup>). Moreover, GO nonaugmented membranes had many times the Langmuir-based E2  
647 adsorption capacity (150 mg g<sup>-1</sup>) elsewhere, even though cellulose acetate (CA) membrane filters  
648 contributed significantly to adsorption (up to 80%) (Jiang et al., 2016a).

649 Recently, Li et al. (2021b) coated and grafted tannic acid(TA)-aminopropyltriethoxysilane  
650 (TAAPTES) on the membrane ferric ions (Fe<sup>3+</sup>) to create a GO membrane with (interlayer gap and

651 transverse defects) (Fig. 10a). The GO membrane is first prepared using Fe<sup>3+</sup>-mediated GO nanosheets.  
652 Then, the surface of the prepared membrane was coated with TA-APTES, which has a high affinity for  
653 Fe<sup>3+</sup>; thicker TA-APTES was used to repair Fe<sup>3+</sup> attributed to larger membrane defects. This "speckled"  
654 TA-APTES coating improved the antifouling capabilities while nanochannels in the GO membrane were  
655 regulated. However, it was unclear whether the presence of Fe<sup>3+</sup> caused the selective placement of TA-  
656 APTES complex on GO membrane defects and or whether the pore size distribution was constrained.  
657 The results showed that TA-APTES modification of the GO-Fe-coated membrane increased the rejection  
658 of the endocrine disruptor BPA (BPA, 228 Da) from 26 to 81%, with minor porousness damage following  
659 BPA separation.

660 In contrast, the permeability cost of the GO-Fe membrane was over 36% after filtering 50 mL BPA  
661 solution. However, TA-APTES coating compromised BPA retention/in the GO-Fe membrane owing to  
662 a considerable rise in antifouling properties. The substantial BPA removal of the GO-Fe-containing  
663 membrane can be related to enhancing size segregation. Neutral OMP size segregation and adsorption  
664 are thought to be the main rejection processes in NF (Chen et al., 2020; Yuan et al., 2020)

### 665 **6.3. GO in biomimetic membrane (BM)**

666 Specific contaminants, like hormones, are resistant to ultrafiltration (Nguyen et al., 2021; Zhang et al.,  
667 2022). A carbon-coated nanofiltration membrane can efficiently remove phenol at pressures up to 8 bar  
668 (Cetinkaya and Ozdemir, 2018). However, a high-pressure membrane is not cost-effective. At 3 bar  
669 pressure, a carbon-coated UF membrane removed 93% of EOMPs, yet UF rarely rejected  
670 micropollutants, and the reactive membrane's removal efficacy was proportional to membrane flow  
671 (Zhang et al., 2022).

672 Increased water flux means less membrane contact time with EOMPs, which necessitates a faster  
673 oxidation rate. Coatings of cobalt carbon, for example, can eliminate 95 percent of EOMPs while  
674 producing more significant fluxes (>120 L/m<sup>2</sup> h) because of the weak oxidation rates of micropollutants  
675 compared to UF flux. Laccase immobilized membranes had a lower water flux (50 L/m<sup>2</sup> h) but a higher  
676 phenol and other EOMPs clearance ratio (82–96%) (Zhu et al., 2020a; Zhu et al., 2020b). They can,  
677 however, increase clearance at higher flux rates when paired with metal NPs and laccase. GO mediated  
678 gravity-driven biomimetic membrane (GDBM) showed 84.32% removal with 25~70 mg L<sup>-1</sup> BPA  
679 removal rate and showed prominent stability (LMH: 27~42 and BPA removal: 55~70%) during 30 days'  
680 operation (Zhu et al., 2020b).

681 Biomimetic membranes with co-immobilized laccase and adsorption transporter on the membrane  
682 have recently been lauded as a "green" and long-lasting membrane technology (Zhu et al., 2020a). On-  
683 membrane enzymes help break down EOMPs and reduce foulants, extending the life of biocatalytic  
684 membranes (Zhu et al., 2020a; Bourassi et al., 2021). The enzyme-fouling layers generated as a result of  
685 enzyme immobilization, on the other hand, significantly improve filtration resistance, but regeneration  
686 is complex (Zhang et al., 2021).

687 According to Soroush et al. (2016), in situ generations of silver nanoparticles (AgNPs) using  
688 graphene oxide nanosheets greatly boosted bacterial inactivation (98%) in forward osmosis (FO), thin-  
689 film composite (TFC) membranes. Ag nanoparticles were successfully regenerated on Go-Ag modified  
690 membranes. Likewise, while carbon nanotube coating and chemical immobilization effectively prevent  
691 biofouling by isolating enzymes, the crosslinking agents used in coating and chemical immobilization  
692 alter permeability (Vanherck et al., 2013). To overcome this problem, Zhu et al.(2020a) employed GO  
693 as carriers and "coated" them with enzymes (laccase) that were physically adsorbed on the "dynamic"  
694 carrier layer on the BDM surface (Fig. 10b). The results showed that when CNT/GO concentrations (20–  
695 45 g m<sup>2</sup>) increased, the CNT/GO&Laccase@UF membrane's EOMPs removal rate increased  
696 proportionally to the biocatalytic membrane's adsorption capacity. Mixed-filtration BDM performed  
697 better than stepwise-filtration BDM.

698 Laccase activity was higher on GO than CNT. Flux and removal rate fluctuated significantly with  
699 certain laccase dosages. Enhancing GO or CNT level increased rejection efficiency but decreased flux,  
700 and after 5 cycles, removal rate and flux reduced by 10–15%. At 65 g m<sup>-2</sup>, the elimination rate for GO-  
701 BDM increased to 98 percent and for CNT-BDM to 87 percent. Increased CNT/GO dosage increased  
702 removal rate but decreased fluxes. In contrast, increased laccase dosage changed both flux rates and  
703 removal rates, implying that a thicker CNT/GO layer results in a smaller pore size and a longer membrane  
704 penetration channel. At the optimal laccase dosage of 50 g m<sup>-2</sup>, the 25 g m<sup>-2</sup> CNT/GO dose with ideal  
705 laccase action (3.97 and 3.24 U cm<sup>2</sup> for GO-BDM and CNT-BDM, respectively) maintained fluxes above  
706 100 L m<sup>-2</sup> h<sup>-1</sup> and a clearance rate greater than 65 percent, demonstrating that laccase growth and catalytic  
707 strength did not become effective by additional CNT/GO. Overall, the most efficient production process  
708 for BDM for water/wastewater treatment was CNT/GO&Lac@UF (a combination of laccase filtration  
709 and CNT/GO on UF).

710 Biomimetic membrane (BM) can effectively remove a variety of MOPs, including dyes, and  
711 endocrine hormones, and antibiotics, in a closed membrane apparatus system while maintaining a low  
712 membrane fouling rate and a permeability greater than  $20 \text{ L m}^{-2} \text{ h}^{-1}$  (LMH)/bar, allowing BM to attain a  
713 "high-pressure membrane effect" with "low-pressure membrane operation." However, these membranes  
714 are only useful in open natural therapy circumstances due to the lack of a filtering driving force. In open  
715 natural water systems, gravity-driven membranes (GDMs) have recently emerged as promising water  
716 filtration alternatives (Zhu et al., 2020b). Due to its independence from an outer accelerating force, high  
717 level of stability, cheap cost of energy, positive financial value, and simplicity of use and servicing,  
718 GDMs with GO showed soy sauce polluted natural water treatment (Pronk et al., 2019).

719 Zhu et al. (2020b) used to GO, CNT (carbon nanotubes), and activated carbon (AC) in GDBM and  
720 found that GO-GDBM had a greater permeate flow (40.33 LMH) than CNT-GDBM (30.95 LMH) or  
721 AC-GDBM (12.52 LMH). Furthermore, GO-GDBM was found to be much more effective at removing  
722 micropollutants due to its enzymatic efficiency and adsorption capacity (exceeding 60 percent). Overall,  
723 after 8 hours, GO-GDBM is stable and feasible, with >75 percent phenol elimination and >15 LMH  
724 fluxes. Laccase enrichment and accumulation are facilitated by the homogeneous mixing of GO and  
725 laccase. The GO is required to catalyze laccase's second copper ion binding T2/T3 active site. In contrast,  
726 azide ions bound the T2/T3 trinuclear copper group stabilized the native intermediate of oxidized laccase,  
727 bolstering GO and laccase's ability to remove micropollutants.

#### 728 6.3.1. GO assisted PES membrane in EMOPs removal

729 Zhang et al. (2021) demonstrated the catalytic oxidation removal of endocrine disruptor chemicals in a  
730 PES membrane employing an enzyme-like ligand, Iron (III)- tetra-amido macrocyclic ligand (Fe(III)-  
731 TAML). Fe(III)-TAML is activated by  $\text{H}_2\text{O}_2$  to produce high valence Fe. The bulk of EOMPs was  
732 efficiently removed using ultrafiltration with a TAML/GO-PEI/PES membrane. Porosity and  
733 hydrophilicity were increased with GO-PEI addition in the PES membrane. The complementary nature  
734 of GO and TAML was identified; TAML's improved oxidation performance improves the UF  
735 membrane's micropollutant removal at a high flux (Fig. 11).

#### 736 **Fig. 11 about here**

737 The degradation routes suggested that the owing to C–C bond cessation, BPA (P1) breakdown into  
738 degraded phenol (P5) and 4-isopropylphenol (P2), and the P2 would again oxidize to hydroquinone (P4).  
739 On the other hand, the GO additive membrane's dense pore structure offered a superior substrate for

740 TAML loading. The membrane's tiny pore size promoted micropollutant contact with the TAML, which  
741 was beneficial for oxidation. Furthermore, 1 mmol L<sup>-1</sup> H<sub>2</sub>O<sub>2</sub> showed sufficient removal (100%) of 10 g/L  
742 BPA from the feed water maintained at the high flux of 192 L<sup>-1</sup>m<sup>2</sup> h. The re-adsorption of the adsorbed  
743 Fe(III)-TAML on the GO-PEI membrane was simple, and the BPA flux and removal efficiency remained  
744 consistent during five cycles. The marketability of immobilized UF with an enzyme-like ligand  
745 highlights the scope of enzyme-like ligand catalytic oxidation in combination with UF to remove EOMPs  
746 at an excellent flux and ensure drinking water safety.

## 747 **7. Enzymatic modified polyethersulfone (PES) membranes**

748 Niebergall and colleagues (2014) used a wet-phase inversion method to introduce nanospheres with  
749 absorbency features into PES matrices (Xu et al., 2017). The produced nanocomposite membrane  
750 absorbent was then employed to remove BPA since it possessed an adsorption capacity of 7.7 mg m<sup>-2</sup> at  
751 a pH of 3.5 (Niedergall et al., 2014). Wu and colleagues discovered that their membrane absorbers were  
752 99.8% effective at removing endotoxins (Wu et al., 2017). However, adsorption saturation and pollutant  
753 leaks occur during regeneration, resulting in considerable secondary contamination.

754 Biocatalytic membranes can directly eliminate micropollutants, avoiding the "adsorption saturation"  
755 limit (Zhang et al., 2019). Immobilized laccase on (1) nano-copper integrated electrospun fibers showed  
756 a 95.4 percent removal efficiency of 2,4,6-trichlorophenol (Xu et al., 2017), and (2) 4-methoxyphenol  
757 was efficiently removed from functionalized CNTs/polysulfone (Costa et al., 2019). Despite this, the  
758 selection ability of membranes is compromised owing to the usual dipping contact manner application  
759 approach.

760 Laccase solution filtration by reverse filtration followed by dopamine (DA) coating layer enable its  
761 application for rejection, sorting, or catalysis (Cao et al., 2016). In an alkaline environment, numerous  
762 substrates can be covered with a durable polydopamine (PDA) coating layer that maintains enzymes  
763 within the membrane subnetwork by various processes (Cao et al., 2016). However, this biocatalytic  
764 membrane has poor solubility and is susceptible to enzymatic leakage and product accumulation, leading  
765 to membrane clogging and product interference (Cao et al., 2018).

766 The development of almost unhindered 2D nanochannels with a powerful rejection capability has  
767 resulted in a greater permeability for GO-based membranes (Han et al., 2018). Authors also offer  
768 excellent transport qualities, low manufacturing costs, elasticity, and chemical tolerance (Chen et al.,  
769 2018a; Chen et al., 2018b; Wang et al., 2018). A sulfonated GO altered PVDF membrane could improve

770 water flux by 146.6% (Ayyaru and Ahn, 2017), and a binding enzyme to GO nanosheets can generate a  
771 significant theoretical SSA ( $2630 \text{ m}^2 \text{ g}^{-1}$ ) with better adsorption potential for EMOPS  
772 (Phatthanakittiphong and Seo, 2016), and a holding enzyme to GO nanosheets can give enhanced  
773 stability and activity (Vanherck et al., 2013; Zhu et al., 2020a; Zhang et al., 2021). The Hanes-Woolf  
774 method was used to measure Laccase's  $V_{\text{max}}$  and  $K_{\text{m}}$  values, which indicated a 47% increase following  
775 immobility on Go, outperforming other immobilized approaches. The  $K_{\text{m}}$  values for GO-Zn-E and GO-  
776  $\text{NH}_2$ -E were lower than that for GO-E, implying a lower negatively charged controlled conditions could  
777 boost substrate attraction. Positive groups like  $\text{Zn}^{2+}$  and  $-\text{NH}_2$  may alter the direction of contacts between  
778 GO derivatives and enzymes, resulting in increased affinity and more active laccase sites exposed to  
779 substrate. GO-E and GO-Zn-E demonstrated enhanced durability and continued capacity to remove BPA  
780 after 72 hours.

781 GOZn-E also had a better removal efficiency than GO-E, which is most likely due to its increased  
782 stability. BPA oxidation derivatives carrying a positive charge may also bind to GO via hydrophobic and  
783 electrostatic adsorption (Lee et al., 2013; Chu et al., 2017). Laccase restriction was reduced in the positive  
784 charge groups  $\text{Zn}^{2+}$  on GO-Zn, limiting the electrostatic bonding of derivatives, while GO and GO-Zn-  
785 based laccase boosted enzyme action and stability relative to soluble laccase. These observations also  
786 validated the inconsistent results. Consequently, GO-Zn and GO were added to boost the biocatalytic  
787 membrane's functionality. The permeable membrane obtained by reverse filtering GO into NF270  
788 improved by 14.8 % to the pristine membrane. Because the PDA layer increased the membrane's  
789 hydrophilicity (Luo et al., 2014b), the PDA coating restored some of the lost permeability. The enzyme  
790 filling of NF-E, reNF-E/GO and reNF-E/GO-Zn was nearly identical, reaching 60%. Reverse filtration  
791 and co-deposition can generate GO-based biocatalytic membranes. Higher permeate flow ought to  
792 accelerate permeate clearance and enhance biocatalytic membrane re-utilization, but it did not appear in  
793 greater BPA rejection by the membrane or adequate interacting time between BPA and laccase through  
794 GO adsorption. BPA may be detected semi-quantitatively using GO-based membranes with increased  
795 storage durability. These findings reveal the involvement of GO and its derivatives in laccase  
796 immobilization and biocatalytic membrane production, which can be used for EOMPs detection and  
797 elimination.

## 798 **8. GO assisted membrane in EOMPs degradation studies**

799 Wastewater influents are typically treated for pathogens and suspended or flocculated debris, but EOMPs  
800 like PhACs remain untreated due to their trace level (Mladenov et al., 2022). Effective ways for removing  
801 PhACs from wastewater include adsorption, advanced oxidation, and membrane filtration. Combining  
802 of more than one strategy is promising and could accelerate the performance of the water treatment  
803 system. Particularly, membrane filtration facilitates the degradation process and can increase 2.15 times  
804 degradation efficiency of the system (Gao et al., 2020). However, GO-based membranes are still not  
805 much efficient for organic contaminated water filtering due to (i) limited catalytic activity, (ii) the  
806 substantial tailoring necessary for intercalated materials, (iii) and delayed catalytic kinetics.

807 Previous studies as shown in table 3, claim the desirable removal capacity of GO for atenolol,  
808 propranolol, caffeine, carbamazepine, diclofenac, acetaminophen, acetylsalicylic acid and ketoprofen  
809 when integrated with catalytic membranes (Xu et al., 2019; Bhatia et al., 2021).

810 **Table 3 about here**

### 811 **8.1. Catalytic Membranes**

812 In EOMPs contaminated water treatment, sulfate radical-mediated advanced oxidation processes (SR-  
813 AOPs) is a modern research hotspot due to the formation of reactive oxygen species (ROS) via  
814 peroxymonosulfate (PMS) stimulation (Ye et al., 2021a). Catalyst performance and practical application  
815 are two essential variables in PMS activation in wastewater treatment (PMS). Ye et al. (2021b) formed  
816 a composite membrane supported by GO (Fig. 12a). Firstly, they formed a 1D/2D nitrogen-doped carbon  
817 array with controlled FexOy by in-situ transmutation of ZIF@MILs-20. Then, this dual metal-organic  
818 framework was intercalated into adjacent GO. The 1D/2D associative porous ZIF@MILs-20 structures  
819 supported by restricted FexOy and increased oxygen vacancies were beneficial for removing NX via a  
820 collaborative "trap-and-zap" approach for PMS stimulation. The ZIF@MILs-20/PMS system eliminated  
821 99.4% of NX after 40 minutes. However, the degradation process generates several intermediate  
822 including hydroxyprolyl-tryptophan, oglufanide, mitromycin despropylene gatifloxacin,  
823 oxonorfloxacin, desethylene norfloxacin hydrochloride, 5-ethyl-5-(4-fluorophenyl)-1,3-diazinane-2,4,6-  
824 trione, flumequine and n-feruloylglycinate. When comparing the removal efficiencies of many persistent  
825 organic pollutants, such as perfluorooctanoic acid (PFOA), CBZ, and BPA catalytic membranes were  
826 shown to achieve ultra-high removal efficiencies (89.3, 95.7, and 100%, respectively) than powder  
827 catalyst (43.5, and 76.3 and 94.3%, respectively). Additionally, after five cycles, the ZIF@MILs-20/GO

828 membrane retained over 90% SMX degradation efficiencies, verifying catalytic membrane (ZIF@MILs-  
829 20/GO) with high stability and reusability.

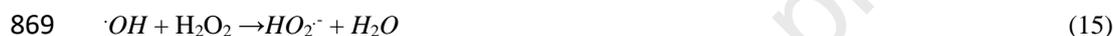
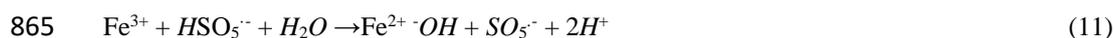
830 **Fig. 12 about here**

831 With a capacity of 165.3 mg g<sup>-1</sup>, the ZIF/MILs-20/GO membrane had the best capacity for NX  
832 adsorption, demonstrating that the GO increased catalyst wettability by lowering the reaction space of  
833 ROS and active sites (Zhu et al., 2018). The ZIF@MILs-20/GO framework exhibited simultaneous  
834 catalytic oxidation and separation of organics with a constant flow of 156.3 Lm<sup>-2</sup>h<sup>-1</sup>bar<sup>-1</sup>, considerably  
835 overcoming membrane fouling. Furthermore, singlet oxygen (<sup>1</sup>O<sub>2</sub>) has been shown to play an important  
836 role in the breakdown of NX. In a nutshell, using catalytic membranes to treat wastewater is a novel  
837 concept.

838 The proposed mechanisms showed Fe<sup>0</sup> peak vanished completely at first, whereas the Fe<sup>2+</sup> peak  
839 shifted slightly, indicating that Fe<sup>0</sup> countered with electrons to free Fe<sup>2+</sup>, as provided in Eqs. 1 and 2. In  
840 particular, Fe<sup>0</sup> can reutilize Fe<sup>3+</sup> by replacing Fe<sup>2+</sup>, as shown in Eqs. 3–5. Furthermore, Fe<sup>2+</sup> regeneration  
841 may occur by electron transport at N species and transition metal NPs interface, boosting catalytic  
842 activity even more (Eq. 6) (Luo et al., 2011). Alongside, sp<sup>2</sup> hybridization of pyridinic N can generate a  
843 -system by connecting two carbon atoms or donating one p electron (Liu et al., 2020a). Furthermore, as  
844 demonstrated in Eqs. 7 and 8, free electrons will combine with the lone-pair electrons of the N dopant,  
845 accelerating PMS activation. Additionally, it has been established that SO<sub>4</sub><sup>-</sup> interacts with H<sub>2</sub>O to yield  
846 OH (Eq. 9).

847 The synthesis of SO<sub>5</sub><sup>-</sup> was aided by the presence of hydroxyl groups on the catalyst surfaces, which  
848 aided in the breakdown of PMS molecules (Eqs. 10 and 11). In contrast, the reaction between H<sub>2</sub>O and  
849 SO<sub>5</sub><sup>-</sup> aided in forming <sup>1</sup>O<sub>2</sub> and accelerated PMS degradation (Eqs. 12 and 13) (Wang et al., 2019).  
850 Because the redox potential of O<sub>2</sub><sup>-</sup> acquired since the catalytic process for pollutant removal was weak,  
851 it was changed to <sup>1</sup>O<sub>2</sub> (Eqs. 14–17) (Hu et al., 2020). Some <sup>1</sup>O<sub>2</sub> can be made by reacting H<sub>2</sub>O with OH  
852 (Eq. 18). While Eq. 19 shows the <sup>1</sup>O<sub>2</sub> formation from oxygen vacancies and PMS molecules (Zeng et al.,  
853 2020). Finally, Eq. 20 shows that a mix of radicals and non-radicals increased NX breakdown, resulting  
854 in totally mineralized NX molecules converted to CO<sub>2</sub> and H<sub>2</sub>O.





875 Overall in the ZIF@MILs-20/GO/PMS system, the degradation process and contribution of relevant  
 876 ROS were discovered, indicating that OH had a minor part in NX degradation whereas  $\text{O}_4^-$  was highly  
 877 essential. Furthermore, NX degradation was aided by both free radicals and non-radicals, with  $\text{SO}_4^-$  and  
 878  $^1\text{O}_2$  being substantial contributors (Fig. 12a).

## 879 8.2. Photo-Fenton catalytic membranes

880 Intercalating photo-Fenton catalytic Fe-based MOFs into GO nanosheets regulates two-dimensional  
 881 nanochannels while simultaneously endowing the membrane with photo-Fenton self-cleaning facility.  
 882 More importantly, the inclusion of GO enhances the photo-Fenton catalytic properties of the membranes  
 883 by increasing the segregation of photogenerated charge carriers due to its big SSA, better electrical  
 884 conduction, and excellent charge carrier capacity (Yang et al., 2017a; Liu et al., 2018). As a result, a  
 885 membrane comprising GO/Fe(III)- intercalating MOF integrated with photo-Fenton activities and  
 886 membrane separation was developed for better water purification. Likewise, MIL-88A intercalated onto  
 887 neighboring GO nanosheets as a restricted catalytic membrane in an oil-water separation system

888 demonstrated a self-cleaning photocatalytic reaction (Xie et al., 2020a). Furthermore, at constant flux,  
 889 the graphene oxide/MIL-88A(Fe) (GO/M88A) composite membrane maintained an excellent MB  
 890 separation (97.87%) even after 12 cycles.

891 Additionally, photo-Fenton catalytic breakdown of pollutants utilizing membranes was 98.81% for  
 892 MB and 97.27% for BPA (Xie et al., 2020a). The activated GO/M88A membrane release  $e^-h^+$  pairs in  
 893 response to visible light irradiation. Impurities (R) were directly oxidized by  $h^+$  while transport of  $e^-$  to GO  
 894 surface enhanced the photoinduced transporter potential for separation and facilitated OH production by  
 895  $H_2O_2$  breakdown. Meanwhile,  $O_2$  can collect  $e^-$  and convert it to the highly active  $O_2^{\cdot-}$  species on  
 896 GO/M88A surface. Contaminants can be oxidized entirely and degraded by the highly reactive OH and  
 897  $O_2^{\cdot-}$  species (R). Fe—O clusters can also use Fenton-like reactions to expedite the decomposition of  $H_2O_2$   
 898 into OH. Furthermore, the GO nanosheets provide many active catalytic sites, resulting in higher  
 899 photocatalytic activity (Li et al., 2018). Impurities collected on the membrane's surface may be quickly  
 900 removed thanks to the strong photo-Fenton catalytic activity, ensuring a better rejection at a high flux  
 901 (Fig. 12b).

902 The following is a summary of the reactions that have been proposed (Eqs. 21-26):



909 The photo-Fenton degradation of AMX by  $TiO_2$ -GO- $Fe_3O_4$  composites revealed a charge carrier  
 910 pathway between  $TiO_2$ , graphene oxide, and  $Fe_3O_4$ , while  $\cdot OH$  and  $H^+$  dominate the photo-Fenton  
 911 degradation of AMX. In situ embedding  $TiO_2$  nanoparticles on GO sheets and UV light exposure create  
 912 surface roughness and wrinkles that facilitate molecular mobility.

913 A submerged magnetic separation membrane photocatalytic reactor (SMSMPR) using ceramic  
 914 membranes was created to isolated suspended  $TiO_2$ -GO- $Fe_3O_4$  from waste effluent, and this reactor was  
 915 integrated with the  $TiO_2$ -GO- $Fe_3O_4$ . AMX degradation was improved by raising the  $H_2O_2$  concentration  
 916 from 0 to 20 mM from 10.0 to 90.1 percent while decreasing it by increasing the pH from 3.0 to 7.0. At  
 917 pH 3, TOC was removed 90% of the time, while at pH 5, it was only 50%. Furthermore, at pH 3 and pH

918 5, leaching Fe concentrations were 0.75 and 0.59 mg L<sup>-1</sup>, respectively, meeting state wastewater release  
919 standards (1.0 mg L<sup>-1</sup>). Backwashing and magnetic separation in the SMSMPR can improve photo-  
920 Fenton catalytic activity, persistence, and filtration support to strengthen wastewater treatment  
921 applications. The SMSMPR appears well suited for treating acid wastewater because of its better  
922 efficiency and less metal leaching under acidic conditions (Li et al., 2019b).

### 923 **8.3. Photocatalytic Membranes**

924 In previous studies, visible light attributed to TiO<sub>2</sub> activation was enhanced with GO (Li et al., 2019b;  
925 Li et al., 2022a). Thus, OH generation and h<sup>+</sup>/e<sup>-</sup> pair separation improved photocatalytic functions of  
926 GO-TiO<sub>2</sub> obtained (Jiang et al., 2016b; Li et al., 2022a). The photo-Fenton process destroys refractory  
927 organic molecules by reducing Fe<sup>3+</sup> to Fe<sup>2+</sup> through a photoinduced electron (Li et al., 2019b; Xie et al.,  
928 2020a). Several research using TiO<sub>2</sub> and Fe<sub>3</sub>O<sub>4</sub> to recover catalysts have been undertaken (Li et al.,  
929 2019b; Li et al., 2022a). However, a photocatalytic activity typically degrades due to the weak charge  
930 transfer speed between Fe<sub>3</sub>O<sub>4</sub> and TiO<sub>2</sub>. Thus, the long life of this reusable photocatalyst must be  
931 extended. Furthermore, the oxygen functional groups can provide a bonding site to form graphene-supported  
932 metal oxide nanocomposites (Zhao et al., 2013). As a result, TiO<sub>2</sub>, CuO, CdS, ZnO, and Fe<sub>2</sub>O<sub>3</sub> are  
933 typically covalently linked using GO (Xu and Wang, 2012; Wang et al., 2013).

934 To demonstrate photocatalytic degradation of CBZ and DCF Heu et al. (2020) utilized GO in a  
935 zirconium-oriented metal-organic framework (UiO-66). They discovered that ultraviolet (UV)  
936 photocatalytic activity reduced membrane fouling from 20.7% to 2.4% and enhanced FRR to 98%. After  
937 five cycles, the UiO-66 GO/NF-10% composite membrane demonstrated superior physical stability with  
938 a comparatively low mass loss of 8.64% compared to those carrying mass of 5 and 15 wt%. In addition,  
939 irreversible fouling of UiO-66 GO/NF-10% composite membrane decreased from 20.7 to 2.4% and  
940 boosted FFR to 98% with improved photocatalytic activity under ultraviolet (UV) irradiation (bandgap:  
941 3.45 eV).

942 They previously used TiOSO<sub>4</sub> as a Ti source to create a multifunctional GO/TiO<sub>2</sub> membrane for  
943 crossflow filtration (Zhu et al., 2017a). The membrane showed significant photocatalytic degradation of  
944 Methylene Blue when exposed to UV light. UV light irradiation of 100 min dramatically reduced the  
945 membrane fouling, resulting in a 96% flux recovery. Likewise, UV light irradiation of 110 min degraded  
946 92% of MB with a photocatalyst dose of 4 mg. The GO/TiO<sub>2</sub> membrane's exceptional performance  
947 revealed its commercial potential in the quality water supply. Similarly, to simultaneously solve TiO<sub>2</sub>'s

948 catalytic performance, durability, and separation property restrictions, Li and coworkers (Li et al., 2019b)  
949 developed a new approach for co-assembling TiO<sub>2</sub> and Fe<sub>3</sub>O<sub>4</sub> nanoparticles on GO (Fig. 13).

950 **Fig. 13 about here**

951 The TiO<sub>2</sub>-GO-Fe<sub>3</sub>O<sub>4</sub> composite membrane showed that (i) doping GO improves catalytic activity  
952 and stability by facilitating charge transport, and (ii) on-site growing Fe<sub>3</sub>O<sub>4</sub> on the GO surface can  
953 increase heterogeneous Fenton degradation of EOMPS and offers photocatalyst affinity for magnetic  
954 separation in the water cleaning system. In the presence of Amoxicillin (AMX), the oxidation efficiency  
955 of sole TiO<sub>2</sub>-GO and Fe<sub>3</sub>O<sub>4</sub> was 14.7%, but TiO<sub>2</sub>-GO-18wt %Fe<sub>3</sub>O<sub>4</sub> was 18.7%. In the presence of H<sub>2</sub>O<sub>2</sub>,  
956 Fe<sub>3</sub>O<sub>4</sub> composites had a 90% oxidation efficiency. On the other hand, the oxidation efficiency of AMX  
957 in TiO<sub>2</sub>-GO-18wt%Fe<sub>3</sub>O<sub>4</sub> is 22.3% due to Fe<sub>3</sub>O<sub>4</sub> activation and 15.2% due to visible light irradiation,  
958 respectively. However, the performance of the photocatalytic system can still be further enhanced by  
959 using appropriate different flow modes, e.g., surface flow, flow-through, cross-flow, and dead-end or  
960 statistic mode (Fig. 14a). Gao et al. (2020) demonstrated that the steady increase of solution flow vertical  
961 to the membrane surface resulted in an overall increase in degradation efficiency of the photocatalytic  
962 system. The membrane degradation efficiency follows an order of FT > CF > SF.

963 **Fig. 14 about here**

964 Ivanković et al. (2021) used adsorption on graphene and functionalized deviations like GO NMs-  
965 based photocatalysis to determine the basic characteristics and behavior of the 18 most commonly found  
966 PhACs (beta-blockers, lipid regulators, psychotropic drugs, and analgesics) in aquatic environments (Ye  
967 et al., 2021b). Metal-organic frameworks (MOFs) were investigated extensively as diverse types of  
968 photocatalysts due to their outstanding ligand-metal charge transfer (LMCT) characteristics when  
969 exposed to light (Yang et al., 2017b; Gao et al., 2018). Due to their Fe<sub>3</sub>-3-oxo clusters and mild toxicity,  
970 Fe-based MOFs have a lot of promise as photocatalysts, as they can initiate a heterogeneous photo-  
971 Fenton-like response in sewage treatment when activated under light (Liu et al., 2018; Ahmad et al.,  
972 2019). In addition, the antifouling potential of the membrane is significantly increased by strengthening  
973 the membrane scheme with a photo-Fenton-like system (Xie et al., 2020b).

974

## 975 **9. Electrified membranes (EMs)**

976 Different structures of EMs membrane (e.g., flat sheet membrane, hollow-fiber, tubular and spiral  
977 membrane) can be formed via free-standing membrane casting, composite membrane assembly process

978 and membrane modification and used as operated in flow-by and flow-through filtration systems (Fig.  
979 15a). These membranes have been demonstrated to be effective for neutral and low molecular weight  
980 EOMPs, exceeding traditional membranes' size- and charge-exclusion separation characteristics (Pollitt  
981 et al., 2019). Electrochemical-based redox processes (Fig. 15b), electrooxidation, electro-adsorption, and  
982 electrostatic rejection (Tang et al., 2017; Zhu and Jassby, 2019) are just a few of the electro-assisted  
983 available technologies, and they overcome fouling and steric hindrance caused by microbes, organic  
984 pollutants, and inorganic salts as shown in Fig. 15c (Liu et al., 2020d). As a result, EMs can effectively  
985 dissolve and/or modify contaminants during membrane separation operations, enhancing charged species  
986 rejection.

987 **Fig. 15 about here**

988 Particular electrode geometry such as nanodots, nanowire, and nanorod arrangement can be used to  
989 facilitate the removal of microbes (e.g., virus) in anodic EM (Fig 15d). The process is further improved  
990 by (i) optimizing flow convection (Ji et al., 2015), (ii) amplifying the electric field (Zhao et al., 2018),  
991 (iii) increasing temperature, and (iv) concentrating reactants (Ji et al., 2019).

992 Yang et al. (2016) incorporated GO into a ceramic composite tubular membrane and utilized it to  
993 eliminate several EOMPs from water. EOMPs includes sulfamethoxazole (SMX), di(2-ethylhexyl)  
994 phthalate (DEHP), di-n-butyl phthalate (DnBP), and cephalexin (CLX). They observed that 99%  
995 rejection of DEHP and DnBP was achieved in crossflow filtration membrane supported by  
996 electrocoagulation and electro-filtration process (EC/EF), whereas those for cephalexin (CLX),  
997 sulfamethoxazole (SMX), and caffeine (CAF) varied from 32 to 97%. The predominant removal  
998 processes of multifunctional membrane system were (i) size exclusion, (ii) electrostatic repulsion, (iii)  
999 adsorption, (iv) electrocoagulation, and (v) electro-filtration (Fig. 15b).

1000 Sulfamethoxazole is an antibiotic that is difficult to remove from wastewater because it produces  
1001 refractory by-products (Du et al., 2018). The complete separation of SMX from sewage is impossible  
1002 with the conventional sludge activation method (Ji et al., 2019) because of its resistant and bio-refractory  
1003 nature. Due to their mild toxicity and high capacity, electrochemical advanced oxidation processes  
1004 (AOPs) are vital for removing these pharmaceuticals (Moreira et al., 2017). Yu et al. (2020) developed  
1005 an electrochemically improved carbon nanofibrous anodic membrane (G/SnO<sub>2</sub>/CFs) that may be used to  
1006 oxidize various developing and refractory medicines electrochemically (Fig. 16a).

1007 Due to SnO<sub>2</sub>'s strong electrooxidation capacity and graphene's excellent electrical conductivity  
1008 (Kretinin et al., 2014), the electrooxidation performance and conductivity of carbon nanofibers (CFs) can  
1009 be considerably improved by adding graphene/tin dioxide (G/SnO<sub>2</sub>) into them in a unique way. It was  
1010 discovered that large amounts of anodic membrane and changing the flow-through mode of the PAN-  
1011 based G/SnO<sub>2</sub>/CFs microfiltration improved its degrading efficiency and electrochemical filtration.  
1012 Furthermore, the composite membrane showed excellent degradation (85%) with promising stability  
1013 after 10 cycles in 50 h operation, indicating that it can manage dangerous refractory organics in  
1014 wastewater.

1015 **Fig. 16 about here**

1016 Electrospinning was utilized by Li et al. (Li et al., 2019a) to create a ZIF-67/polyacrylonitrile (PAN)  
1017 fibrous membrane that enhanced the simultaneous removal of impurities as a membrane reactor (Wang  
1018 et al., 2020). However, most catalysts were partially deposited or gathered disorganized in polymers,  
1019 which obstructs electron movement and necessitates the development of more effective supporting  
1020 materials in catalytic membrane systems (Zhong et al., 2018). Although GO's mechanical strength and  
1021 ultra-flexibility make it a strong choice for membrane manufacturing, pollutant buildup on the membrane  
1022 surface lowers penetration flux (Sun and Li, 2018). Tunable mass transfer channels can separate oil and  
1023 water while also auto-cleaning photo catalytically by linkers active units into adjoining GO (Cai et al.,  
1024 2020). However, in PMS stimulation, the utilization of GO stacked frameworks in combination with  
1025 active catalysts to generate an auto-standing GO-assisted membrane for synchronized screening and  
1026 catalytic oxidation of EMOPs is unusual.

1027 Recently, an auto-assembly approach was developed for sandwiching active species, lawn-like 2D  
1028 Fe<sub>2</sub>O<sub>3</sub>@Co<sub>0.08</sub>-Fe<sub>1.92</sub>@nitrogen-doped CNT/reduced GO (rGO) composites, into GO membranes (Ye  
1029 et al., 2021c). Because of the enhanced interlayer distance of GO nanosheets, the FeCo@GCTs/GO film  
1030 improved permeability and displayed catalytic auto-cleaning features. The permeation flux of the  
1031 FeCo@GCTs/GO membrane was enhanced to 487.3 L m<sup>-2</sup> h<sup>-1</sup>bar<sup>-1</sup>, which was more than 7.5 times that  
1032 of the GO membrane (64.6 L m<sup>-2</sup> h<sup>-1</sup>bar<sup>-1</sup>), and the antifouling performance of the composite membrane  
1033 was improved as the interlayer gap of GO nanosheets was increased.

1034 Meanwhile, in the SMX-HA combined setup, their removal efficiencies were 67.5% (by GO  
1035 membrane), and 99.2% (by a composite membrane) were achieved, suggesting that the FeCo@GCTs/GO  
1036 membrane can also segregate micro molecules. After 30 minutes, the FeCo@GCTs/GO/PMS system

1037 successfully removed up to 98.4% of SMX. Moreover, following five successive cycles, the  
1038 FeCo@GCTs/GO membrane sustained above 90% SMX elimination and Cr (VI) removal under 30 min,  
1039 showing that the FeCo@GCTs/GO membrane was adaptive to repeated use (Fig. 16b). The results  
1040 revealed that the catalytic membrane/PMS setup could efficiently prevent metal loss.

1041 As per the participation of soluble metal ions, the cleaning rates of  $\text{Co}^{2+}$  and  $\text{Fe}^{2+}$  were moderate (<  
1042 7%), showing a diverse mechanism for SMX breakdown (Fig. 16b). Additionally, once the catalytic  
1043 membrane was deployed to a range of contaminants mixture, about 100% degradation rates were  
1044 achieved for MO, BPA and TC in 15 min, 25 min, and 30 min (Ye et al., 2021c). This self-contained  
1045 FeCo@GCTs/GO catalytic membrane can be utilized in numerous microchannels membrane substrates  
1046 in challenging circumstances.

## 1047 **10. Conclusions and outlooks**

1048 OMPs have been found in tap water, wastewater, and surface water. Uncontrolled OMPs threaten already  
1049 constrained water supplies. The conventional strategies used in most WWTP cannot remove OMPs like  
1050 PPCPs. Therefore, developing efficient and environment-friendly novel supporting technologies is  
1051 crucial to ensuring water quality and future-proofing potable recovered water use. Membrane filtration  
1052 has been pragmatic to water treatment contaminated with numerous OMPs. However, owing to cost,  
1053 fouling control, poor dispersion in aqueous solution, and low sorption affinity, its application on a broad  
1054 scale remains a challenge.

1055 Furthermore, each membrane technology has its own characteristics that must be considered while  
1056 deciding on the best treatment. Likewise, the combination of two or more strategies also improved overall  
1057 performance. For instance, degradation combined with filtration is 2.15 times more effective. GO  
1058 additive strengthens majorities of available porous polymeric substrates membranes (e.g., PVDF, PC,  
1059 PAN, and PSF), but the incorporation of GO into inorganic membranes is still limited.

1060 Adsorption is another membrane-based filtration strengthening factor that was improved by  
1061 incorporating GO nanosheets which provide a hydrophilic layer and increase surface area and adsorption  
1062 capacity. However, adsorption may accelerate biological fouling and limits water flux. Herein,  
1063 bactericidal characteristics of GO may promise improved flux recovery (~6 folds) while mixing of other  
1064 chemical species (e.g., Ag, APTES,  $\text{SiO}_2$ , ZnO) and incorporation degradation can further enhance (7.5  
1065 folds) and make membrane useable for many cycles.

1066            However, chemical and mechanical stability is the most demanding feature of membrane life. For  
1067 this, hollow metal fibers such as stainless steel (SS) membranes are gaining significant attention. Still,  
1068 surface passivation is required to prevent oxidation and advanced decoration routes. A similar issue can  
1069 also be addressed using nanoscale coatings of GO to accelerate electrochemical processes and avoid the  
1070 early corrosion of bare metal reinforcements during electro-Fenton (EF) reactions.

1071            More studies are warranted to assess the enhancement of GO-assisted photo-Fenton catalytic and  
1072 Fe-based MOFs catalytic properties. AOPs frequently promote the release of a number of different  
1073 oxidants, increasing the electrolysis effectiveness of other anode materials. Resultantly make it less  
1074 effective in less polluted medium with a lower level of pollutants. This can be explained by establishing  
1075 a limiting phase in the reaction caused by the mass transfer of target contaminants to the anode surface  
1076 from the liquid. It has been shown that sono-electrolysis increases the mass transfer rate and encourages  
1077 the destruction of hydroxyl radicals, which are responsible for the production of water, as well as several  
1078 oxidants produced when these radicals interact with other species found in wastewater (Oturán et al.,  
1079 2008; Sánchez-Carretero et al., 2011). Likewise, bio-based polymers (used in matrix membranes to  
1080 improve hydrophilic and permeability) and carboxymethyl cellulose GO may boost tensile strength and  
1081 Young's modulus by 2 to 3 times. Therefore, GO-based improved mechanical stability in polymeric  
1082 membranes could appear as a better alternative to ceramic membranes and reduce the operational cost  
1083 by up to 24-54% (Sagle and Freeman, 2004).

1084            Moreover, the microorganisms and organic matter in wastewater are also limiting factors in the  
1085 membrane filtration of EOMPs. Improving the operating matrices (washing, conditioning cycles and  
1086 hydrodynamic optimization) may minimize this issue, and operations and functionality might endure  
1087 longer.

1088            Achieving a balance between EOMPs, environmental implications, and economic feasibility is  
1089 essential for constructing an economical and environmentally benign solution. GO mediated gravity-  
1090 driven membranes for low pressure. Likewise, incorporating GO-ZnO decreases membrane-solute  
1091 hydrophobic attraction, hinders fouling by sodium alginate, increases rejection rate, and provides high  
1092 fluxes. The membranes usability at minimal pressure (4 bar) and optimum photocatalytic stand out as  
1093 ideal technology for low-energy applications in rural parts of developing countries where untreated water  
1094 is collected and used for various domestic purposes.

1095 GO integration substantially not only facilitate EOPMs removal but also shows an excellent flux  
1096 recovery ratio in a diverse type of system, which is imperative for long-term performance and reusability.  
1097 For instance and in the MBR system, 96% atrazine was removed by UF (FRR:98.6%), in photocatalytic  
1098 degradation, 92% removal of methylene Blue was achieved (FRR:96%), in PVDF membrane, reduced  
1099 extracellular polymeric substance and removed 97% COD (FRR:98.6%), and along poly (acrylonitrile-  
1100 methyl acrylate) 99.3% of BSA removed (FRR:98.6%)

1101 GO incorporation has also demonstrated high visible light attributed to TiO<sub>2</sub> activation, saving  
1102 energy. The modified polyvinylidene fluoride GO membrane with Ag<sub>2</sub>CO<sub>3</sub> disposition exhibited minimal  
1103 irreversible fouling ratio (*R<sub>ir</sub>*) and total fouling ratio (*R<sub>t</sub>*) readings, indicating excellent antifouling  
1104 membrane characteristics. This response was attributed to the formation of a hydration layer on the  
1105 surface of the membrane as a function of mass deposited CO<sub>3</sub><sup>2-</sup> (hydrophobic group), which protects the  
1106 membrane surface and inhibits the attachment of pollutants (Qin et al., 2015). As with other  
1107 photocatalytic materials like TiO<sub>2</sub>, Ag<sub>2</sub>CO<sub>3</sub> photocatalytic activity under UV or visible irradiations may  
1108 also be employed to improve the self-cleaning ability of modified membranes.

1109 GO integrated membrane performance can be further optimized by other strategies such as  
1110 electrocoagulation and electro filtration (phthalate esters, ECs, and PhACs). This approach offers the  
1111 door for the scalable design of improved combinatorial electro-reactive materials for the oxidation  
1112 treatment of highly polluted effluents and simultaneous filtration. Instead of screening across the pores,  
1113 this method permits selectivity across vast pore-sized membranes.

1114 These strategies in conjugation with mixed matrix membrane have enabled the fabrication of GO  
1115 ultrafiltration membrane, which attains better permeability and excellent selective screening and shows  
1116 improved antifouling potential and promising mechanical traits. Due to pseudocapacitance properties,  
1117 MMMs can work as in-situ monitors for membrane fouling which offers long working life, and  
1118 membranes capable of producing high-efficiency filtration in environmental application separation  
1119 processes. However, removal rates are often highly variable for target EOMPs (i.e., CAF, SMX, and  
1120 CLX), particularly in the presence of multiple pollutants, making complexities in applying these  
1121 membranes. To overcome this shortcoming, further study is needed to figure out how to enhance the  
1122 removal performance and keep it consistent for various ECs.

1123 More research is needed to ensure the efficient technical transfer of PES-GO membranes in MBR.  
1124 This additional research could concentrate on (i) exploring the economic level of GO to increase

1125 permeability without jeopardizing membrane antifouling potential; (ii) coupling joint physical and  
1126 mechanical strategies to remove/reduce the loose cake layer formed on the PES-GO membrane during  
1127 mixed liquor filtration; and (iii) investigating alternative material for chemical cleaning to cope these  
1128 challenges.

1129 Finally, although it hasn't been proven that low-level exposure to these compounds is harmful,  
1130 potable water treatment plants should aim to give pharmaceutical-free water. Water rules ensure water's  
1131 credibility and safety. More research into the prevalence of these compounds in the biosphere and their  
1132 impact on quality of life could lead to future legislation.

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## Figures Captions

**Fig. 1** Schematics illustration for different synthesis of graphene oxide (GO) (a-c) Hummers method, pyrolysis of carboxyl compounds method and, pyrolysis of carbon-based waste (Adapted from Ref.(Lebepe et al., 2020) with permission under creative commons CC-BY license, 2020), and (d) GO with functional groups after exfoliate from graphite and acid catalysis.

**Fig. 2** Diagram illustrating the various synthesis procedures utilized to manufacture GO membranes. (a) Strategy of vacuum filtration (Baskoro et al., 2018) (b) Pressure-, vacuum-, and evaporation-assisted self-assembly method Adapted from Ref. (Kwon et al., 2021) with permission under creative commons CC-BY license, 2021, (c) spin-coating approach (Nair et al., 2012), (d) LBL assembly method (Zhang et al., 2015) (e) Spray-evaporation assembly methods (Guan et al., 2017) (f) Immersion coating method (Rollings et al., 2016). Reproduced with Elsevier and Royal Society of Chemistry's permission.

**Fig. 3** SEM micrographs of surfaces of: tubular ceramic substrate (a), GO membrane (b), SG@GO membrane (c), cross-section of: tubular ceramic substrate (d), GO membrane (e), SG@GO membrane (f), graphical depiction of WCA and SZP of SG@GO laminate with unlike SG levels (Synthesis settings: GO: 0.2 g L<sup>-1</sup>; immersing time: 0.5 h) (g); Graphic illustration of manufacture of SG@GO membrane on tubular ceramic substrate (h), and, EOMPs retaining of GO membrane and SG@GO composite membrane (i). Reprinted with permission from ref (Shen et al., 2017). Elsevier holds the copyright for 2017.

**Fig. 4** SEM micrographs of surface and cross-section morphologies of: Ceramic tube (a), PEI membrane (b), GO/PEI membrane (c), ZIF-8/PEI membrane (d), ZIF-8@GO/PEI composite membrane (ZIF-8@GO:0.005 wt.%) (e), AFM images of GO/PEI membrane, ZIF-8/PEI membrane and ZIF-8@GO/PEI membrane (f), and graphical presentation of vacuum-assisted assembly method based synthesis of crosslinked ZIF-8@GO/PEI composite membrane on tubular ceramic substrate and EOMPs molecules retention (g). Reproduced with permission from ref. (Yang et al., 2018) Copyright 2018, Elsevier.

**Fig. 5** Graphical illustration of the two-phase method applied to prepare the GO-functionalized zwitterionic polyampholyte hydrogel PES membrane. WP (b), PEG rejection of the pristine PES, p-PES, and GO-p-PES membranes (values expressed: means  $\pm$  SD, n = 3) Impact of 3 h surface connection of PES, p-PES, and GO-p-PES membranes to *E. coli* ( $\sim 4 \times 10^4$  CFU/1.54 cm<sup>2</sup>), the values are stated as means  $\pm$  SD, n = 8) (c), Biovolume of the live cells, EPS, and dead cells for the three membranes the values are stated as means  $\pm$  SD, n = 12 arbitrary sites in two repetition experiments (d), time-sensitive reutilizing fluxes of the pristine PES, p-PES, and GO-p-PES membranes in the BSA ultrafiltration test (BSA: 1 g L<sup>-1</sup> in 10 mM Phosphate-buffered saline solution, initial flux: 50 L m<sup>-2</sup> h<sup>-1</sup>, transmembrane pressure :0.2–0.7 bar) (e) Reproduced with permission from ref. (Zhang et al., 2018) Copyright 2018, Elsevier.

**Fig. 6** (a) Practical GO membrane formulation for EOMPs/salts co-contaminated refractory wastewater treatment, (b) Removal of typical EOMPs. Reproduced with permission from ref. (Han et al., 2018) Copyright 2018, Elsevier, (c) Graphical depiction of PVP-GO nanocomposites and PVP-GO/PVDF hollow fiber membrane synthesis, (d) methodology of MPBR system and (e) flux and EPS study of proteins and polysaccharides using pristine PVDF and PVP-GO/PVDF membrane in the MPBR arrangements. Reproduced with permission from ref. (Wu et al., 2020) Copyright 2020, Elsevier.

**Fig. 7 a** Time sensitive infiltration of GS-DIMs and NIMs for TC (a), the expected permselectivity action for template molecule by GS-DIMs (values expressed: means  $\pm$  SD, n = 3) (b), Schematic illustration for the preparation method of GS-DIMs. Real-time filtration fluid curves for GS-DIMs and the corresponding level of different target molecules through the GS-DIMs (values expressed: means  $\pm$  SD, n = 3). Adopted with permission from ref. (Wu et al., 2021b) Copyright 2021, Elsevier, **b** Time sensitive infiltration of GT-DIMs and NIMs for TC (a), the expected permselectivity action for template molecule by GT-DIMs (values expressed: means  $\pm$  SD, n = 3) (b), Schematic illustration for the preparation method of GT-DIMs. Real-time filtration fluid curves for GT-DIMs and the corresponding level of TC, OTC, CTX and SMZ through the GT-DIMs (values expressed: means  $\pm$  SD, n = 3) reproduced with permission (Wu et al., 2022b) Copyright 2022, Elsevier.

**Fig. 8 a** Graphical depiction of GO/Co-Al LDH composite membranes synthesis (a), performance of different membranes based on rejection efficiency and flux for 10 ppm MB solution (b), performance of GO/Co-Al-2 based on rejection efficiency and flux for MB solution at different level (10–100 ppm) (c), performance of different membranes based on flux for different EOMPs at 10 ppm level (d), performance of different composite membranes based on rejection efficiency for different EOMPs at 10 ppm (e) reproduced with permission (Wang et al., 2021b) Copyright 2021, Elsevier, and comparison of CS\_GLU and CS\_GOX\_GLU composite membranes for recovery potential of the pesticides (in ascending order of log Kow) (f) reproduced with permission (Silvestro et al., 2021) Copyright 2021.

**Fig. 9** Salient features of membrane separation technology. (a) Selective separation of MF, UF, RO, and NF membranes reproduced with permission (Ren et al., 2021), Copyright 2021 Elsevier, (b) Mechanisms involve in

membrane filtration process (b) Reprinted with permission from Khanzada et al. (Khanzada et al., 2020). Copyright 2020 Elsevier.

**Fig.10 a** Graphene design arrangement (a) (E2 molecules denoted estradiol adsorption sites), Routes of steroid hormone absorption by GO dependent on closeness to the hormone (GO is represented symbolically as a GO portion as an marker) (b) reproduced with permission (Nguyen et al., 2021) Copyright 2021 Elsevier; **b** Illustration of the targeted surface change of the GO NF membrane induced by  $\text{Fe}_3^+$  (a); Representation of a potential reaction between the mix of APTES and TA and associated contacts with the  $\text{Fe}_3^+$ - facilitated GO membrane (b), BPA rejection (c) and WP of GO, GO-Zn and GO-Fe membranes coated and noncoated by TA-APTES layer (d) reprinted with permission (Li et al., 2021b) Copyright 2021 Elsevier.; **c** SEM micrographs BDMs: (a) CNT + Lac@UF(a-f): (b) Lac&CNT@UF, (c) CNT&Lac@UF, (d) GO&Lac@UF, (e) Lac&GO@UF and (f) GO&Lac@UF, BDM assembly methods (g), Elimination rate, flux, and laccase activities for diverse BDMs manufactured by various techniques. (h) Elimination rate, flux, and laccase activities at different filtration cycles for CNT/GO&Lac@UF. The experimental conditions were CNT/GO:25 g m<sup>-2</sup>, laccase dosage:50 g m<sup>-2</sup>, pollutant concentration:25 g L<sup>-1</sup>, pressure: 0.1 Mpa, 100 rpm (i) reprinted with permission (Zhu et al., 2020a) Copyright 2020 Elsevier.

**Fig.11** graphical illustration of TAML/GO-PEI/PES working performance in filtration and removal of BPA (a), WF and BPA content in permeate solution in filtration cycles trial of TAML/GO-PEI/PES membrane (b); TAML/GO-PEI/PES membrane adsorption capacity in filtration cycles trial (c); PES, GO-PEI/PES and TAML/GO-PEI/PES membranes performance in EOMPs removal (1 mg L<sup>-1</sup>) (d), and TAML/GO-PEI/PES membrane filtration attributed degradation pathway of BPA (e) Reproduced with permission from ref. (Zhang et al., 2021) Copyright 2021, Elsevier.

**Fig.12 a** The manufacture method of ZIF@MILs-20/GO catalytic membrane (a), Conceptual depiction of the PMS activation process on the ZIF@MILs-20/GO membrane (b), NX degradation mechanisms in the PMS system, GO membrane, and ZIF@MILs-20 (c), comparative analyses with other catalysts (d), The effects of catalysts (e), WCA (f), catalysts performance at varying system pH (g), catalysts performance at varying reaction temperature (h), the matching activation energy (d), conditions: [catalyst] = 0.01 g L<sup>-1</sup>, [NX] = 20 mg L<sup>-1</sup>, [PMS] = 0.3 g L<sup>-1</sup>, [T] = 25 °C, the catalytic membrane's capacity for cleanup following adsorption (i) reprinted with permission from (Ye et al., 2021a) Copyright 2021 Elsevier, **b** Schematic drawing of the assembly of GO/M88A membrane (a). Schematic presentation of photo-Fenton self-cleaning system in GO/M88A membrane-H<sub>2</sub>O<sub>2</sub>-visible light arrangement (b), the effort of separating BPA from GO/M88A membrane without and using photo-Fenton method (c), curves of BPA degradation by photo-Fenton catalytic (d), digitized images of the color shift of real textile effluent during photo-Fenton/removal operations (e), the elimination of pollutants from true textile effluents using GO/M88A membranes (f) reprinted with permission (Xie et al., 2020) Copyright 2020 Elsevier.,

**Fig. 13 (a)** Graphical depiction of the SMSMPR, (b) Mineralization mechanism of AMX in water by SMSMPR, (c) Elimination proficiency of AMX in contrasting reaction schemes, (d) diverse light sources' impacts, (e) H<sub>2</sub>O<sub>2</sub> dosages and (f) different solution pH on AMX degradation in the TiO<sub>2</sub>-GO-18 wt.% Fe<sub>3</sub>O<sub>4</sub> & H<sub>2</sub>O<sub>2</sub> arrangement by the SMSMPR. Reaction settings: [H<sub>2</sub>O<sub>2</sub>] = 20 mM (for c, d, f), pH 3 (for c, d, e), [catalysts] = 0.5 g L<sup>-1</sup> (for c-f), [AMX] = 20 mg L<sup>-1</sup> (for c-f), and visible light irradiation (for c, e, f) adopted from ref. (Li et al., 2019) with permission, Copyright 2019 Elsevier.

**Fig. 14 (a)** Diagram illustrating the four functional modes of the photocatalytic filtering reactor, adopted from ref. (Gao et al., 2020) with permission, Copyright 2020 Elsevier (b) Diagram illustrating the suggested methods for eliminating target pollutants via a ceramic composite tubular membrane containing Go combined with the EC/EF process, adopted from ref. (Yang et al., 2016) with permission, Copyright 2016 Elsevier.

**Fig. 15 (a)** Synthesis of electrified membrane (EM), electrified circuits, and installation Illustration of EM preparation, electrified membrane modules, and cross-flow filtration function, as well as the influence of water convection and diffusion on operating modes, (b) EM filtration for water purification and filtration; (c) various forms of membrane fouling; and (d) improving microbial elimination in anodic EM filtration with improved membrane electrode design. Reprinted with permission from Ref. (Sun et al., 2021). Copyright 2021 American Chemical Society.

**Fig. 16 a** Left: the matching images of electrochemical reactor; middle: complete arrangement of electrochemical membrane device comprises the membrane stacks; right: photos of the unfolded membrane sandwich stacks with three layers from vertically (anode: PAN-based G/SnO<sub>2</sub>/CFs membrane maintained by RuO<sub>2</sub> plated titanic (Ti/RuO<sub>2</sub>) net, filter: an insulating silicone rubber O-ring separated the electrodes, cathode: RuO<sub>2</sub> plated titanic (Ti/RuO<sub>2</sub>) net) (a), Projected conversion pathways for SMX electrocatalytic degradation (b) adopted from ref. (Yu et al., 2020) with permission, Copyright 2020 Elsevier. And **b** FeCo@GCTs/GO membrane reusability in SMX and Cr(VI) solution (a); and the breakdown productivities of metal ions (b) adopted from ref. (Ye et al., 2021b) with permission, Copyright 2021 Elsevier.

**Table 1 GO mediated membranes in filtration and adsorption**

Compound name	Target pollutant	GO level;	Pollutants removal efficiency and mechanism	Flux/hydrophilicity/permeability	Major findings and recommendations	Reference
Polyvinylidene fluoride graphene oxide silver carbonate (PGA membranes)	BSA	0.4 wt.%	79%; UF	PWF: PGA-3 (254 LMH) > 2-fold than pristine PVDF membrane (126 LMH); FDR: 83.1	The PG membrane coated three times with Ag <sub>2</sub> CO <sub>3</sub> (i.e., PGA-3) showed higher FRR, FDR and antibacterial performance than other membranes and provide best quality of lake water in multi-cycle filtration experiments.	(Wu et al., 2021a)
ZIF-8@GO/PEI	Organic solvents	GO: 0.1-0.4 g L <sup>-1</sup>	99.1%; NF	3.5 L m <sup>-2</sup> h <sup>-1</sup> bar <sup>-1</sup>	Owing to its exceptional compaction, tubular membrane modules are useful in industry.	(Yang et al., 2018)
PVP-GO/PVDF membrane	extracellular polymeric substance (EPS) COD, NH <sub>4</sub> <sup>-</sup> N <sup>+</sup> and NO <sub>3</sub> <sup>-</sup> N	0.5 wt.%	> 97.8, 93.1 and 68.7% removal COD, NO <sub>3</sub> <sup>-</sup> N, NH <sub>4</sub> <sup>-</sup> and N <sup>+</sup>	MP: 1.7 times > PM; WCA: decreased from 97 to 62°, FRR: 96%	PVP-GO function as wipers to remove EPS foulants from membrane surface. PVP-GO/PVDF shown excellent antifouling properties for high-density microbe establishment and ammonia-nitrogen wastewater remediation.	(Wu et al., 2020)
GO additive solvent green (SG)	dye molecules Eriochrome black T	0.2 g L <sup>-1</sup>	98% similar to PM; NF	330 L m <sup>-2</sup> h <sup>-1</sup> bar <sup>-1</sup> nearly 6- fold more than PM GO (56 L m <sup>-2</sup> h <sup>-1</sup> MPa <sup>-1</sup> )	This approach may provide a straightforward technique for optimizing the interlayer spacing of GO laminates for efficient molecular sorting.	(Shen et al., 2017)
GO/Co-Al LDH	RhB, MB, CR, petroleum ether, n-hexane	Al LDH: GO= 4:1	Rejection>90%; VF	718.8 L m <sup>-2</sup> h <sup>-1</sup> bar <sup>-1</sup>	Co-Al LDH may be altered in GO membranes to improve extraction efficiency and wettability.	(Wang et al., 2021b)
HGO	three typical organics in NOM (BSA, SA and HA)	2 mg mL <sup>-1</sup> GO 0.08 g m <sup>-2</sup>	NOM 55%; BSA 29%; SA:58–85%; and HA: 72–92%; UF	WP of HGO >26 time than rGO and >2 times to GO; FFR: 96.04%; WCA: reduced from 71° to 35°	Increased HGO dosage may delay the transition from pore blockage to cake layer. Ultimately, HGO coating provides a possible use for the treatment of foulants.	(Ding et al., 2021)
GO/aminated GO (NGO)	petrochemical	GO: 0.05 wt.%	6% more rejection than PM; NF	PWF of PM was > 24.8% increased by 15.6% and 26.9% i.e., 44.93 and 49.30 L m <sup>-2</sup> h <sup>-1</sup> with GO and NGO, respectively	The interaction between the amino group of NGOs and TMC lowered the thickness size of TFCNGO. Thin film composite NGO offers a substantially finer layer over TFCGO owing to its larger pore dimensions and increased wettability.	(Kong et al., 2021)
Ceramic-based graphene oxide framework (GOF) membranes	Chloramphenicol (CAP)	0.2 mg mL <sup>-1</sup>	53.3%; NF	increase in permeability	Cross-linking GOF CM with MPD (m-phenylenediamine) lead in the synchronous rejection of salts and CAP, so overcoming the normal trade-off dilemma.	(Yuan et al., 2020a)

					Involved in the Donnan effect, the molecule/ion screening mechanism were size sieving (steric hindrance), electrostatic interaction, and fractional dehydration.	
GO membrane	Organic matter (COD)	GO:14.4 mg m <sup>-2</sup>	76%; RO/NF	Flux decrease 7.8 to 3.1 L m <sup>-2</sup> h <sup>-1</sup> bar <sup>-1</sup> at 250 layers of GO	Rejection and flux were optimum up to 100 GO layers, achieve balance for the support membrane, optimum up to 100 Go layers. Negatively charged OM better removed with GO than the electrically neutral polyethylene glycol (PEG).	(Han et al., 2018)
Tannic acid-crosslinked graphene oxide membranes	COD, TDS, TSS and turbidity	0.5 mg mL <sup>-1</sup>	COD:91.67%; VF		The phenolic hydroxyl segments of tannic acid are plentiful throughout the GO nanosheets, rendering the membrane super oleophobic and ultra-hydrophilic. Low oil adhesion, maximum oil rejection, and high-pressure water filtration.	(Singhal et al., 2020)
Ceramic membrane functionalized with GO (ceramic GO)	NOM (humic acid and tannic acid), pharmaceuticals (ibuprofen and sulfamethoxazole)	1 g	GO improved rejection: 74.6 to 93.5% (NOM); 15.3 to 51.0% (pharmaceuticals); 2.9 to 31.4% (inorganic salts)	MP: 14.4–58.6 vs. 25.1–62.7 L m <sup>-2</sup> h <sup>-1</sup> bar <sup>-1</sup>	Hydrophobic attraction and charge effects by GO removed EOMPs. GO mediated ceramic membrane is effective in removal of multiple pollutant with contrasting nature from water.	(Chu et al., 2017)
GO-TA-APTES-x tannic acid (TA)-aminopropyltriethoxysilane (APTES) nanoparticles	Congo red	0.25 mg mL <sup>-1</sup>	99.9%; NF	TA-APTES increase 10 times MP; PWF was up to 111. L m <sup>-2</sup> h <sup>-1</sup> bar <sup>-1</sup> , (15 times more to GO)	The crosslinking agent TA-APTES NPs encourage and facilitate resilience in aqueous conditions and is conducive to the transfer of GO nanofiltration membrane from controlled system to industrial use.	(Liu et al., 2022)
fluffy-like poly (acrylonitrile-methyl acrylate) amphiphilic graphene oxide (f-GO/P(AN-MA))	BSA	0.5 wt.%	Increased 70.59% (PM) to 99.33% (MM); UF	PWF 200 to 326 L m <sup>-2</sup> h <sup>-1</sup> , while the WCA declined from 92.8° to 68.3°.	FRR: 98.16%. poly (acrylonitrile-methyl acrylate) entangled hydrophobic segment on f-GO f-GO/P(AN-MA) showed long-term resistance to organic matter fouling	(Zhang et al., 2022a)
GO-PEG/PVDF graphene oxide-polyethylene glycol (UF)	BSA	0.5 wt.%	94%; UF	Flux: 93.0 L m <sup>-2</sup> h <sup>-1</sup> twice than PM	FRR: 78.0%; MM exhibits superior membrane wettability, porosity, and antifouling ability in commercial systems.	(Ma et al., 2020)
Perfluorosulfonic acid (PFSA) grafted on graphene oxide	BSA	PFSA: GO mass	BSA:93.9%; HA:79.6%; UF	587.4 L m <sup>-2</sup> h <sup>-1</sup> 1.5 time higher than PM	FRR: 90.8% Membrane alteration increased hydrophilic nature, limited surface roughness, and	(Liu et al., 2020)

(GO) PFSA-g-GO/PVDF		ratio of 2.5:1			augmented porosity and pore diameter. The mass ratio of PFSA to GO in membrane nanocomposites is the key determinant of pore properties and porosity.	
GO@UiO66/PVDF	BSA	1.0 wt.%	93%; UF	263.3 L m <sup>-2</sup> h <sup>-1</sup> WCA: reduced from 78.49 (PVDF) to 63.61 and 58.31 with GO /PVDF and GO@UiO66/PVDF	FRR 81.3% Brilliant structural properties and chemical resistance; Extra hydrophilic active groups of the GO@UiO-66 visibility and PVDF phase transformation from contributed to the formation of a compact hydration sheet on the membrane surface and lowered Zeta potential, thereby enhancing organic invasion and reducing membrane fouling.	(Liang et al., 2021)
PES-HPEI-GO hyperbranched polyethylenimine	BSA	5.0 wt.%	92.0%; UF	153.5 L m <sup>-2</sup> h <sup>-1</sup>	FRR 88.7%; WCA 63. Antibiotic efficacy against <i>Escherichia coli</i> ( <i>E. coli</i> ) is excellent. The tensile strength and Young's modulus of hybrid membranes were greater.	(Yu et al., 2013)
PVDF/GO	BSA	1.0 wt.%	92.6%; UF	398.7 L m <sup>-2</sup> h <sup>-1</sup>	FRR 79.3%; WCA 62.5	(Xu et al., 2016)
PSF-GO	BSA	1.3 wt.%	Rejection was similar to PM-UF	430.0 L m <sup>-2</sup> h <sup>-1</sup>	FRR 82.5%; GO nanoplatelets can change the pore dimensions of porous media produced by inversion by up to 20%. Hydrophilicity and electrostatic repulsion boosted anti biofouling, while GO treatment showed higher mechanical strength and water permeability.	(Lee et al., 2013)
PSF-SiO <sub>2</sub> /GO	BSA	0.3 wt.%	98.0; UF	185.0 L m <sup>-2</sup> h <sup>-1</sup>	FRR 84.4%; WCA 72.0. A spongy and uniform film of silica nanoparticles applied to the surface of the GO as a GO space blanket results in high wettability and dispersibility, as well as an extended in water permeation rate, protein elimination, and antifouling ability.	(Wu et al., 2014)
PVDF/GO	BSA	0.5 wt.%	~ 97%; UF	~ 80 L m <sup>-2</sup> h <sup>-1</sup>	FRR ~ 75%; WCA ~ 76. The formation of hydrogen linkages between GO and PVP allows for improved efficiency.	(Chang et al., 2014)
PVDF/GO/PVP	BSA	0.5 wt.%	85; UF	104.3 L m <sup>-2</sup> h <sup>-1</sup>	FRR 90.5%; WCA ~ 67. The synergistic impacts of integrated GO and PVP improved surface hydrophilicity and antifouling performance.	(Chang et al., 2014)

PVDF/GO/PVP	BSA	0.3 wt.%	~ 75; UF	~ 800 L m <sup>-2</sup> h <sup>-1</sup>	FRR ~ 70%; WCA 65.93 The adhesion forces for BSA was 0.78 mN m <sup>-1</sup>	(Zhu et al., 2017b)
PVDF/SGO sulfonated graphene oxide (SGO)	BSA	0.12 wt.%	98%; UF	740 L m <sup>-2</sup> h <sup>-1</sup> Improvement in PWF 146.6% in P-SGO was for better than 53.3% P-GO blend membranes	FRR 88.7%; WCA 50. SGO has strong hydrogen interaction and electrostatic repulsion towards proteins foulants, together with increased hydrophilicity, water flux, and mechanical performance, pave the way for PVDF ultrafiltration membranes.	(Ayyaru and Ahn, 2017)
graphene oxide immobilized membrane	-	-	-	Improved water vapor flux (30–40%).	Biocidal impacts revealed that fewer energy (20-25 percent) was consumed by thermophilic and mesophilic cells i.e., 90.1% and 59.4%. Both the thermal and nonthermal impacts of microwave irradiation enhanced biocidal efficacy and flux. The ideal temperatures for eliminating thermophilic and mesophilic cells, accordingly, are 80 and 60 °C.	(Gupta et al., 2022)
GO-multi-walled carbon nanotubes (MWCNTs)	mineral and ionic species	0.1 wt.%	108.37% improved; VF	GO and MWCNTs decreased 32% contact angles	GO/MWCNT increases PVDF membrane surface characteristics and water purification functionalities.	(Chae et al., 2021)
GO modified polypropylene fiber filter membrane	COD and SS	-	COD: 84.8%; SS: 84.6%	-	Water-based paint wastewater flocculation + GO-PP fiber filter membrane could be a suitable material.	(Zhang et al., 2022b)
Europium-metal organic framework (Eu-MOF)/GO CM	organic phosphine	0.079 mg/cm <sup>2</sup>	100% rejection; GO based rejection:85.46%; VF	PWF: 1.27 to 16.76 L m <sup>-2</sup> h <sup>-1</sup> ; PF increased from 0.76 to 5.01 L m <sup>-2</sup> h <sup>-1</sup>	The permeability-selectivity computation of molecularly imprinted membrane materials could be addressed if the template molecule has a greater standard of tolerance and penetration.	(Li et al., 2021a)
Diphenyl phosphoryl azido (DPPA) / PSf (DPPA-GO/PSf)	BSA	0.5 wt.%	95.8%; UF	254.1 L h <sup>-1</sup> m <sup>-2</sup>	FRR 80.3%. Persistence anti- <i>E. coli</i> and <i>S. aureus</i> action. Strong interactions between oxygen-containing AGO nanosheets and polymer matrix eliminate interfacial flaws, resulting in good dispersity and interfacial compliance for PSF membranes.	(Xu et al., 2022)
PES-GO	HA, BSA	0.6 wt.%	>98%, MBR	GO incorporation reduce contact angle reduced from 59.6 to 46.9 and permeability from 288 to 161 L m <sup>-2</sup> h <sup>-1</sup> bar <sup>-1</sup>	PES-GO membranes are effective in removing organic pollutants, but are susceptible to damage from prolonged contact with sodium hypochlorite solution and chlorine.	(Lemos et al., 2021)

GO/PA-NF Polyamide (PA)	Congo red and cationic red X- GTL	0.05 wt.%	Congo red: 100%; cationic red X- GTL: 99.8%; NF	PF:44.2 L m <sup>-2</sup> h <sup>-1</sup> , PWF: 15.5 L m <sup>-2</sup> h <sup>-1</sup> bar <sup>-1</sup>	Piperazine (PIP) and trimesoyl chloride (TMC) significantly enhanced the PA NF membrane's wettability and antifouling abilities.	(Wei et al., 2021)
CGO-M/PES cysteine- functionalized GO (CGO)	BSA	0.5 wt.%	99.8%; UF	82.6 L m <sup>-2</sup> h <sup>-1</sup> at 0.2 MPa	FRR 92.1% (CGO/PES) > 47.5% (PM) WCA 50.5° at 2.0 wt.% CGO Antifouling polymer-based UF membranes treat protein-rich wastewater.	(Kong et al., 2020)
CA/MOF@GO Cellulose acetate Metal-organic frameworks	BSA	0.03, 0.06, 0.09 and 0.12 g	91.4; UF	183 L m <sup>-2</sup> h <sup>-1</sup> at 0.15 MPa	FFR 88.13%; WCA 49.5° HKUST-1 was used as MOF effectively overcome the drawbacks of GO and the properties of GO can be fully utilized.	(Yang et al., 2019)
PVDF/ OMWCNT/GO mixed matrix membranes oxidized multi- walled carbon nanotubes	BSA	0.5 g L <sup>-1</sup>	85%; UF	125.6 L m <sup>-2</sup> h <sup>-1</sup> at 0.1 MPa	WCA and FFR of MM were 67.6° and 60.57% while PM 79.6° and 51.96% Better surface pores, roughness, hydrophilicity, and antifouling. Pollutant layer thickness was proportional to membrane resistance/conductivity, allowing for prompt cleaning and prolonging membrane life.	(Yuan et al., 2020b)
M-GO/CNT-0.5- polyaniline (PANI)	CR	0.00274 g	99.9%; UF	496.4 and 230.4 L m <sup>-2</sup> h <sup>-1</sup> at 0.1 and 0 MPa	WCA of M-PES is 74.7° reduce to 46.6° with M-GO/CNT-0.5-PANI which increased permeability, selective separation, antifouling, mechanical strength, and extended life.	(Wang et al., 2021a)
Cu(tpa)@GO/ PES	CR	GO: 0.5 g (Cu(tpa)) 0.9@GO: 0.9:1	90.0%; UF	PWF: 140.0 L m <sup>-2</sup> h <sup>-1</sup> at 0.2 MPa	Cu(tpa) was used as MFO. WCA and FFR of (Cu(tpa))0.9@GO were 16° and 70% while PM (PES) was 84° and 43%. The nature of inserted MOFs boosted anionic dyes' selective rejection ratio (CR and MO). Better for wastewater EOMPS.	(Makhet ha and Moutloa li, 2018)
Guanidyl- functionalized graphene/polysulfon e mixed matrix	Antibacterial and antimicrobial0	0.5 wt.%	93.7%; UF	More than 200 L m <sup>-2</sup> h <sup>-1</sup>	FRR of PM PSF 73.4% increased to 82.4% with 1 wt.% of GFG. GFG/PSF CM had a porosity of 72.5-72.9%, an average pore width of 30.93-30.97 nm, and a WCA of 86.9-83.3°. Remarkable antimicrobial efficacy and endurance against Escherichia coli and Staphylococcus aureus.	(Zhang et al., 2019a)
Polyamide (PA) + GO	Norfloxacin	0.1 wt.%	53.32%; NF	12.78 L m <sup>-2</sup> h <sup>-1</sup> bar <sup>-1</sup>	GO reduced WCA of PA reduced from 60.82 to 49.3. GO enhanced membrane anti-fouling and norfloxacin and sulfamethoxazole rejection. GO inclusion makes active layer thinner,	(Wang et al., 2017)

					smoother, and denser and increases hydrophilicity and negative surface charge.	
PES/GO	BSA	0.1,0.5 and 1.0 wt. %	96.0; NF	$5.1 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$	FRR 90.5%; WCA: 53.2. Irreversible resistance reduced from 66.1% (PM) to 9.5% (MM) PES membranes with magnetic nanoparticles and a magnetic field improve FRR, antifouling, and PWF efficiency.	(Zinadini et al., 2014)
GO/Fe <sub>3</sub> O <sub>4</sub> /PES mixed matrix membrane	BSA	Fe <sub>3</sub> O <sub>4</sub>	98.0; UF	$175.6 \text{ L m}^{-2} \text{ h}^{-1}$	FRR 87.9%; Fe <sub>3</sub> O <sub>4</sub> Used as hydrophilic additives This membrane reinforcement for elevated NF and RO membranes works successfully.	(Mirzaei et al., 2021)
PVDF/GO-Fe <sub>3</sub> O <sub>4</sub>	BSA	Fe <sub>3</sub> O <sub>4</sub> + magnetic field	92.0; UF	$595.0 \text{ L m}^{-2} \text{ h}^{-1}$	FRR 86.4%; WCA 55.9 Magnetic field driven casting transported Fe <sub>3</sub> O <sub>4</sub> /GO nanocomposites from the membrane matrix to the surface, creating high-performance hybrid membranes.	(Xu et al., 2016)
M-GO/Fe <sub>3</sub> O <sub>4</sub> /PES mixed matrix membrane	BSA	Fe <sub>3</sub> O <sub>4</sub> + magnetic field	92.0; cross-flow filtration	$252.0 \text{ L h}^{-1} \text{ m}^{-2}$	FRR 80.1%	(Mirzaei et al., 2021)
PVDF/MGO magnetic Fe <sub>3</sub> O <sub>4</sub> /graphene oxide	BSA	Fe <sub>3</sub> O <sub>4</sub> + magnetic field	77.7; UF	$484.0 \text{ L h}^{-1} \text{ m}^{-2}$	FRR 83.0%; WCA 62.0; Outstanding magnetic susceptibility and hydrophobicity	(Huang et al., 2018)
Graphene oxide (GO)-based composite ultrafiltration (UF) membranes	atrazine	0.1 wt. %	95% atrazine elimination, total reduction of TSS, turbidity, and microbes; MBR and UF	At a backwash pressure of $4 \text{ kg cm}^{-2}$ for 1 hour of MBR examination, PF drop was reduced by approximately 5% following back pulsing of different duration and resting period.	FRR:98.6%. Membrane fouling lowers flow, and MLSS induces complete and intermediate blockage mechanisms. Compared to direct membrane filtration or biodegradation, this membrane improves the quality of the permeate.	(Mukherjee et al., 2018)

PWF: pure water flux; WP: water permeability; FRR: flux recovery ratio, FDR: flux decay rate; Rt: reaction time; T: Temperature; SRT: standard room temperature;

MP; membrane permeability; PM: pristine membrane; MM: modified membrane; CM: composite membrane, WCA: water contact angle, PF: Permeation flux; RS:

rebinding selectivity; SF: suction filtration; CFF: cross-flow filtration; MAC: membrane adsorption capacity

**Table 2GO mediated membranes in degradation membranes**

Compound name	Target pollutant	Supporting components and removal mechanism	Pollutants removal efficiency	Flux/hydrophilicity/permeability	Major findings and recommendations	Reference
GO-loaded PDA-TC-SiO <sub>2</sub> /AC@PVDF	TC, CEX, SMZ and OTC	Adsorption/filtration/ degradation	106.44 mg g <sup>-1</sup>	3600 L h <sup>-1</sup> m <sup>-2</sup>	DI-MMMs retained 93.5% of initial rebinding capacity after 10 cycles. Improved DI-MMM identification and permeability could help molecularly imprinted membranes overcome the permeability-selectivity compromise.	(Ma et al., 2022)
graphene oxide (GO)/SiO <sub>2</sub> -loaded dual-imprinted membranes (GS-DIMs)	antibiotics TC; cefalexin (i.e., sulfamethazine, and oxytetracycline)	0.15 g GO nanosheet	RS: > 3.5; Rejection: (CEX, 98%; SMZ, 99%, and OTC, 97%); SF, adsorption	TC: 4.27, CEX: 3.88 SMZ: 4.11 OTC: remain unchanged in 5 cycles	The CM GO/SiO <sub>2</sub> -loaded nano CM worked well in industrial-scale applications and effluent samples. 65.61 mg/g was the total efficiency of elimination (after 20 min)	(Wu et al., 2021b)
GO/CNTs-modified membrane graphene oxide (GO)/carbon nanotubes (CNTs)	DOC	GO/CNTs: 1:4	69%; CFF/adsorption	DOC water flux of PM and MM was 1280 and 667 kg m <sup>-2</sup> h <sup>-1</sup>	MM exhibits good antifouling and rejection rates stability. The wide exterior diameter of CNTs degraded their antifouling efficacy and removal efficiency, whereas small MW cross-linking agents produced total blockage. In the case of HA + Ca <sup>2+</sup> , the linking effect of Ca <sup>2+</sup> led to intermediate fouling for all membranes. The WF inhibition increased with increasing CNT mass percent in pure GO, 2:1, 1:2, and 1:4 mixtures.	(Zhao et al., 2021)
PVDF/GO@SiO <sub>2</sub> /PVP	BSA	adsorption	78.5%; adsorption	1232 L h <sup>-1</sup> m <sup>-2</sup>	FRR: 77.5%; WCA 63.02; Nanohybrid particle (GO@SiO <sub>2</sub> ) has the best antifouling properties and boosts PVDF membrane performance.	(Zhu et al., 2017b)
chitosan-graphene oxide (CS_GO) CMs	20 types of Pesticides	1% to 20%, w/w	95% removal of rifampicin; adsorption	-	The affinity for the most hydrophilic contaminants was decreased by GO, and the R% was enhanced. CS demonstrated Low pollutant extraction with logK <sub>ow</sub> values between 0.8 and 2, and integration of GO at a low level (1%)	(Silvestro et al., 2021)
Ultem + 2.5 wt.% GO	Ibuprofen (IBU) non-steroidal anti-inflammatory drugs	0.5 wt.%	53%; adsorption	-	Through non-covalent $\pi$ - $\pi$ contacts between the GO interface and the benzyl ring of IBU, the GO sheet can facilitate faster electron/ion transport.	(Bourassi et al., 2021)

GO-Fe-TA-APTES	endocrine disruptor, BPA, 6-methoxytetralin, naphthalene and pyrene	Fe <sup>3+</sup> ; tannic acid-aminopropyltriethoxysilane (TA-APTES).	PM:26% MM:81%; adsorption	-	GO-Fe-coated membranes can be used in pharmaceuticals, coking, and municipal wastewater treatment. GO-Fe-TA-APTES had no membrane permeability loss after 50 mL filtration, while GO-Fe had 36%. After filtration of 50 mL BPA solution MTHN (70%)	(Li et al., 2021b)
GO-GDBM gravity-driven biomimetic membrane (GO-GDBM)	BPA	CNT/GO dosages (20–65 g m <sup>-2</sup> ) Supported by: biodegradable CA	55~70%; adsorption; Static filtration;	72.1 L h <sup>-1</sup> m <sup>-2</sup> at 12 cm of hydrostatic height; After 30 days: 27~42 L h <sup>-1</sup> m <sup>-2</sup>	As a GDM, GDBM, specifically CNT-GDBM and GOGDBM, with extraction rate more than 60% and fluxes bigger than 35 Lm <sup>2</sup> h can appear as a feasible water purification method in open natural water systems.	(Zhu et al., 2020b)
graphene, graphene oxide, carbon nanotubes and C60	steroid hormones	Supported by: biodegradable CA	GP-AO <sub>2</sub> : 6.6 mg/g; adsorption;	-	CNT and GO demonstrated high Biodegradable CA UF membrane screening abilities (over 10 mg g <sup>-1</sup> ) for hormones which remained undetectable in isotherm tests.	(Ngunyen et al., 2021)
GO-p-PES membrane (UF)	polyethylene glycol (PEG)	GO:10 mg mL <sup>-1</sup> ; Supported by: biodegradable CA	70% adoption reduce with GO; >90% rejection; adsorption;	Flux decline in PES (65%) was reduce to 25% with GO. FRR in PES (46% while with GO ~ 80%)	In the long run, GO-functionalized polyampholyte hydrogel appears promising for treatment of wastewater. GO loading promoted antibacterial and antibiofouling properties.	(Zhang et al., 2018)
carbon-coated nanofiltration membrane (NF90)	Phenol	Supported by: biodegradable CA	99%; MF, UF, NF; adsorption	36 L h <sup>-1</sup> m <sup>-2</sup> 10 bar <sup>-1</sup>	The NF90 membrane can remove phenol under low pressure 6 bars. GO-coated membranes have a low removal rate due to their high polarity (hydrophilicity).	(Cetinkaya and Ozdemir, 2018)
co-deposition of DA/GO and DA/GO-Zn	bisphenol A (BPA)	Laccase; biocatalysis; NF	No effect on rejection.	permeate flux coNF-PDA/GO, 7.46 L m <sup>-2</sup> h <sup>-1</sup> ; coNF-PDA/GO-	GO's enrichment and catalytic characteristics increased laccase activity and stability. The modified membrane showed promising results for removing and detecting EOMPS.	(Zhang et al., 2019b)

				Zn: 29.85 L m <sup>-2</sup> h <sup>-1</sup>		
GO&Lac@UF GO additive Biomimetic dynamic membrane (BDM)	MB	Supported by: biodegradable CA	80%; adsorption; UF	≥120 L h <sup>-1</sup> m <sup>-2</sup>	WCA: decreased from 86.43 (PM) to 77.78 (GO&Lac). Laccase activity was higher on GO than CNT. Appropriate production method, GO, and laccase dosage can maximize biomimetic layer and enzyme immobilization, increasing its sustainability.	(Zhu et al., 2020a )
poly(ether sulfone) PES + GO (UF)	Ibuprofen	Supported by: biodegradable CA	44.9%; UF, biodegradable adsorption;	flux 5.27 L h <sup>-1</sup> m <sup>-2</sup> bar <sup>-1</sup>	GO membrane layers gaps affects steric hindrance and elimination rate. GO removed PPCPs in their molecular state using biodegradable CA ultrafiltration and molecular sieving rather than conventional nanofiltration membranes, which rely on electrostatic repulsion.	(Lou et al., 2020)
β-cyclodextrin (β- CD) modified graphene oxide (CDGO) membrane	bisphenol A (BPA)	β-CD molecules; Supported by: biodegradable CA	100%; adsorption, catalytic degradation	water fluxes increase with operating pressure, flux was one or two order of higher than RO and NF	CDGO membrane has stable structures β-CD molecules enhanced surface areas and winding 2D nanochannels and BPA removal. CDGO biodegradable CA ultrafiltration membrane separation capabilities are many times higher than PM, allowing for a larger range of applications in molecular separation and easy regeneration after multiple cycles using ethanol washing.	(Chen et al., 2020)
graphene- containing ceramic composite tubular membrane (TGCCM)	emerging contaminants (ECs) in water, (DnBP, DEHP, CLX, SMX, CAF)	GO nanosheet: 2 g L <sup>-1</sup> ; Electrocoagulation on adsorption, Biodegradable CA	DnBP and DEHP removed by 99%; CLX, SMX and CAF by 32-97%; adsorption	140 L h <sup>-1</sup> m <sup>-2</sup>	Size exclusion, electrostatic repulsion, Biodegradable CA ultrafiltration membrane separation, electrocoagulation, and electrofiltration are the removal mechanisms	(Yang et al., 2016)

**Table 3GO mediated membranes in catalytic and degradation membranes**

Compound name	Target pollutant	supporting components/ Experimental condition	Pollutants removal efficiency	Flux/ hydrophilicity/ permeability	Major findings and recommendations	Reference
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graphene oxide (GO)	micropollutants DCF (2,6-dichlorophenyl)	K <sub>2</sub> FeO <sub>4</sub>	FeVIO <sub>4</sub> <sup>2-</sup> :20%; GO-FeVIO <sub>4</sub> <sup>2-</sup> : 100%; oxidation	-	The better removal of EOMPs is dictated by their structure (or moieties), which may require a reaction with high-valent iron species (FeIV/FeV) to produce the accelerated action brought about by carbonaceous substances.	(Pan et al., 2020)
G.O and rGO	CBZ	K <sub>2</sub> FeO <sub>4</sub> <sup>-</sup>	Removal by FeVIO <sub>4</sub> <sup>2-</sup> :30%; GO: <10%; GO-FeVIO <sub>4</sub> <sup>2-</sup> : 100%; oxidation	-	GO with FeVIO <sub>4</sub> <sup>2-</sup> showed 60% and 50% more removal of ZBZ compared to rGO and with rGO with FeVIO <sub>4</sub> <sup>2-</sup>	(Pan et al., 2020)
graphene oxidepolyethyleneimine (GO-PEI)	(Fe(III)-TAML)	H <sub>2</sub> O <sub>2</sub>	100%; UF; Catalytic oxidation	192 L h <sup>-1</sup> m <sup>-2</sup>	GO-PEI boosted porosity and hydrophilicity. H <sub>2</sub> O <sub>2</sub> activates enzyme-like ligand, Iron (III)- Tetra-amido macrocyclic ligands to produce high-valence Fe. Five cycles of BPA removal were steady.	(Zhang et al., 2021)
TAML/GO-PEI/PES graphene oxide polyethyleneimine (GO-PEI)	BPA	Iron (III)- Tetra-amido macrocyclic ligand (Fe(III)-TAML) H <sub>2</sub> O <sub>2</sub> ;	100% remove; Catalytic oxidation; UF	192 L h <sup>-1</sup> m <sup>-2</sup>	GO-PEI modification in PES membrane enhanced porosity and hydrophilicity and modification increased SSA from 20.03 to 22.36 m <sup>2</sup> /g, FRR from 34% to 68% while contact angle dropped from 86.5° to 63.4°.	(Zhang et al., 2021)
graphene oxidepolyethyleneimine (GO-PEI)	(Fe(III)-TAML)	H <sub>2</sub> O <sub>2</sub>	100%; UF; Catalytic oxidation	192 L h <sup>-1</sup> m <sup>-2</sup>	GO-PEI boosted porosity and hydrophilicity. H <sub>2</sub> O <sub>2</sub> activates enzyme-like ligand, Iron (III)- Tetra-amido macrocyclic ligands to produce high-valence Fe. Five cycles of BPA removal were steady.	(Zhang et al., 2021)
N-doped GO	phenol in aqueous solution.	persulfate (PS); catalytic oxidation	Oxalic acid (100%) phenol(94%); Catalytic oxidation	-	Negligible elimination by adsorption. The catalytic mechanism is driven by both radical and nonradical degradation of contaminants, however singlet oxygen species were discovered to contribute more significantly.	(Pedro sa et al., 2019)
GO-MWCNTs Co@NCNT-MS membrane (2–2–20)	TC, RhB, and MO		Average: 90% 100%; Filtration/ Fenton degradation	40 L h <sup>-1</sup> m <sup>-2</sup> (TC); 70 L h <sup>-1</sup> m <sup>-2</sup> (TC, RhB, and MO)	In addition to adsorption sites, radical (SO <sub>4</sub> <sup>-</sup> , OH <sup>-</sup> , and O <sub>2</sub> <sup>-</sup> ) and nonradical (IO <sub>2</sub> ) species help in degradation. The Co@NCNT-MS cobalt-carbon heterogeneous Fenton-like catalyst eliminates toxins in complex wastewater effluents.	(Li et al., 2022b)

GO nanosheets additive 3D Fe-based metal-organic framework heterostructured membrane	Prussian blue	Fe(III)/Fe(II)	98%; separation and Fenton oxidation	27 L m <sup>-2</sup> h <sup>-1</sup>	Sandwich type membrane sheets arrangement provide high flux. In Fenton, GO nanosheets provided enough catalytic reaction time for PB to break down, resulting in excellent oxidation efficiency. Unique isolation and degradation method for EOMPs in wastewater.	(Song et al., 2022)
GO	Octadecyltrichlorosilane (ODTS) Solution Concentration (1%) PPCPs (TC)	0.5 and 1% wt	50%; Fenton degradation/adsorption	520 L h <sup>-1</sup> m <sup>-2</sup> bar <sup>-1</sup>	GO attributed the membranes' changes increased the efficacy of extracting PPCPs from water.	(Polak et al., 2021)
Co <sub>1</sub> -GO membrane	1,4-dioxane (1,4-D)	PMS, vitamin C as a mild reducing agent; membrane filtration and AOPs	GO: 99% Co <sub>1</sub> -GO: and Co <sub>1</sub> -GO 3 mM PMS: 100%; Catalytic degradation	2.1 ± 0.4 L h <sup>-1</sup> m <sup>-2</sup>	The membrane's capacity to reject big organic molecules mitigated their radical-scavenging effects. The kinetics of 1,4-dioxane breakdown were substantially quicker (>640 times)	(Wu et al., 2022a)
Fe <sub>2</sub> O <sub>3</sub> @Co@nitrogen-doped reduced graphene oxide@carbon nanotube composites	SMX	Peroxymonosulfate	98.5% (0.128 min <sup>-1</sup> ); Catalytic degradation	64.6 and 487.3 L h <sup>-1</sup> m <sup>-2</sup> bar <sup>-1</sup> i.e., 7.5-fold in MM than PM	2D confined catalytic membranes with oxygen vacancies are used to purify sewage. FeCo@GCTs is an ideal electron receiver; turf design and increased oxygen vacancies completely remove SMX and promote Cr (VI) reduction.	(Ye et al., 2021b)
GO-TiO <sub>2</sub>	naproxen (NAP), carbamazepine (CBZ) and diclofenac (DCF)	H <sub>2</sub> O <sub>2</sub> , UV-LED irradiation,	CBZ: 90% DCF: 100% NAP:80%; photodegradation/adsorption	92.6 ± 3.6 L h <sup>-1</sup> m <sup>-2</sup> Bar <sup>-1</sup>	300 nm UV-LEDs showed energy efficiency and optimum removal. UV-LED irradiation, modified membrane eliminated 100% of NAP, DCF, and CBZ in 20 min. Surface flow and dead-end filtering degraded CBZ by 15% and 75% in 120 min. Filtration quadrupled deterioration efficiency. The addition of H <sub>2</sub> O <sub>2</sub> to 365 nm/H <sub>2</sub> O <sub>2</sub> increased CBZ elimination by 20%.	(Li et al., 2022a)
GO-ZnO/ PES	trace organic compounds (TOrcs) CBZ, metoprolol and theophylline	0.05% GO-ZnO	PM:<40%; PES + 0.05% GO-ZnO: >60%; CFF/photodegradation	>78 L h <sup>-1</sup> m <sup>-2</sup>	The introduction of GO-ZnO enhanced trace organic refusal by decreasing membrane-solute hydrophobic affinities; the membranes degraded BB dye; and the modified membranes were less vulnerable to organic fouling by sodium alginate.	(Mahlangu et al., 2017)

N-TiO <sub>2</sub> /graphene oxide particles	MB	TiO <sub>2</sub>	Initial MB 35.8 mg L <sup>-1</sup> after filtration 12.5 mg L <sup>-1</sup> , degrade 22.3 mg L <sup>-1</sup> ; photocatalysis, UF	70 L m <sup>-2</sup> h <sup>-1</sup>	WCA of is 81° reduce to 64° with GO. The kinetics for NTG-M in sunlight is 4% faster than in UV.	(Chen et al., 2017)
UiO-66_GO/NF photocatalytic Nanofiltration Membrane	CBZ and DCF	photocatalytic activity under ultraviolet (UV) irradiation	CBZ & DCF: 90%/20 min; CBZ (70%) and DCF (93%) after 2 h; photocatalytic degradation	Water flux (up to 63 kg m <sup>-2</sup> h bar)	FRR: 80%; fouling tolerance:33% In water treatment, the UiO-66 GO nanocomposite is a promising addition to enhanced photocatalytic NF membranes for addressing constrained flow, high energy consumption, and short membrane lifetime.	(Heu et al., 2020)
GO/TiO <sub>2</sub> membrane	Methylene Blue	UV light irradiation, photocatalytic activity under ultraviolet (UV) irradiation	92%; photocatalytic degradation	7.5 L m <sup>-2</sup> h <sup>-1</sup>	FRR 96%; UV radiation has the potential to greatly reduce membrane fouling. Adsorption and cake layer were shown to be the main causes of GO/TiO <sub>2</sub> membrane fouling, accounting for 46.3% and 46.0% of membrane resistance, respectively.	(Zhu et al., 2017a)
TiO <sub>2</sub> -graphene oxide (GO)-Fe <sub>3</sub> O <sub>4</sub>	refractory antibiotic amoxicillin organic compounds in aqueous solution	Fe <sub>3</sub> O <sub>4</sub> /photodegradation	TiO <sub>2</sub> -GO and Fe <sub>3</sub> O <sub>4</sub> :14.7% TiO <sub>2</sub> -GO-18 wt.% Fe <sub>3</sub> O <sub>4</sub> :90%; 22.3% oxidation by Fe <sub>3</sub> O <sub>4</sub> and 15.2% by photocatalytic degradation	-	GO enhances the catalytic cycle and efficiency. When the concentration of H <sub>2</sub> O <sub>2</sub> was increased from 0 to 20 mM, the removal performance climbed from 10% to 90%. SMSMPR CM has excellent photo-Fenton catalytic activity and AMX degradation stability. The AMX intermediates disintegrated into carbon dioxide, water, and inorganic ions.	(Li et al., 2019)
electrochemically modified carbon nanofibrous anodic membrane (G/SnO <sub>2</sub> /CFs)	refractory toxic organic - sulfamethoxazole (SMX)	SnO <sub>2</sub> ; electrooxidation	85%; degradation	PMF is 74 144.9 and 216.4 L h <sup>-1</sup> m <sup>-2</sup> at 10, 20 and 30rpm peristaltic pump	Degradation is reduced by increasing the quantity of anodic membrane and using flow-through mode. Nine non-toxic SMX intermediates are discovered using LC-MS/MS. Membrane stability was excellent even at 10 cycles, 50 h reaction. electro oxidative performance and membrane conductivity can be increased by SnO <sub>2</sub> and graphene.	(Yu et al., 2020)

electrochemically active cathodic membrane (CFC/PVDF/GO-Fe <sup>3+</sup> )	Rhodamine B	Fe <sup>3+</sup> ; electrooxidation	10 mg L <sup>-1</sup> Rhodamine B was ~75% in CSTR; degradation	200 L h <sup>-1</sup> m <sup>-2</sup>	In-situ Fe <sup>2+</sup> regeneration on membrane cathode causes decolorization at pH > 4.0. Only avoiding Fe <sup>3+</sup> -oxalate complexes improved membrane oxalate oxidation stability.	(Li et al., 2018)
Graphene nanoplatelets: CNT/PTFE filter	[Fe(CN) <sub>6</sub> ] <sup>4-</sup> , TC, Phenol, Oxalate, COD, TDS, Nephelometric, Turbidity Unit	electrooxidation and electrochemical filtration	(COD, 91.67%); (TDS, 91.3%), (TSS, 98.85%) and (NTU, 96.45%) degradation	52 L h <sup>-1</sup> m <sup>-2</sup>	The promise of graphene electrode in flow-through purifiers is demonstrated by a graphene-based electrochemical filter using CNTs as conducting binders.	(Liu et al., 2014)
Hybrid graphene-decorated metal hollow fibre membrane reactors	pharmaceutical pollutant, paracetamol	Electro-Fenton; electrochemical filtration	membrane: 37%; Membrane: 165% wit degradation	High flux and selectivity; comparatively low water flux reduce from 5000 to 4500 L m <sup>-2</sup> h <sup>-1</sup> after three cycles	GO cathode performance were highest than all other and showed Tri-cycle stability. Low-cost catalytic membrane reactors with high flux and selectivity from materials reactivity, with promise for mineralizing persistent pollutants.	(Le et al., 2019)
ZIF@MILs-20/GO	norfloxacin (NX)	singlet oxygen ( <sup>1</sup> O <sub>2</sub> ), electro-Fenton batch experiments,	99.4% degradation	stable flux of 156.3 L m <sup>-2</sup> h <sup>-1</sup> bar <sup>-1</sup> over 10 h	The primary cause of NX degradation is <sup>1</sup> O <sub>2</sub> . Supplemented oxygen vacancies restrict FexOy, and 1D/2D hierarchically porous ZIF@MILs-20 frameworks enhance NX removal via a synergistic "trap-and-zap" method for PMS activation. Catalytic membranes eliminate fouling via filtration and catalytic oxidation of organic compounds.	(Ye et al., 2021a)

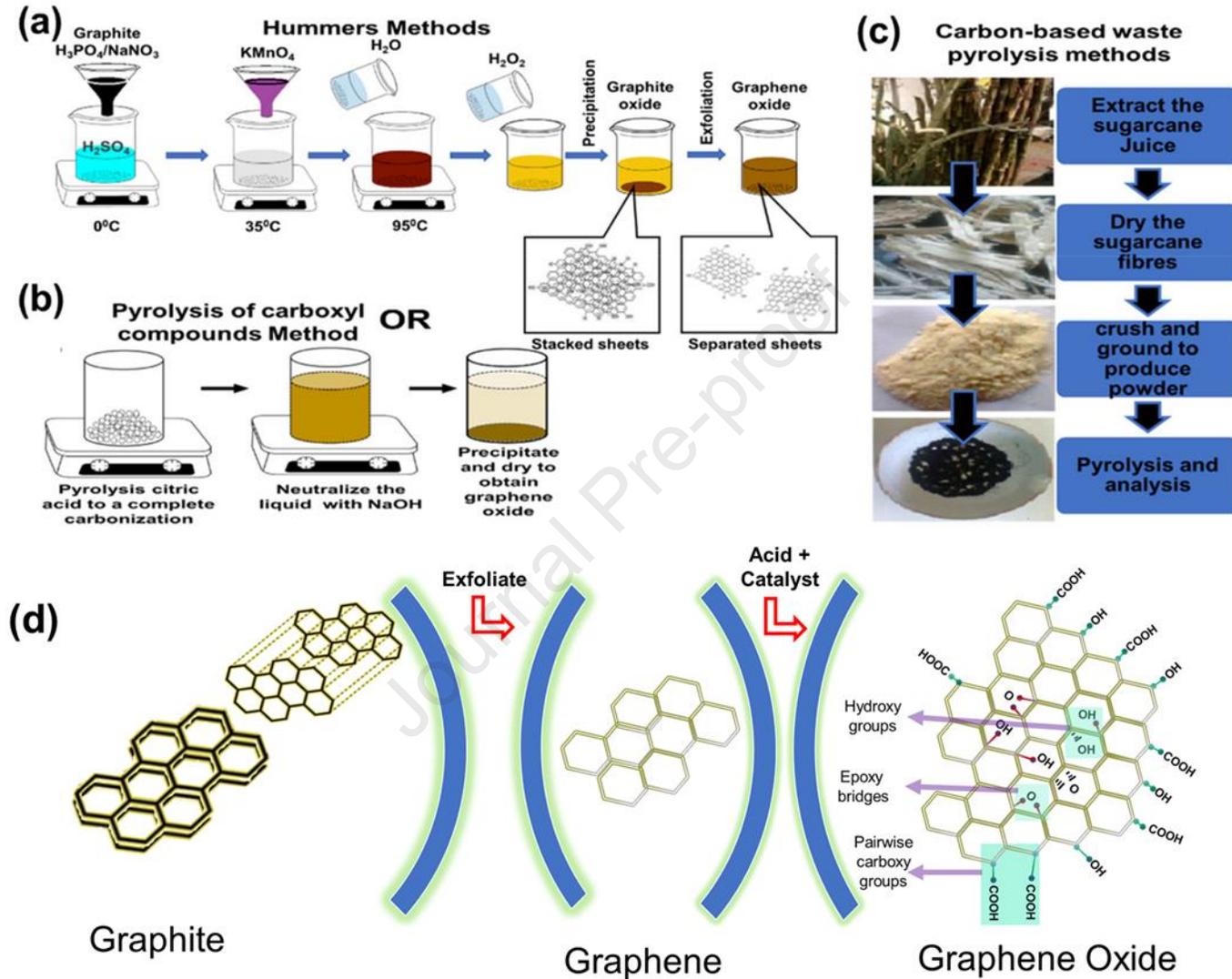


Fig.1

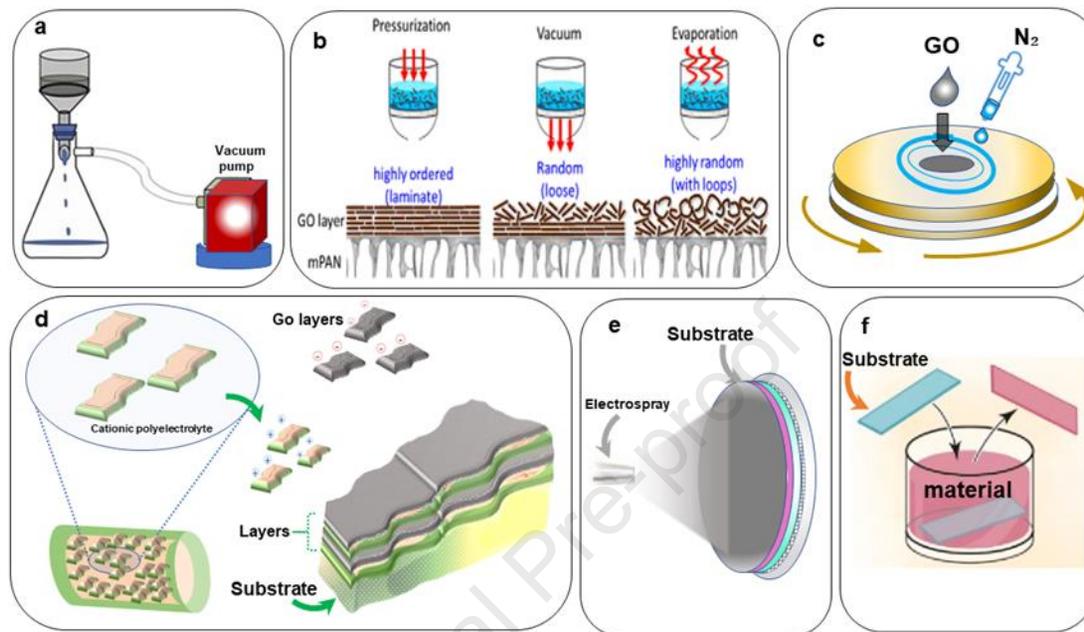


Fig.2

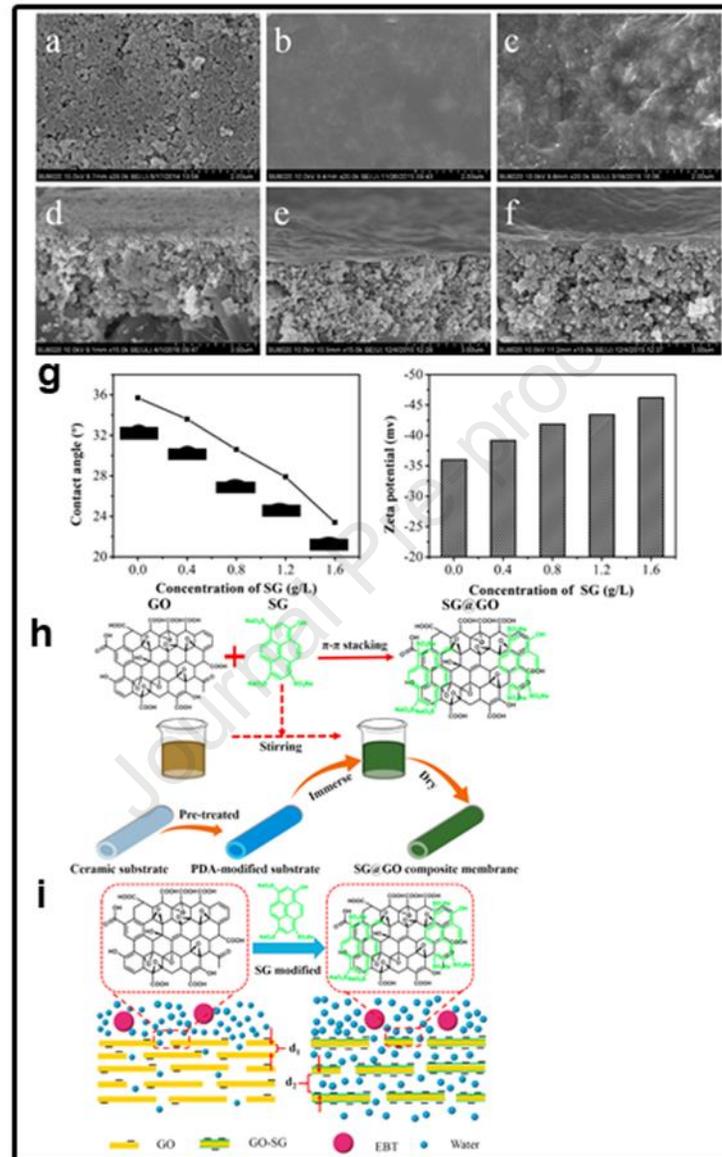


Fig.3

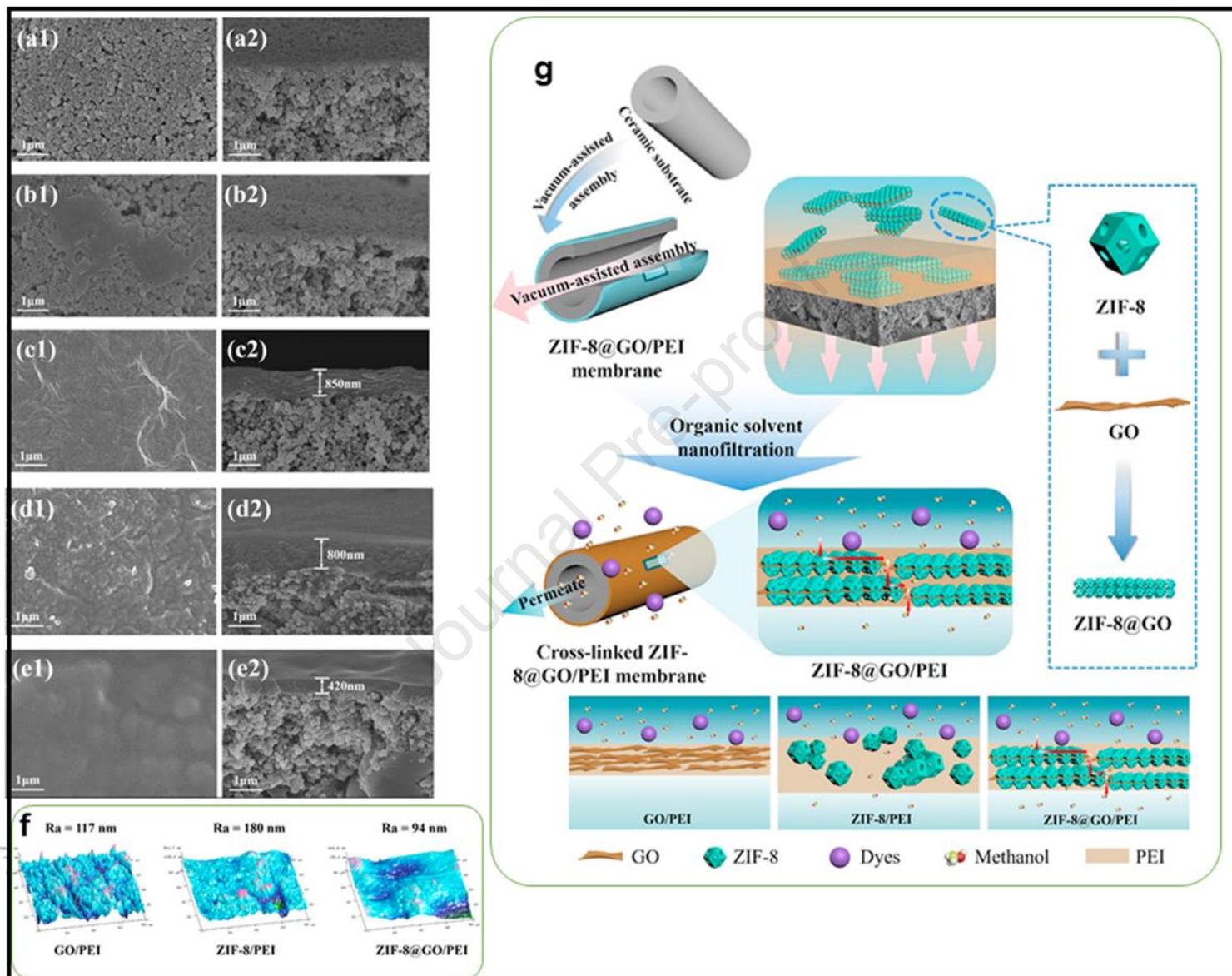


Fig.4

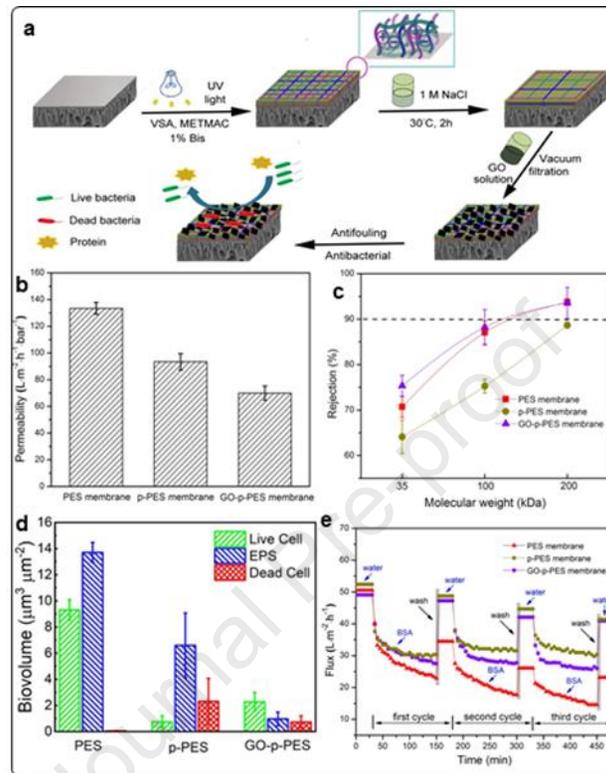


Fig.5

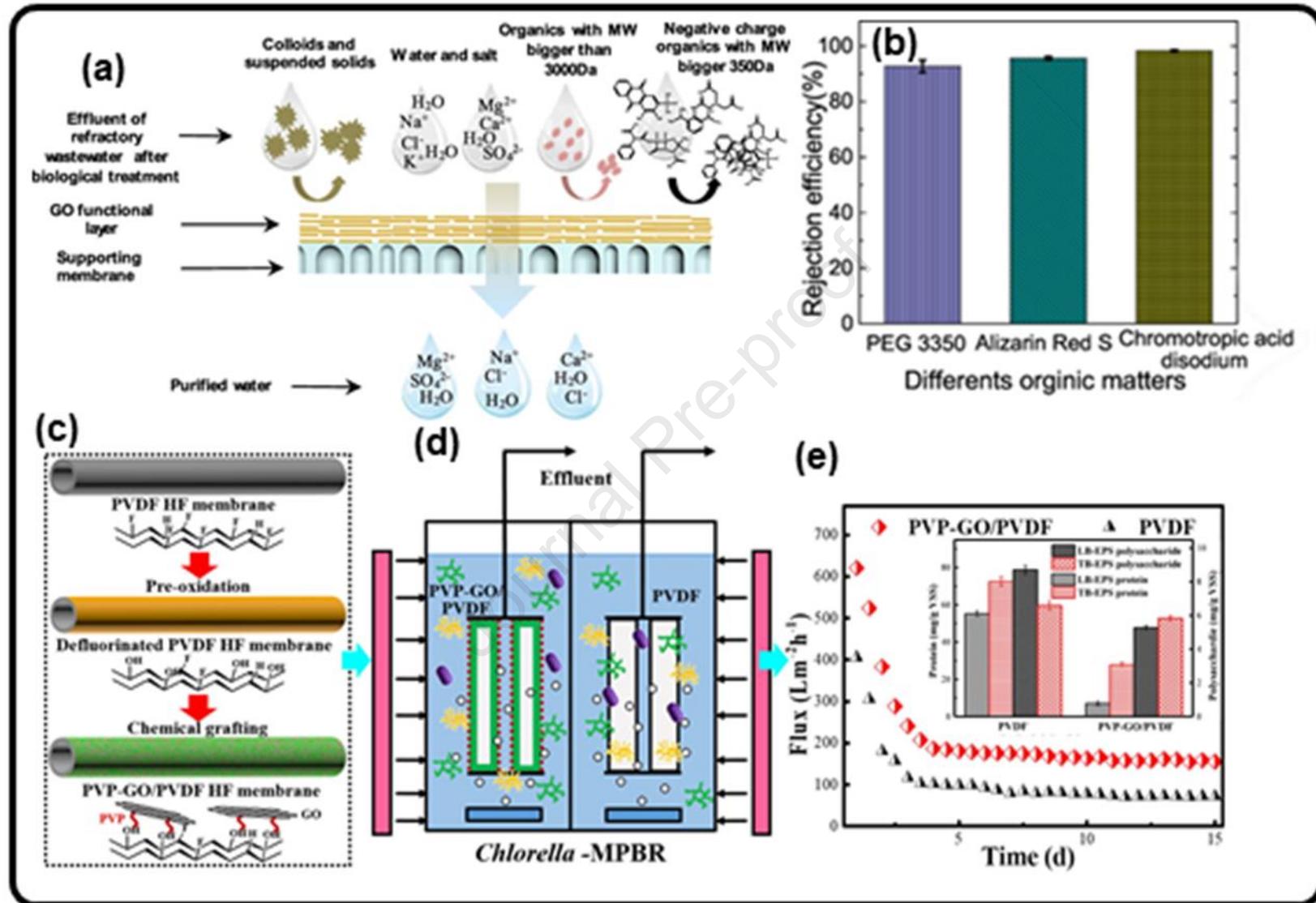


Fig.6

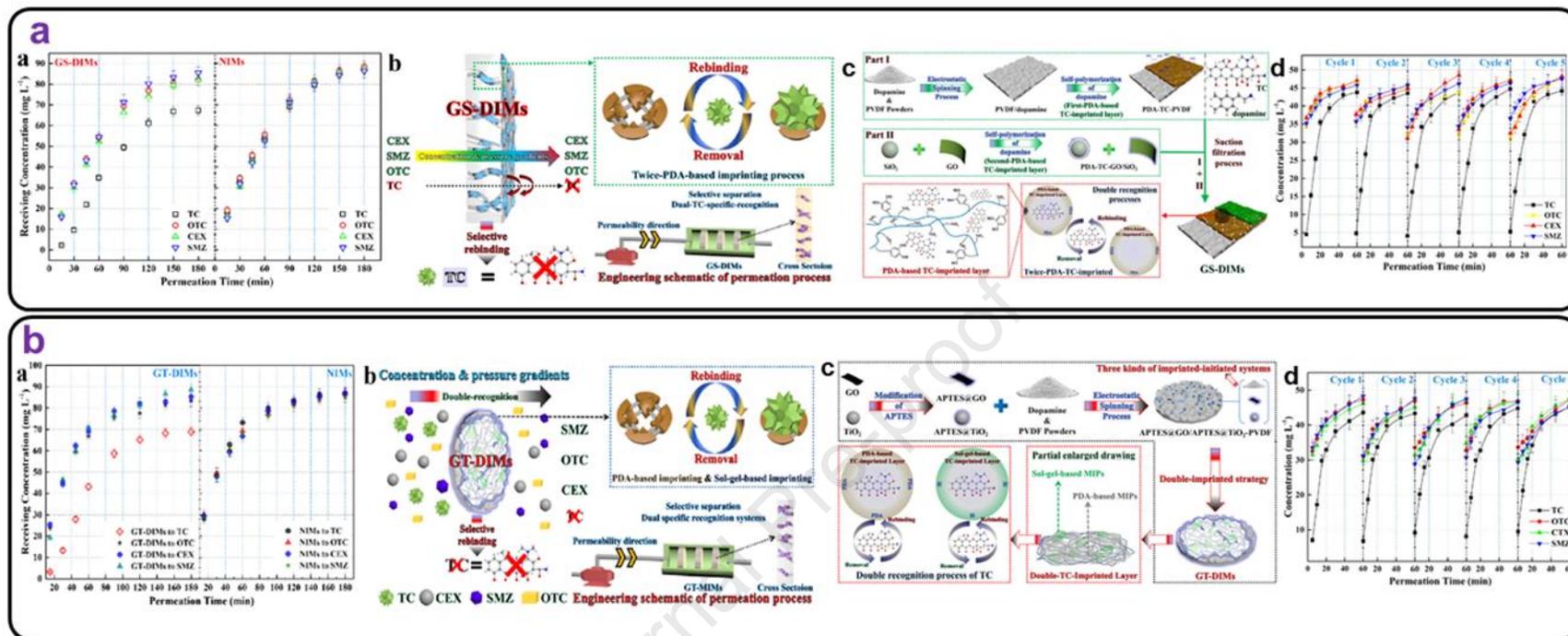


Fig7

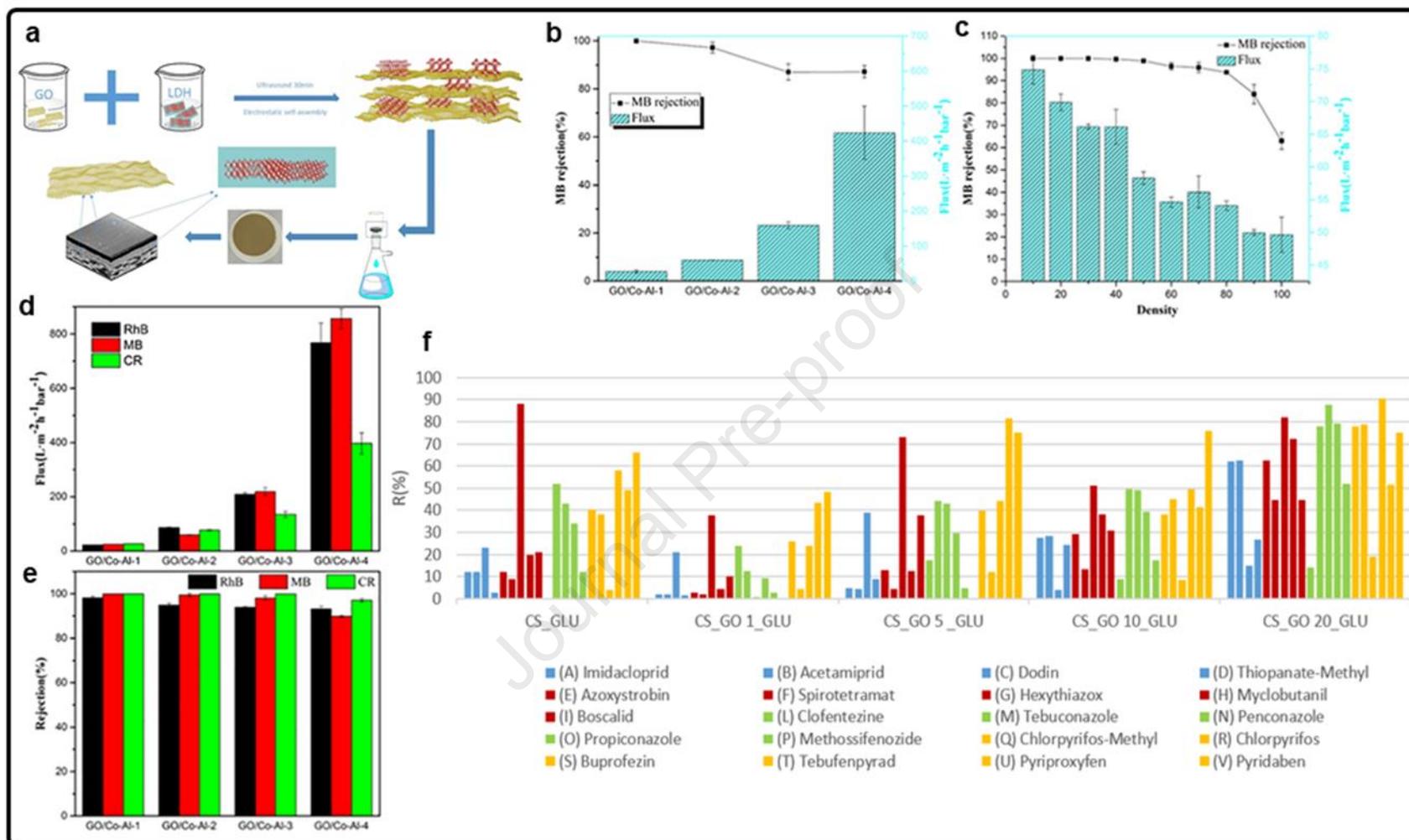


Fig.8

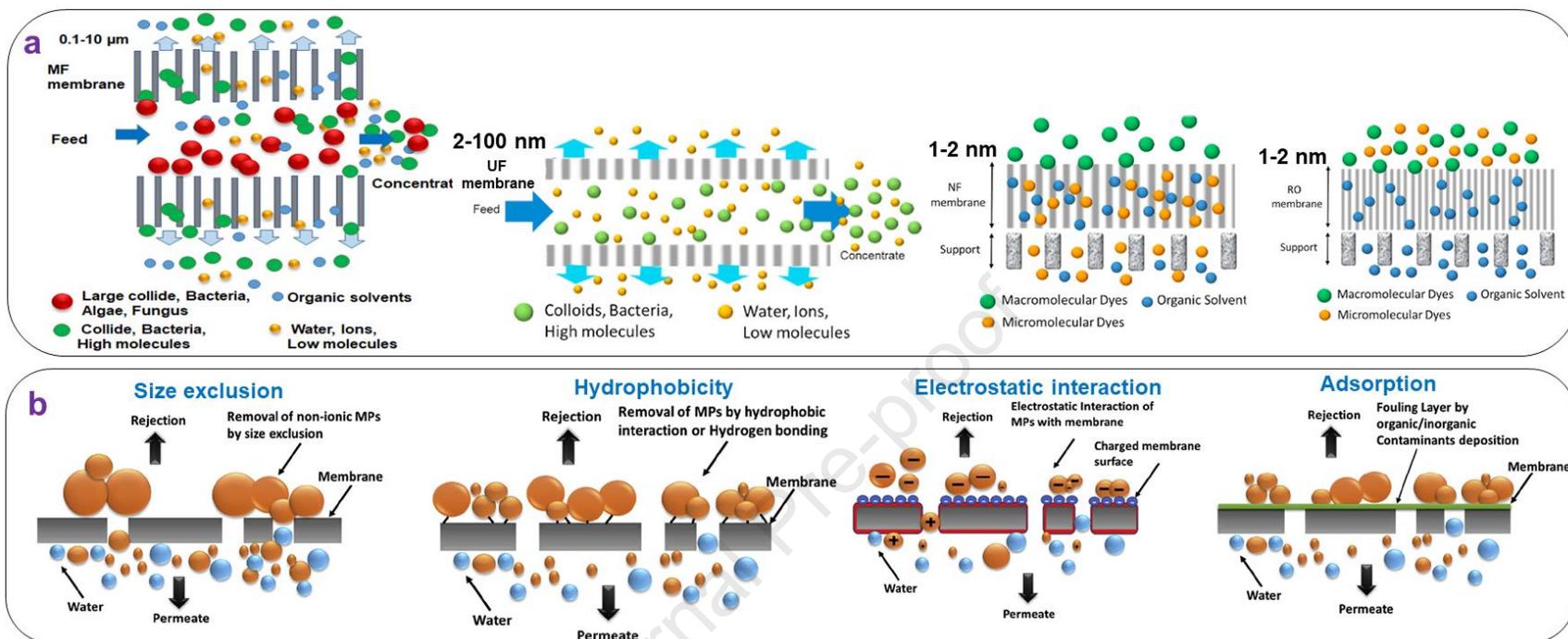


Fig.9

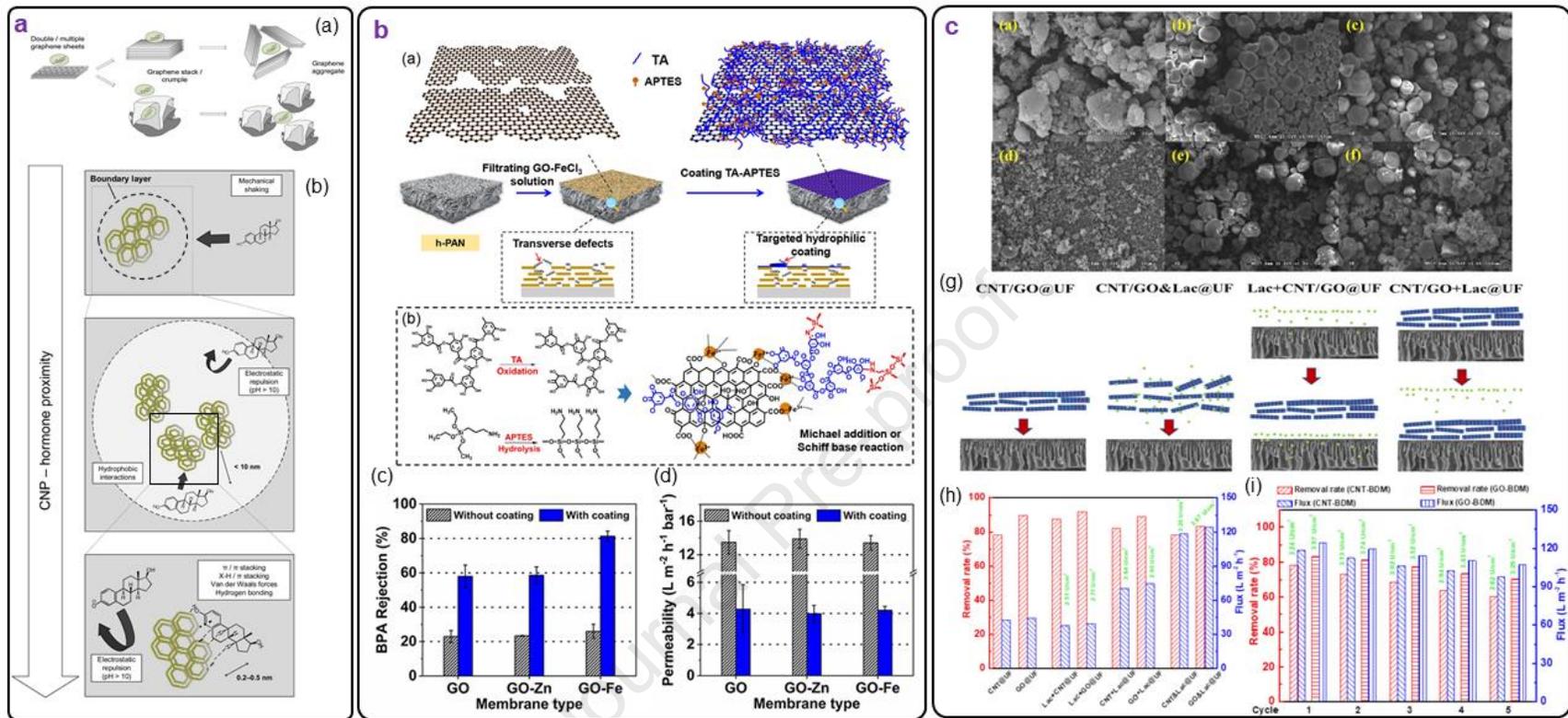


Fig.10

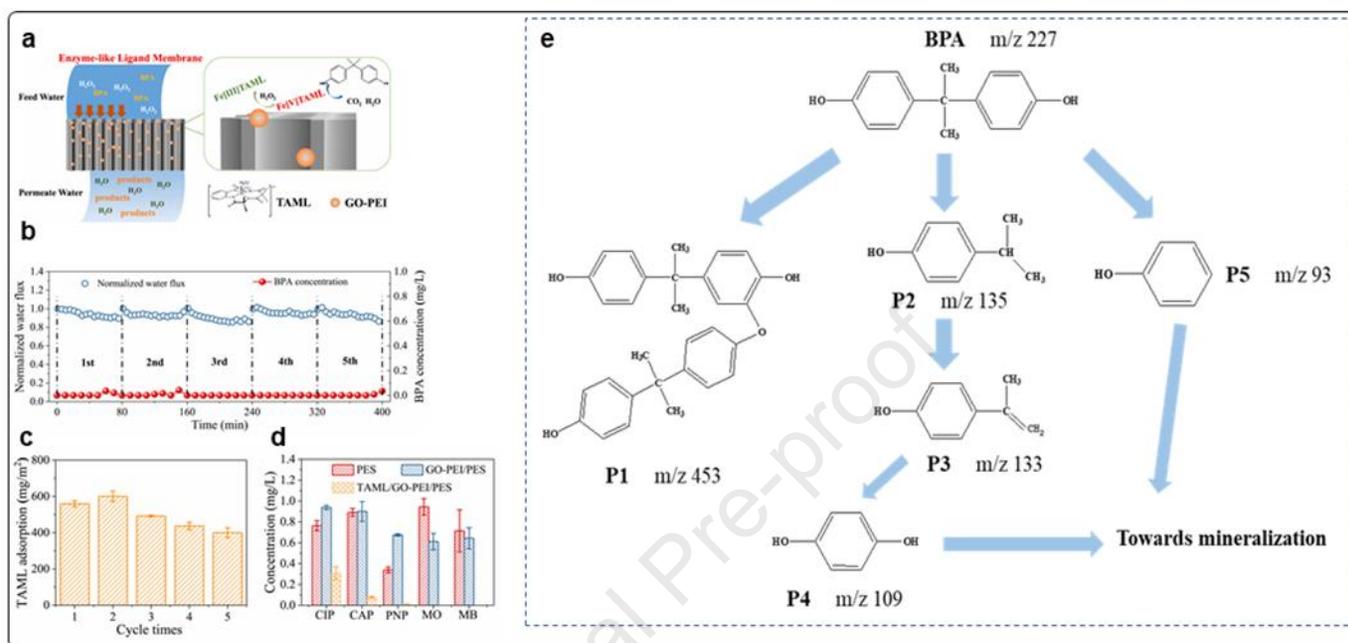


Fig. 11

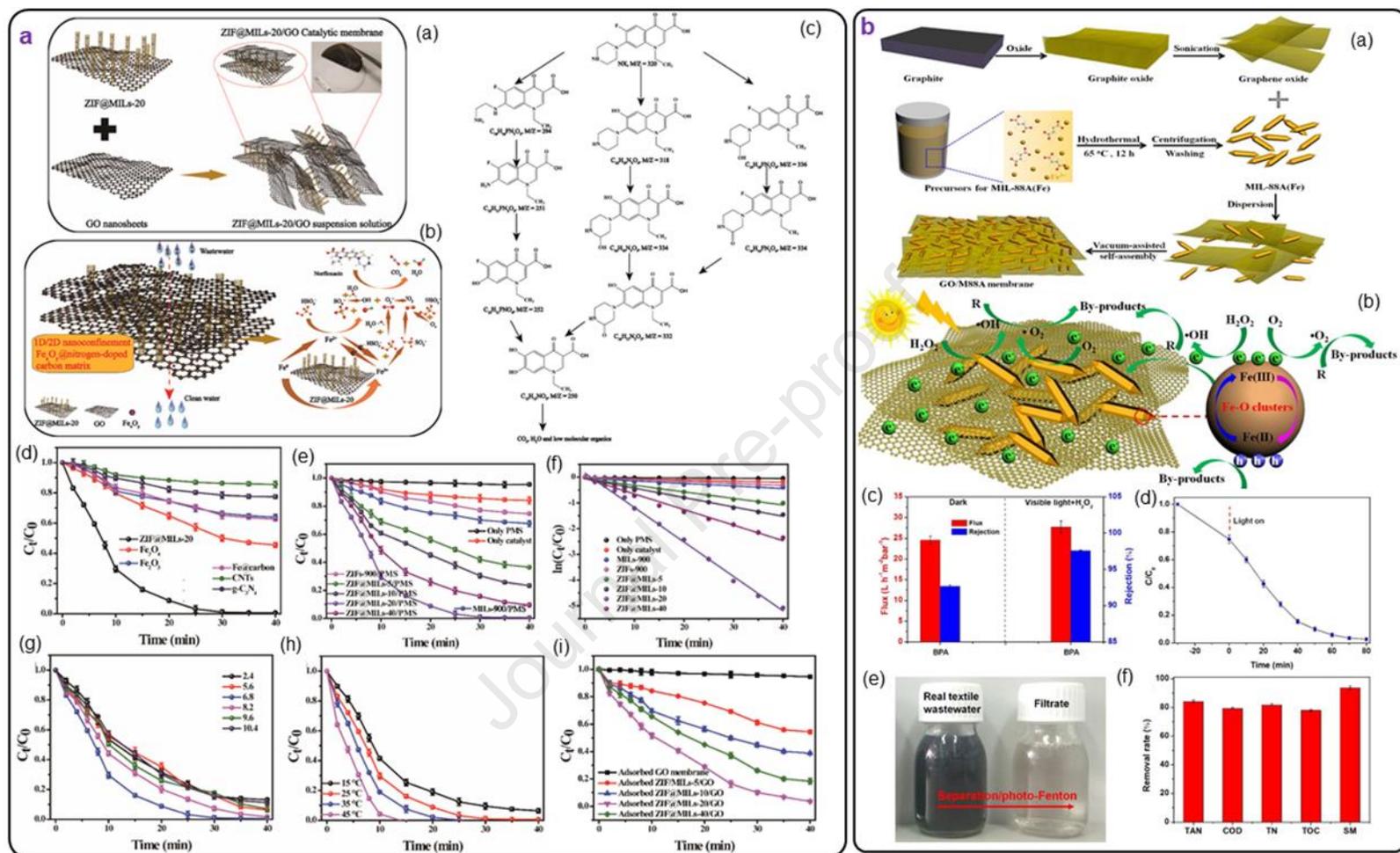


Fig.12

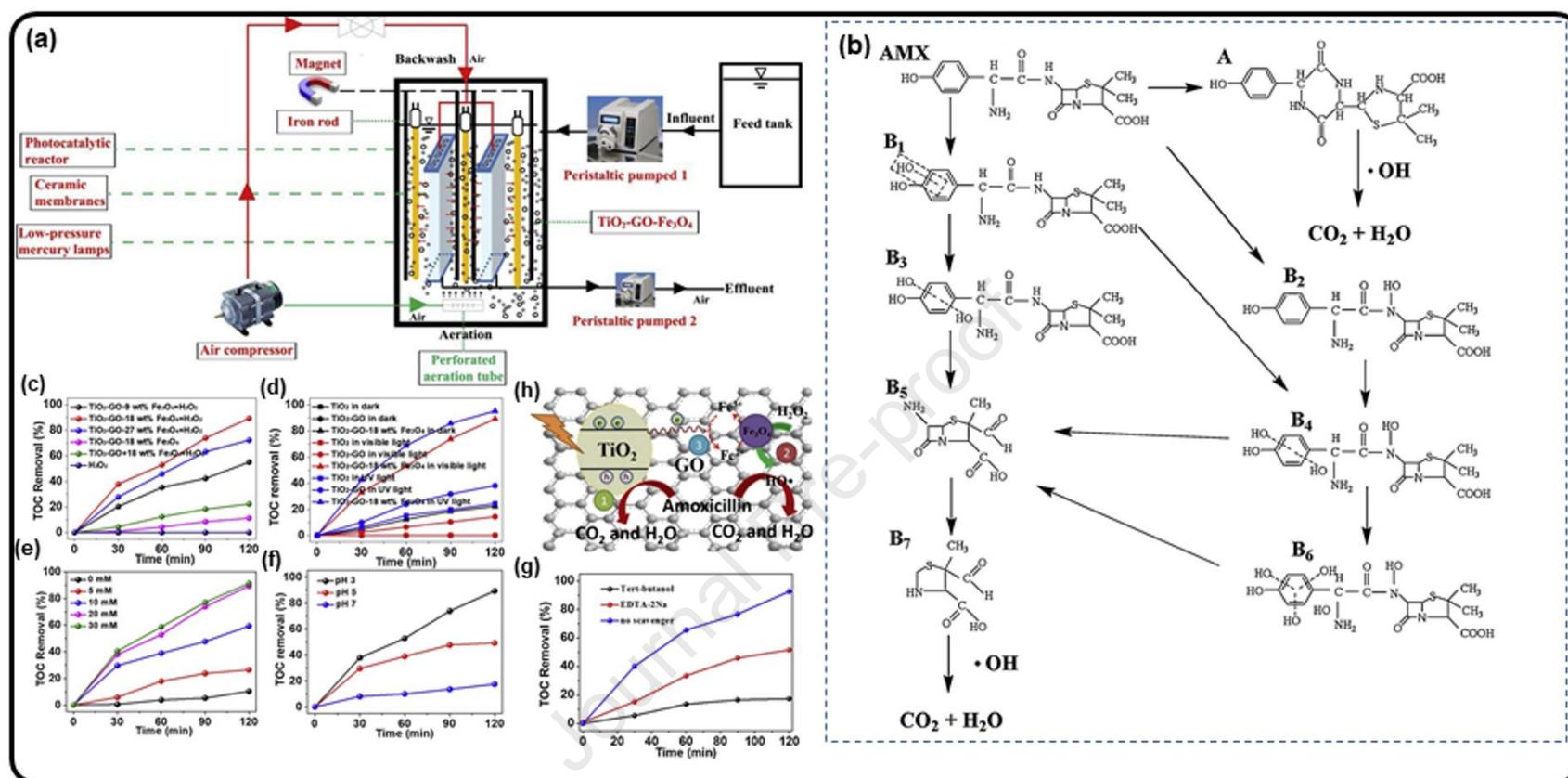


Fig.13

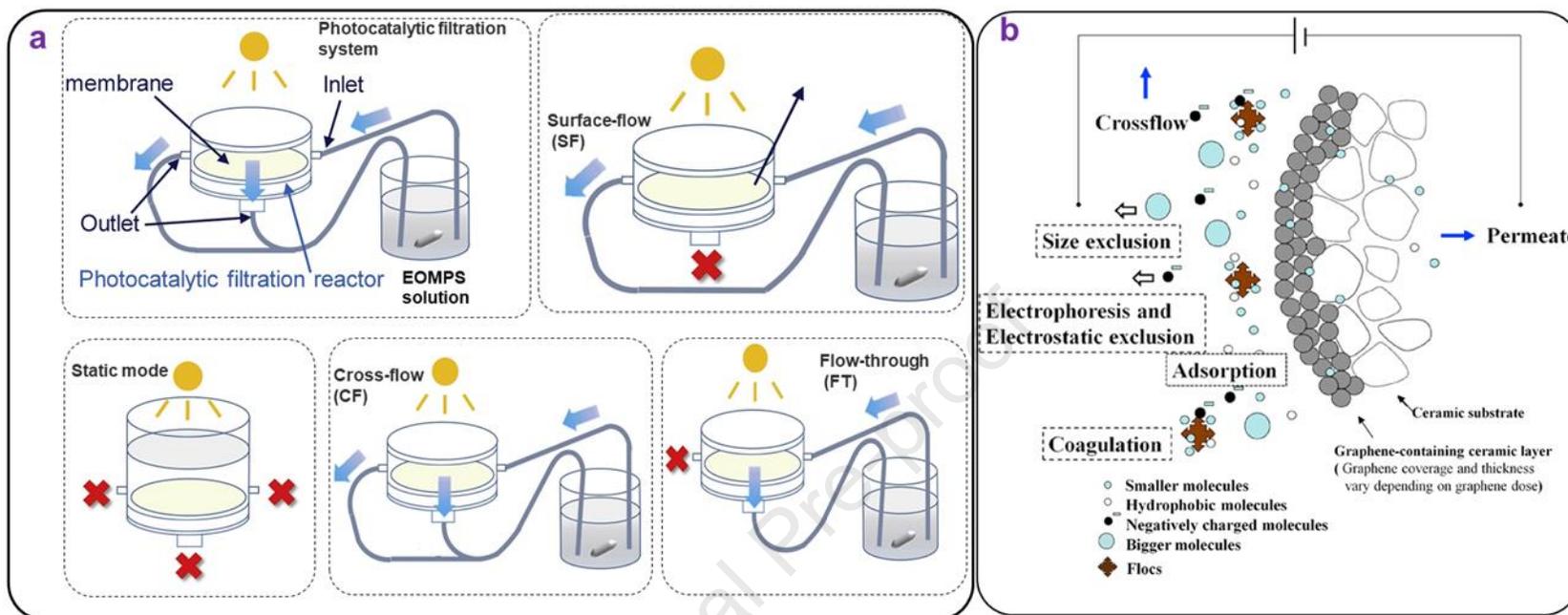


Fig.14

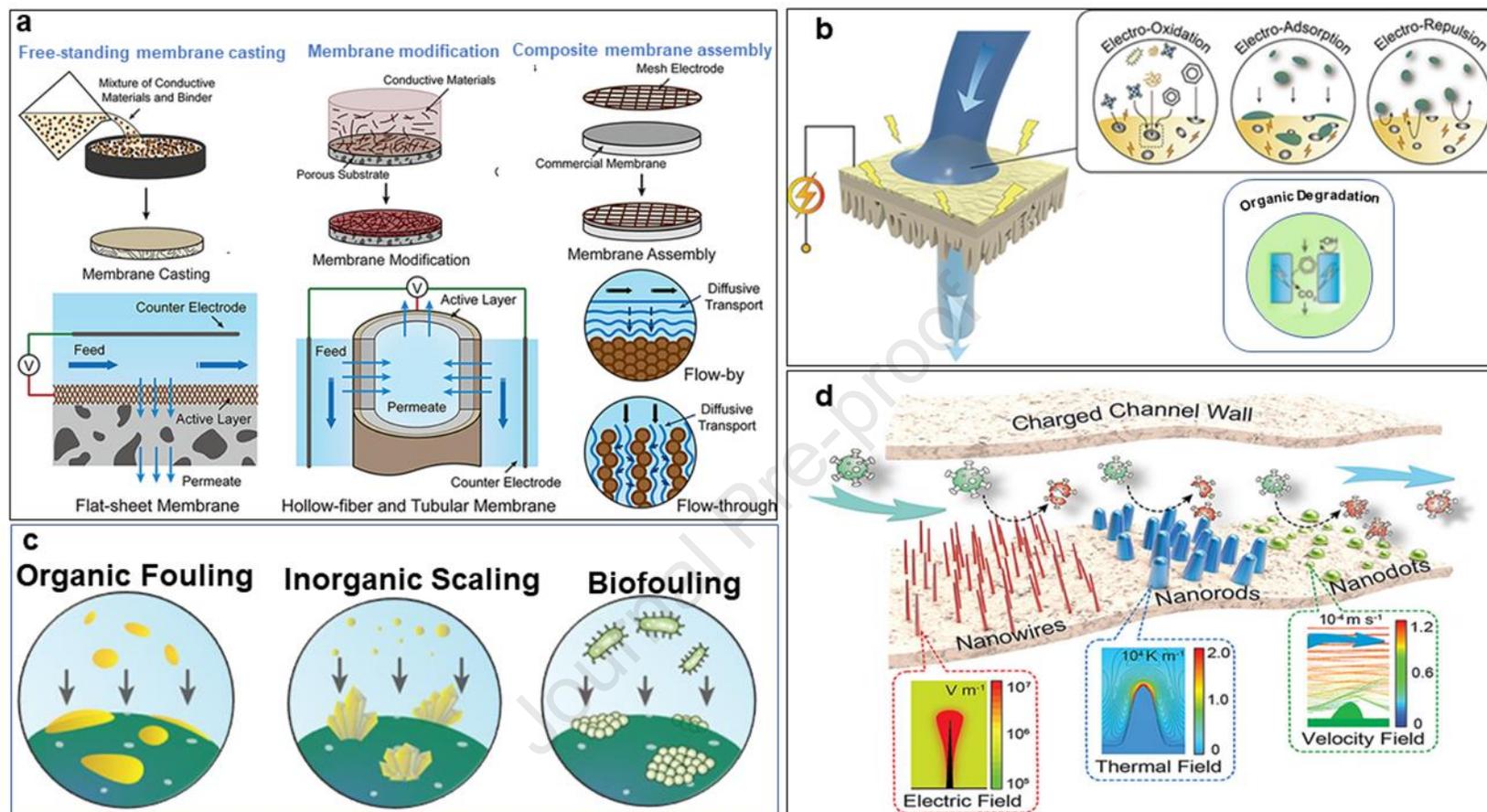


Fig.15

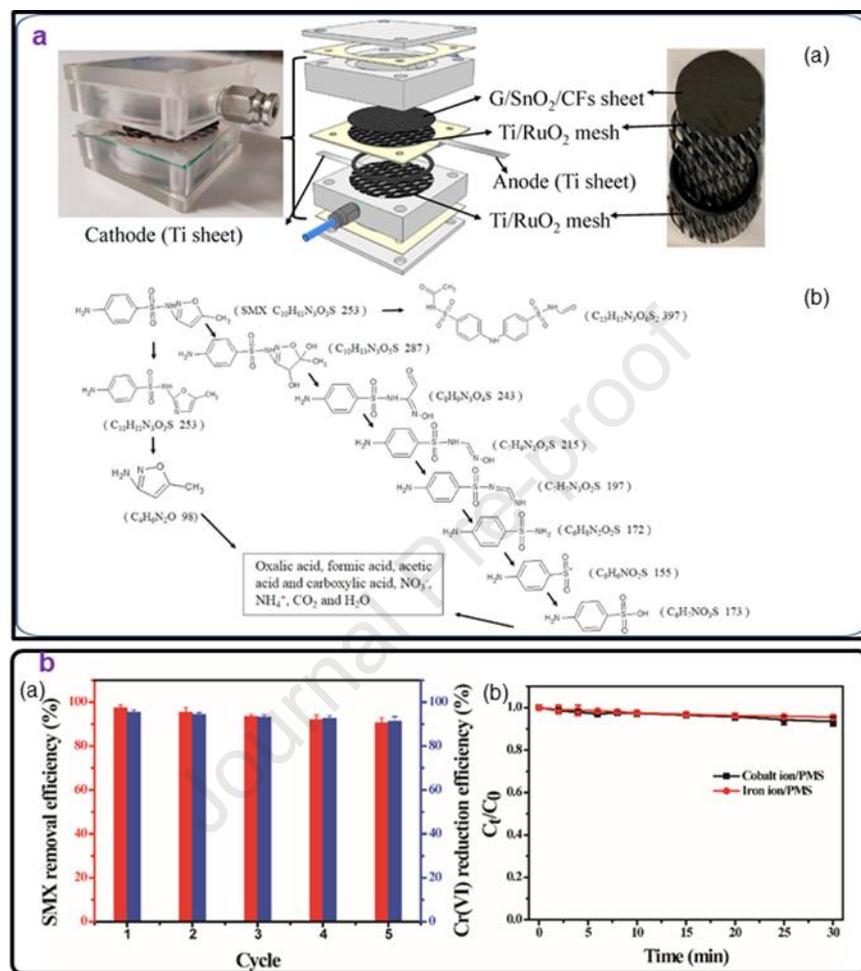


Fig.16

**Highlights**

- Next-Generation GO membranes showed promising removal of emerging organic micropollutants.
- GO membranes and substrates together act synergistically in water treatment.
- GO can easily accommodate adsorptive, degradative, biomimetic, and filtration membranes.
- Low price and multicycle operational GO membranes have scope for industrial application.
- Present status and future-oriented assessment of GO-based membrane technology are discussed

**Declaration of interests**

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.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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