Removal of organic micropollutants using advanced membrane-based water and wastewater treatment: A review

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Abstract

The rising consumption of pharmaceuticals, personal care products, and endocrine disruptive compounds for healthcare purposes and improving living standards has resulted in the widespread occurrence of organic micropollutants (MPs) in water and wastewater. Conventional water/wastewater treatment plants are faced with inherent limitations in tackling these compounds, leading to difficulties in the provision of secure and safe water supplies. In this context, membrane technology has been found to be a promising method for resolving this emerging concern. To ensure the suitability of membrane-based treatment processes in full-scale applications, we first need to develop a better understanding of the behavior of MPs and the mechanisms behind their removal using advanced membrane technologies. This review provides a thorough overview of the advanced membrane-based treatment methods available for the effective removal of MPs, including reverse osmosis, nanofiltration, ultrafiltration, forward osmosis, and membrane distillation.

Keywords: Membrane technologies, Organic micropollutants, Pharmaceuticals, Personal care products, Endocrine disruptive compounds.

1. Introduction

The ongoing and widespread occurrence of organic micropollutants (MPs) – including pharmaceutical active compounds (PhACs), personal care products (PCPs), and endocrine disrupting chemicals (EDCs) - presents a formidable challenge to the water industry [1]. MPs are generally present in water at trace concentrations ranging from ngL^{-1} to μgL^{-1}, however; with

increasing human population and higher reliance of modern societies on PhACs and PCPs, the release of organic MPs into the water bodies is foreseen to increase in the future [2]. Recently, there are growing concerns over the complexity of MPs in terms of their potential adverse effects on human health, especially upon chronic exposure via the water supply system [3,4]. The European Union (EU) has raised concerns about the increasing levels of organic MPs in water bodies and enforced strict regulations on their discharge. Similarly, the United States Environmental Protection Agency (USEPA) has listed several organic MPs on a Contaminant Candidate List to monitor their occurrence levels, routes of human exposure, and the potential health risks [5].

The removal of MPs using advanced membrane technology has, therefore, become a topic of significant interest in water/wastewater treatment and water reuse [6–10]. In reflection of such interest, several dedicated review papers [11–16] have overviewed the extensive efforts devoted to this issue. However, previous reviews have focused mostly on commercial reverse osmosis (RO) and nanofiltration (NF), and the coverage of new and emerging membrane technologies, such as forward osmosis (FO) and membrane distillation (MD), has been very limited. In particular, there has not yet been any critical review to date, which addresses the emerging membrane technologies specifically for the removal of organic MPs despite the importance and eminence of this topic.

This review aims to address this literature gap by providing an overview of the state-of-the-art membrane-based technologies and their application in the removal of MPs. The present review mainly highlights the removal of MPs via non-biological membrane-based treatment methods such as RO, NF, UF, FO, and MD rather than the conventional/membrane-based biological processes i.e., activated sludge or membrane bioreactors [17–19]. The review starts with a brief account of the occurrence of MPs, their pathways to the aquatic environment, and their adverse environmental and health impacts (Section 2). The removal of MPs from water/wastewater by different membrane technologies (RO and NF (Section 3.1), UF (Section 3.2), FO (Section 3.3), and MD (Section 3.4)) is then systematically evaluated. In particular, the roles of molecular properties, operational conditions, and membrane properties are critically assessed, and their underlining mechanisms are discussed. Some of the latest advances from recent literature (e.g., the effect of the functionalization and incorporation of nanomaterials in the polymeric membrane and the effect of organic, inorganic, and complex fouling on the removal of MPs) are also introduced. The current review provides a roadmap for further research by highlighting the factors that may influence process performance and demonstrating ways in which these processes can be improved.

2. MPs in the aquatic environment

2.1. Classification of MPs

MPs include a wide range of contaminants of emerging concern. They can be categorized into multiple groups such as PhACs, PCPs, pesticides, and industrial products according to their properties and utilization purposes. Based on these categorical arrangements, further classifications can be made. Some of the representative products of the above-defined categories

are mentioned below. Antibiotics, hormones, analgesics and anti-inflammatory drugs, antiepileptic drugs, cytostatic drugs, blood lipid regulators, contrast media, and β-blockers can be classified as pharmaceutical products, while antimicrobial agents/disinfectants, fragrances, insect repellants, detergents, preservatives, and sunscreen UV filters fall under personal care products [20,21]. Similarly, organochlorine insecticides, organophosphorus insecticides, herbicides, fungicides are representative pesticides, and plasticizers and fire retardants are major industrial products which are considered as MPs. To date, over 100,000 MPs have been utilized by humans and animals for health care reasons and lifestyle improvement [22]. The detailed classification of selected MPs groups, along with some of their representative products, are summarized in Table 1 [14,21,23,24].

Table. 1. Classification of MPs. Source: Modified from [14,21,23,24]

Groups	Sub-groups	Representative Compounds	MW ($gmol^{-1}$)	Molecular formula	Charge at pH 7	pKa	Log K_{ow}	Log D
PhACs	Antibiotics	Erythromycin	733.93	$C_{37}H_{67}NO_{13}$	Neutral	8.9	3.06	1.55
		Roxithromycin	837.05	$C_{41}H_{76}N_2O_{15}$	Neutral	9	2.7	
		Ofloxacin	361.36	$C_{18}H_{20}FN_3O_4$	-	5.8	-2	-0.25
		Sulfamethoxazole	253.3	$C_{10}H_{11}N_3O_3S$	-	1.7;5.6	0.89	0.45
	Analgesic and anti-inflammatory drugs	Acetaminophen	151	$C_8H_9NO_2$	Neutral	9.5	0.46	0.23
		Ibuprofen	206.29	$C_{13}H_{18}O_2$	-	4.47	3.97	1.44
		Naproxen	230	$C_{14}H_{14}O_3$	-	4.2	3.18	0.34
		Mefenamic acid	241.285	$C_{15}H_{15}NO_2$	-	3.8	5.12	2.04
		Fenoprofen	242	$C_{15}H_{14}O_3$	-	4.21	3.9	0.38
		Ketoprofen	254.28	$C_{16}H_{14}O_3$	-	4.29	3.12	0.41
		Indometacin	357.78	$C_{19}H_{16}ClNO_4$	-	3.8	4.23	0.75
		Diclofenac	296.15	$C_{14}H_{11}C_{12}NO_2$	-	4.08	4.51	1.59
	Antiepileptic drugs	Primidone	218	$C_{12}H_{14}N_2O_2$	-	P1=-1, P2=12.2	0.91	0.83
		Carbamazepine	236.27	$C_{15}H_{12}N_2O$	Neutral	13	2.45	2.58
	Blood lipid regulators	Clofibric acid	214.65	$C_{10}H_{11}ClO_3$	-	3.35	2.57	1.08
		Gemifibrozil	250.34	$C_{15}H_{22}O_3$	-	4.45	4.77	2.22
		Bezafibrate	361.82	$C_{19}H_{20}ClNO_4$	-	3.44	4.25	0.69
		Pravastatin	24.53	$C_{23}H_{36}O_7$	-	4.2	3.1	-1.21
	β-blockers	Propranolol	259.34	$C_{16}H_{21}NO_2$	Neutral	9.6	3.48	1.15
		Metoprolol	276.37	$C_{15}H_{25}NO_3$	+	9.49	1.88	-0.61
	Contrast media	Iopromide	790.0	$C_{18}H_{24}I_3N_3O_8$		P1=2 P2=13	-2.10	-
		Iopamidol	777.1	$C_{17}H_{22}I_3N_3O_8$		10.7	-2.42	-
		Iohexol	821.1	$C_{19}H_{26}I_3N_3O_9$		11.7	-3.05	-
	Hormones	Estrone	270.36	$C_{18}H_{22}O_2$	Neutral	10.3	3.13	- 3.6 at pH9
		17B-estradiol	272.38	$C_{18}H_{24}O_2$	Neutral	10.4	4.01	4.12at pH9
		17α-ethinyl estradiol	296.4	$C_{20}H_{24}O_2$	Neutral	10.3	3.9	-
		Estriol	288	$C_{18}H_{24}O_3$	Neutral		2.45	-
	Cytostatic drugs	Cyclophosphamide	260	$C_7H_{15}Cl_2N_2O_2P$		0.5	0.97	- -

PCPs	Anti-microbial agents/ Disinfectants	Triclosan	289.6	$C_{12}H_7Cl_3O_2$	Neutral	7.8	5.34	5.28
		Triclocarban	315.6	$C_{13}H_9C_{l3}N_2O$	Neutral	11.4	4.90	-
	Preservatives	Propyl-paraben	180.2	$C_{10}H_{12}O_3$	Neutral	8.5	3.04	-
		Methyl-paraben	152.15	$C_8H_8O_3$	Neutral			1.86 at pH6
	Insect repellent	N,N-diethyl-m-toluamide	191.3	$C_{12}H_{17}NO$		< 2	2.18	-
	Sunscreens	Oxybenzone	228	$C_{14}H_{12}O_3$			3.79	-
Pesticides	Herbicides	Atrazine	215.68	$C_8H_{14}C_lN_5$	Neutral	1.7	2.6	-
		Diuron	233.1	$C_9H_{10}C_{l2}N_2O$	Neutral	2.68		
	Insecticides	Diazinon	304.35	$C_{12}H_{21}N_2O_3PS$		2.6	3.8	
	Fungicides	Clotrimazole	344.84	$C_{22}H_{17}C_lN_2$				
		Tebuconazole	307.82	$C_{16}H_{22}C_lN_3O$				
Industrial Chemicals	Plasticizers	Bisphenol A	228.29	$C_{15}H_{16}O_2$		9.6	3.32	
		DBP	278.34	$C_{16}H_{22}O_4$				
		DEHP	390.564	$C_{24}H_{38}O_4$				
		DMP	194.184	$C_{10}H_{10}O_4$				
	Fire Retardants	Tri(2-chloroethyl) phosphate	250.187	$C_9H_{15}O_6P$			1.44	
		Tri(chloropropyl) phosphate	327.57	$C_9H_{18}C_{l3}O_4P$			2.59	

2.2. Pathway of MPs to the environment

MPs can enter the environment through various pathways, including domestic wastewater, untreated/treated effluent discharged from wastewater treatment facilities and processing industries, agriculture and farmyard runoff mixing with fresh/surface water, and manure/biomass sludge applications [25,26]. Among these, wastewaters from hospitals, domestic residences, and manufacturing industries are considered as a major point source of the MPs which have trickled into the environment [21]. Some of the PhACs are not readily and completely metabolized by humans and/or animals and are excreted via urine and feces [27,28]. In the case of many MPs, their metabolites and byproducts are poorly removed by conventional treatment methods [29–36]. Such treated/untreated effluents are discharged into the freshwater bodies (i.e., lakes, rivers, and coastal water) to be reused for irrigation, horticulture, and other non-potable purposes, and in this process, the occurrence of MPs have become steadily increased from parts-per-trillion (ngL^{-1}) to parts-per-billion (μgL^{-1}) to result in the deterioration of soil, surface water, and groundwater qualities [37–39]. Other pathways through which water bodies are exposed to MPs may include swimming and recreational activities, disposal of unused-medicines, and veterinary medicine runoff from farmyards, which end up mixing with freshwater bodies [40,41]. The typical pathways of MPs in water and wastewater through identified potential sites are shown in Figure 1.

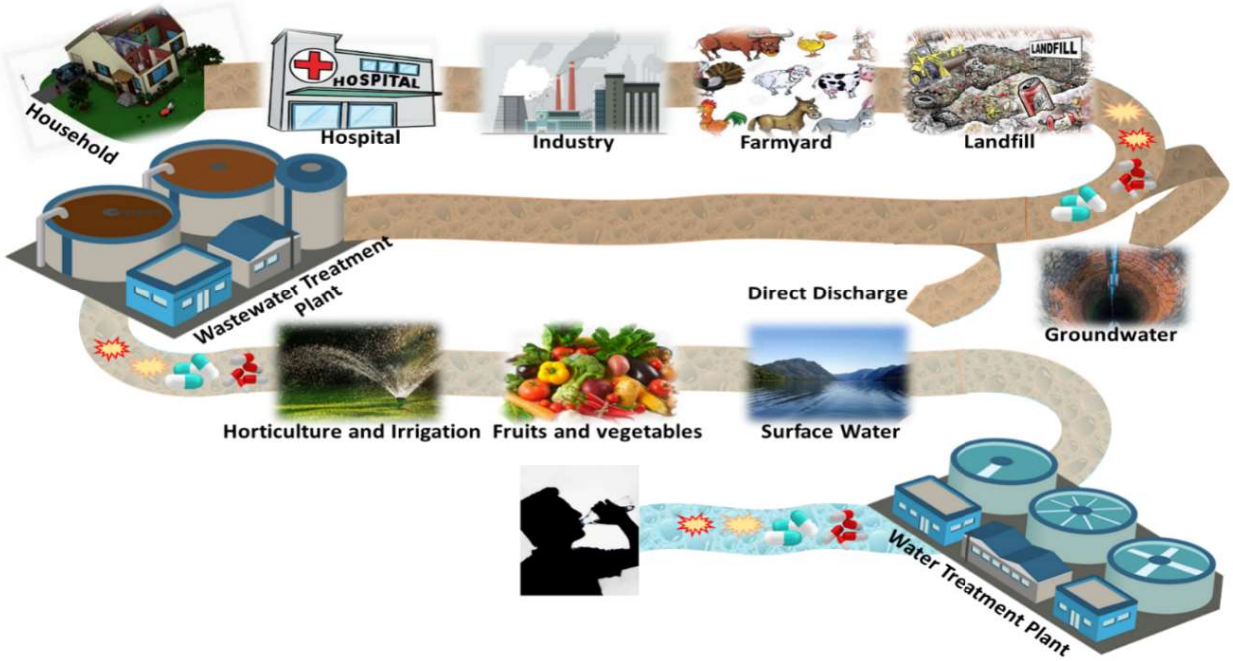

Figure 1. Potential sources and pathways of MPs. Source: Modified from [13,41].

2.3. Effects of MPs on human health and the ecosystem

The ubiquitous occurrence of MPs in freshwater bodies is of rising concern due to their potential adverse effects on human health and the environment. A study conducted on post-mortem brain material obtained from 24 individuals (12 obese and 12 under-weight with a body mass index >30 and <25 kg/m^2, respectively) showed the accumulation of bisphenol A, triclosan, triclocarban, methyl-paraben, ethyl-paraben, n-propyl-paraben, and benzyl-paraben in the hypothalamus, while bisphenol A, benzophenone-3, triclocarban, methyl-paraben, and n-propyl-paraben were detected in white-matter brain tissues [42]. In a separate study, an environmental working group in the U.S. conducted a survey on 20 teenage girls, from 14 to 19 years in age, and observed the accumulation of 16 hazardous chemicals including triclosan, synthetic musk, and 2-benzenedicarboxylic salt related to the use of cosmetic products [41,43].

In addition, the studies have revealed that the MPs, in particular, EDCs have the ability to modulate endocrine functioning by damaging the normal physiological reactions related to the male and female reproductive system (i.e., menstrual cycle irregularities, impaired fertility, endometriosis, polycysticovarian syndrome, spontaneous abortion, and alteration of hormone concentration) [44–46]. Desai et al. [47] elucidated the role of EDCs in metabolic disorders such as obesity, insulin resistance, type2 diabetes, hepatic injury, dyslipidemia, and cardiovascular diseases in humans. Giulivo et al. [48] also described the potential role of EDCs (i.e., bisphenol A, parabens, and phthalates) on the pathogenesis of breast cancer even at very low concentrations. The effect of acute and chronic exposure on the histopathological changes, reproductive system, and body organs of birds, fishes, mud snails, and mammals has also been reported elsewhere [49–53].

3. Removal of MPs from water and wastewater

Numerous studies have revealed that existing conventional water and wastewater treatment facilities are unable to achieve adequate removal of MPs [54–64]. To ensure the appropriate subtraction of MPs and to assess the performance of treatment systems for their complete exclusion from the treated effluents, both engineers and environmental scientists need to understand the mechanism for removing MPs to design a more specific and appropriate system. Many authors reported on the improved performance in MPs removal when using advanced treatment methods, including activated carbon (AC), ultraviolet-radiation (UV), ozonation (O_3)/advanced oxidation process (AOP), and membrane filtration [63–67]. Table 2 summarizes the estimated performance anticipated by the different processes for PhACs, EDCs, and PCPs removal from water and wastewater based on existing studies conducted on specific classes of compounds or compounds which are similar to trace pollutants. Among membrane processes, RO, NF, and ultra-filtration (UF) were found to be promising for the complete and near-complete removal of a variety of MPs from water/wastewater [12,68–70]. In addition, FO and MD have also gained significant attention from researchers as potential candidates for future implementation due to their low operating cost and high-quality performance (Table 2) [71–75].

Table 2. Removal performance of different membrane and non-membrane-based treatment processes. Source: Modified from [16,76,77]

Pollutants		Removal Performance of Non-Membrane Based Process						Removal Performance of Membrane Based Process					
Groups	**Classification**	**AC**	**BAC**	**O_3/ AOPs**	**UV**	**Cl_2/ ClO_2**	**Coag/ Floc**	**FO**	**RO**	**NF**	**UF**	**MD**	**Degradation (B/P/AS)**
EDCs	Pesticides	E	E	L-E	E	P-E	P	F-E	E	G	P-F	G-E	E {P}
	Industrial Chemicals	E	E	F-G	E	P	P-L	F-E	E	G	P-F	F-E	G-E {B}
	Steroids	E	E	E	E	E	P	F-E	E	G	P-F	G-E	L-E {B}
	Metals	G	G	P	P	P	P-G	F-E	E	G	P-F	G-E	P {B}, E {AS}
	Inorganics	P-L	F	P	P	P	P	F-E	E	G	P-F	E	P-L
PhACs	Antibiotics	F-G	E	L-E	F-G	P-G	P-L	F-E	E	E	P-F	L-E	E {B} G-E {P}
	Antidepressants	G-E	G-E	L-E	F-G	P-F	P-L	F-E	E	G-E	P-F	G-E	G-E
	Anti-inflammatories	E	G-E	E	E	P-F	P	F-E	E	G-E	P-F	F-E	E {B}
	Lipid regulators	E	E	E	F-G	P-F	P	F-E	E	G-E	P-F	E	P {B}
	X-Ray Contrast Media	G-E	G-E	L-E	F-G	P-F	P-L	F-E	E	G-E	P-F	-	E {B and P}
	Psychiatric Control	G-E	G-E	L-E	F-G	P-F	P-L	F-E	E	G-E	P-F	E	G-E
PCPs	Synthetic Scents	G-E	G-E	L-E	E	P-F	P-L	F-E	E	G-E	P-F	-	E {B}
	Sunscreens	G-E	G-E	L-E	F-G	P-F	P-L	F-E	E	G-E	P-F	F-E	G-E
	Anti-microbials	G-E	G-E	L-E	F-G	P-F	P-L	F-E	E	G-E	P-F	G-E	F {P}
	Surfactants/ Detergents	E	E	F-G	F-G	P	P-L	F-E	E	E	P-F	E	L-E {B}

BAC = biological activated carbon; B = biodegradation; P = photodegradation (solar); AS = activated sludge.
E = excellent; > 90%, G = good; 70-90%, F = fair; 40-70%, L = low; 20-40%, P = poor; < 20%.

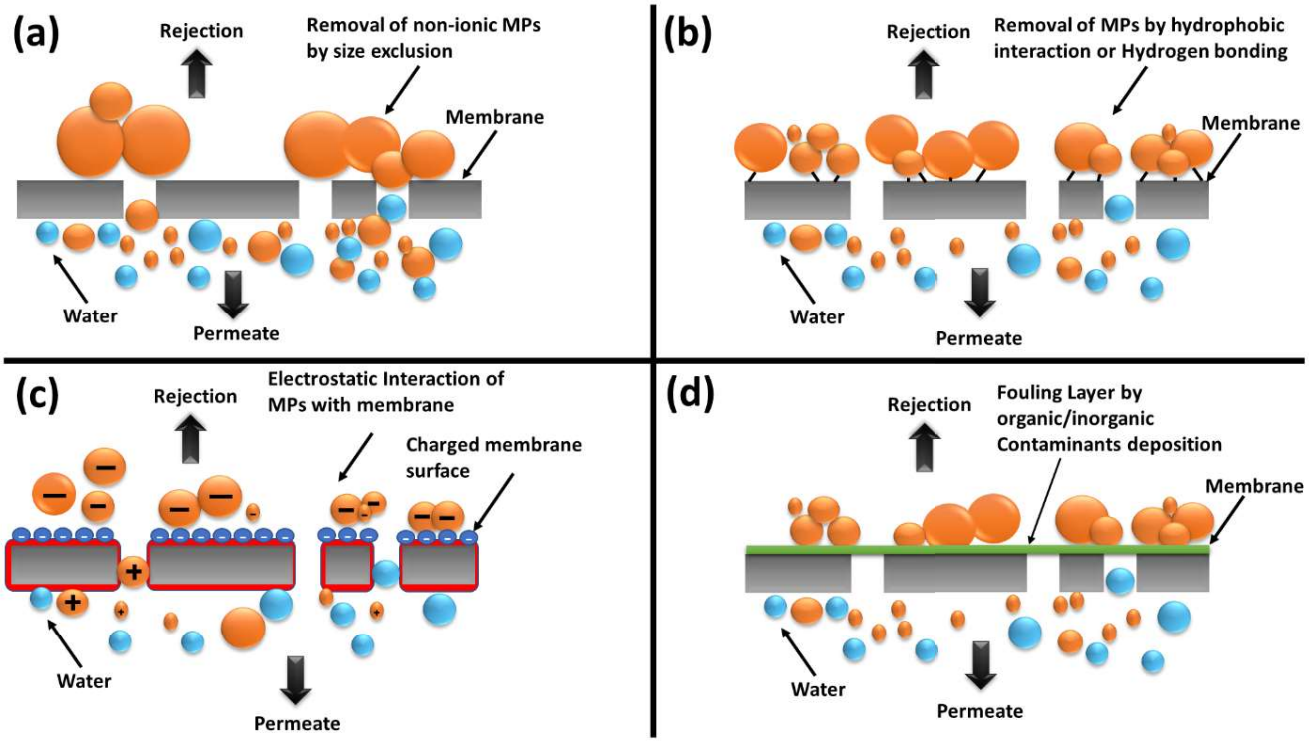

Figure 2. Mechanism of MPs removal from membrane; (a) Size exclusion; (b) Hydrophobic interaction; (c) Electrostatic interaction; (d) Adsorption. Source: Modified from [78]

3.1. MPs Removal by RO and NF

3.1.1. Influence of MPs characteristics on their rejection

RO and NF are pressure-driven processes and are more energy-intensive compared with other pressure driven membrane-based treatment systems such as MF and UF [13,79]. Despite their high pressure requirement, the use of RO or NF for water and wastewater treatment and desalination has been increasing steadily [13,80]. In addition, the use of NF/RO for tertiary treatments at wastewater/sewage treatment facility is also being encouraged due to the high purity of the NF/RO-treated effluents [81]. However, for NF/RO processes to obtain the effective removal of MPs, which have different physicochemical characteristics (i.e., size, charge, solubility, diffusivity, and hydrophobicity), it is imperative to understand the fundamental mechanisms involved (i.e., electrostatic interaction, size exclusion, and hydrophobic interaction) (Figure 2) [78,82]. Among the studies conducted in this regard, Licona and his co-workers observed a strong relationship between molecular weight (MW) and hydrophobicity in the rejection of MPs [83], identifying size exclusion and adsorption as the dominant mechanisms for their removal. Also, the electrostatic repulsion between the MPs and the negatively-charged membrane surface helped to remove negatively-charged MPs such as ibuprofen, dipyrone, and diclofenac more effectively compared to those which were neutrally-charged (i.e., acetaminophen and caffeine). Similar findings were observed by Albergamo et al. [84], who reported a strong reverse correlation

between the size and passage of neutral-hydrophilic and anionic MPs. This correlation was weaker for the moderately hydrophobic MPs. The author attributed this low removal to the affinity between hydrophobic moieties such as aromatic rings and hydrocarbon chains and the active layer of RO membranes, as illustrated in Figure 3a.

In the case of some MPs, size exclusion works as their main removal mechanism. For instance, the nonionic structure of bisphenol A for the selected pH led to its low removal (74.1%) compared to ibuprofen (98.1%) and salicylic acid (97%) [85]. Although, the MW of bisphenol A (MW=228 $gmole^{-1}$) is higher than ibuprofen (MW=206 $gmole^{-1}$) and salicylic acid (MW=138 $gmole^{-1}$), bisphenol A's high pKa value (pKa=9.6-10.2) conferred to an insignificant contribution of the electrostatic interaction by the negatively-charged membrane surface and the absence of electrostatic contribution as compared with ibuprofen (pKa=4.9) and salicylic acid (pKa=2.9) which have a deprotonated/negatively-charged appearance. Therefore, unlike ibuprofen and salicylic acid, the dominant mechanism for bisphenol A removal was only the size exclusion. Similar findings were observed by Reznik et al. [86], who reported size exclusion as the dominant mechanism for hydrophilic neutral compounds with high water partitioning coefficients. In the case of the positively-charged MPs, the removal efficiency is considerably decreased due to their electrostatic interaction with the negatively-charged membrane surface and subsequent diffusion.

Adding to the chemical speciation administered by the ionic structure, pKa, and K_{ow} values, the rejection of MPs is also substantially influenced by their associated functional groups [87]. In the absence of electrostatic mechanisms (i.e., attraction/repulsion), the characteristics of MPs and other compounds may also play a prime role in their removal by the membrane. A different rejection behavior for selected PhACs (carbamazepine, ibuprofen, and sulfamethoxazole) was observed by NF membranes (i.e., NF-90 and NF-270; Filmtech) due to their different physicochemical nature [88]. A relatively constant rejection by both the NF membranes was detected for carbamazepine (pKa=2.3), as size exclusion was the dominant mechanism for the nonionic/uncharged compounds. However, the rejection of (uncharged) sulfamethoxazole by loose structure NF-270 membrane was significantly lower, despite its high MW, instigated by the presence of two functional moieties at both sulfonamide linkage sides. Likewise, >85% removal was recorded by RO membranes for both estrogenic hormones, i.e., estrone and 17-β-estradiol containing 17-keto and 17-hydroxyl groups, respectively [89]. The reason for their high rejection is presumably the occurrence of hydrogen bonding between the polyamide membrane surface and the 3-oxygen atom in the first ring of estrone and 17-β-estradiol [89]. Similarly, molecules with high dipole moment (polarity) could easily diffuse into membrane pores; therefore, despite having similar MW, the removal of high polar molecules was significantly lower compared to low/non-polar compounds [90]. These findings illustrate the significant role of the compound's dipole moment, which alters the molecule orientation as the compound approaches the membrane pores, in the removal via membranes. Although the removal performance of RO membranes for MPs is likely to be high in contrast with NF due to their smaller MWCO value and denser polymeric

surface, the use of NF is also attractive as it has comparable performance, a high flux, and a low-pressure requirement.

3.1.2. Effects of operating conditions on MPs rejection

Adding to the significant influence of MPs characteristics (i.e., MW, hydrophobicity, K_{ow} value, associated functionalities, and dipole moment) on their removal, the change in the chemical (i.e., presence of organic and inorganic matters, feed solution pH, etc.) and physical (temperature, pressure, cross-flow velocity, etc.) operating conditions may also significantly affect the rejection of MPs. The presence of specific inorganic ions, such as Na^+, Ca^{2+}, Mg^{2+}, SO_4^{2-}, can significantly affect the membrane rejection behavior due to their specific interactions with MPs, their ability to modify membrane properties, and their impact on the solution ionic strength [91]. Higher concentrations of inorganic ions (particularly divalent and multivalent ions) result in higher ionic strength, and thus reduced Debye length, as a result of electric double layer compression [92]. Consequently, the effect of charge interaction will be greatly weakened. Divalent ions, including Ca^{2+} and Mg^{2+}, also have a strong tendency to bind to the carboxyl groups of polyamide RO and NF membranes, which leads to charge neutralization or even charge reversal of membrane surfaces [91,93–96].

In their investigation of the removal of a series of halogenated acetic acids, Yang et al. [91] reported that increasing Ca^{2+} concentration could greatly reduce the rejection of these negatively-charged disinfection by-products by a loose NF270 membrane, as a result of the neutralization of negative surface charge of the membrane by Ca^{2+}. Nevertheless, their rejection by NF90 was not significantly affected by charge neutralization, which is explained by the greater dominance of the size exclusion effect by the tight NF membrane. Changes in ionic strength and ion-membrane specific interaction can also potentially alter the structure of a membrane, including its pore structure, and thereby affect the membrane transport properties [97]. Likewise, the interaction of inorganic ions with MPs can change physical properties (e.g., physical size by forming dimers and aggregates [98] or solubility and hydrophobicity [99]). In addition to the above-mentioned direct effects, the presence of inorganic ions can also indirectly affect membrane rejection by influencing membrane fouling [100]. For example, Ca^{2+} can accelerate membrane fouling by humic acid to form a thicker, denser, but less negatively-charged foulant cake layer [101,102], leading to a loss of solute rejection as a result of enhanced concentration polarization in this cake layer [100]. In contrast, the mild fouling in the absence of Ca^{2+} was found to enhance solute rejection, possibly due to the sealing of membrane defects and additional charge repulsion by the negatively-charged humic acid macromolecules [101].

Similar to monovalent and divalent ions, the effect of silica particles, which are abundant in natural water, have been evaluated on MPs rejection. The removal of 17-β-estradiol and progestogen hormones by RO membranes (LFC-1, Hydranautics, Oceanside, CA) was reported to be affected severely in the presence of silica particles during the initial 40 hr followed by a moderate decline, whereas a linear declining trend was observed in the absence of silica [103]. The possible reason

was presumably the negative effect of the silica fouling layer formation on the back diffusion of the compound after their diffusion through the polymeric membrane, hence resulting in poor rejection. For tightly bound NF90 and XLE RO membranes (Filmtec), electrostatic repulsion and size exclusion worked synergistically and resulted in the improved rejection of MPs (i.e., carbamazepine, triclosan, ibuprofen, sulfamethoxazole, sulfadiazine, and sulfamethazine) after silica fouling. However, destructive performance was observed in the case of a loose NF270 membrane [104], presumably due to the additional steric barrier by silica fouling, accompanied by the cake-enhanced concentration polarization, subsequently decreasing the rejection performance.

Like the inorganic contaminants, the solute-solute interaction between macro organic molecules and MPs was found to have a substantial effect on MPs rejection by NF membranes (NF90 and NF270) [105]. As illustrated in Figure 3b, the presence of the organic macro-molecules had an influence over the removal of several PhACs which could be attributed to (i) the association of organic macromolecules with PhACs; (ii) the modification of membrane surface by organic fouling; and (iii) the steric hindrance and/or electrostatic interaction due to the negatively-charged membrane surface [105,106]. A study examined the influence of organic, biological, and colloidal fouling and their complex over the rejection of six MPs (i.e., carbamazepine, ibuprofen, sulfadiazine, sulfamethoxazole, sulfamethazine, and triclosan) using commercial membranes (NF-90 and NF-270). The results revealed that the removal performance of the NF-90 membrane was improved for all fouling conditions and ascribed this high performance to the cumulative effect of steric hindrance and electrostatic repulsion. However, for the NF-270 membrane, the rejection of MPs was notably decreased by all fouling mechanisms due to the cake-enhanced concentration polarization effects [107].

Dolar et al. also investigated the effect of water matrixes (Milli-Q water, model water, tap water, and real pharmaceutical wastewater) on the removal of five veterinary pharmaceuticals (i.e., sulfamethoxazole, trimethoprim, ciprofloxacin, dexamethasone, and febantel) using four different NF (NF90, NF270, NF (Dow Filmtech) and HL (Desal, Osmonics, GE Infrastructure Water Process Tech., Vista, CA)) membranes and two RO membranes (LFC-1 (Hydranautics, Oceanside, CA) and XLE (Dow Filmtec, Midland, MI)). In general, the rejection of the selected compounds was increased with the complexity of the water matrix from XLE, LFC, and NF90 membranes. However, a reverse trend was observed from the loose NF membranes (i.e., NF270, NF, and HL). The authors attributed this deteriorating performance to the bigger pore size of the NF membranes, plugging of the tight network pores and their disappearance during fouling, and enlargement of the wider aggregate pores [108].

Xu et al. also examined the influence of multi-influent matrices on the removal of six MPs using a DF30 NF membrane (Beijing Origin Water Technology Co., Ltd. China). The presence of inorganic ions in the feed resulted in an improved rejection of neutral (carbamazepine and chloramphenicol) and positively charged (metoprolol and trimethoprim) MPs. However, a declined rejection was observed for negatively-charged compounds (diclofenac sodium and indomethacin). This biased phenomenon mainly occurred due to the change in membrane surface

by the deposition of divalent cations (Ca^{2+} and Mg^{2+}), which consequently weakened the electrostatic attraction and repulsion between the positively and negatively charged MPs and negatively-charged membrane, respectively [109]. On the other hand, the improved removal of neutral MPs (carbamazepine and chloramphenicol) was mainly attributed to the enhanced sieving effect caused by the deposition of divalent cations over the membrane. Similarly, the addition of organic matter (15 mgL^{-1} of HA) resulted in the improved rejection of positively-charged metoprolol and trimethoprim, however, no significant change was observed for anionic and non-ionic compounds. This was presumably due to the electrostatic interaction between the negatively-charged HA and positively-charged MPs, since no change was observed in the rejection of six MPs after the addition of SA in the same amount. In addition, no fouling formation occurred after HA, SA addition as validated from the stable flux value. These findings suggested that the species-dependent electrostatic effect was the primary reason behind the improved rejection of MPs [110]. In addition to the influence of organic/inorganic fouling on MPs rejection, the effects of MPs on NF (NF90) and RO (DOW 1812-50) membrane fouling were investigated during the filtration of synthetic and real wastewater spiked with three PhACs (i.e., ibuprofen, carbamazepine, and sulfamethoxazole). It was observed that the presence of PhACs mitigated membrane fouling and led to a smaller decrease in flux and salt rejection [111].

Since the formation of a fouling layer changes surface properties of the membrane, affecting rejection performance, chemical cleaning is used to restore membrane permeability once fouling has become excessive. However, chemical cleaning can have a considerable impact on MP rejection. A discernible decrease in MPs rejection can often be observed immediately after caustic cleaning [112–117]. Simon et al., [113,114] ascribe this observation to conformational change within the polymeric matrix of the membrane active layer due to exposure to the caustic cleaning solution. Under this highly caustic condition (pH >11), electrostatic repulsion between carboxylic functional groups of the polyamide layer could enlarge the membrane pore size, resulting in the observed decrease in MP rejection. The effect is hysteresis. In other words, it is not permanent, and MP removal efficiency is gradually restored over time. In fact, several studies have demonstrated that the negative impact of chemical cleaning can be minimized by applying acidic cleaning immediately after caustic cleaning [114,115]. Since chemicals used for membrane cleaning are often prescribed by membrane manufacturers, permanent membrane damage beyond normal wear and tear is unlikely even with repetitive chemical cleaning as long as the recommended chemical cleaning procedure of the manufacturer is followed [117,118]. Nevertheless, the hysteretic impact of chemical cleaning on MP rejection becomes more severe as the frequency and cleaning temperature increase. Data from Kallioinen et al. suggest that the membrane could be permanently damaged when the cleaning temperature is increased to 70 ^{0}C [117].

The influence of pH variation on MPs rejection has also been investigated (Figure 3c). Soriano et al. observed deteriorating performance from an NF270 membrane against perfluorocarboxylic acid at low solution pH (i.e., pH=3.1-4.4) and attributed this to low electrostatic repulsion between the

loose NF membrane and MP [119]. In contrast, higher rejections were observed from NF90, XLE, BW30, and SW30XLE membranes due to their low MWCO value, which demonstrate size exclusion as being the dominant mechanism. A study on the MPs rejection of NF and XLE membranes with and without silica fouling in varied pH conditions revealed that pH level had marginal influence over the rejection of both hydrophobic and hydrophilic compounds by NF90 and an insignificant effect on XLE membrane while, in contrast, a significant impact on NF270 membrane performance was observed particularly for low pH conditions (pH=3-8). Since electrostatic repulsion was the dominant mechanism for the NF270 membrane, the low removal of MPs was ascribed to the change in the membrane (as more amide and carboxyl groups on the membrane surface were dissociated as the pH increased) and MPs charge at low pH value. Although the formation of a silica fouling layer could enhance the removal performance of loose NF270, its effect was overwhelmed by the accompanied cake-enhanced concentration polarization phenomenon which impeded back diffusion of MPs into the feed solution, causing them to become trapped and accumulate on the membrane surface, so as to increase their diffusion across the membrane [104]. Similarly, the pH dependence speciation of estrone (pKa=10.4 approximately) resulted in a declined rejection of a TFC SR2 membrane at elevated pH, which was attributed to the decreasing-adsorption/increasing-repulsion effect with increasing membrane surface charge values (negative) at high pH (>pKa) [120]. The change in pH value did not affect the membrane property as verified by unaffected flux during the entire examined pH range; however, the dramatically decreasing rejection was solely due to the dominant size exclusion mechanism for estrone removal. From these findings, it was concluded that the less zeta potential value (i.e., +5 to −5 < pH 4 > −5 to – 22 mV) could ultimately result in high adsorption of estrone over the membrane surface, arbitrated by hydrogen bonding between the membrane and carbonyl and/or hydroxyl groups of estrone. Since adsorption curtailed at high pH and size exclusion were the prevalent mechanisms, the rejection would be affected by an upsurge in electrostatic repulsion [120].

Similar to the chemical conditions, the understanding of physical operating variables is also of great importance for the design and operation of NF/RO processes, as well as have significantly influence MPs removal. In general, an increase in cross-flow velocity (CFV) in the RO process results in an increased removal performance by affecting the concentration polarization mechanism occurring at the solution-membrane interface. However, CFV was observed to have an insignificant effect on estrone removal for the examined CFV range (0.073 - 0.24 m/s) [89]. This was presumably due to the higher estrone concentration within the membrane (XN-40; Trisep Corporation, Goleta, USA) compared to the polarization layer, hence depicting the minimal effect of concentration polarization. Generally, the rejection of the solute likely increases with increasing operating pressure, however, a 15% decline was observed for estrone when the pressure value was increased for the selected operating range (10 - 25 bar) [89]. A possible reason for this deteriorating performance could be the strong interaction of organic pollutants with the membrane polymer [121,122]. The solute membrane interaction could be altered by friction and diffusion, which are governed by hydrodynamic conditions and chemical concentration gradient, respectively. Since

the average pore radius of XN-40 membrane was of the same magnitude as the molecular size of estrone (0.7nm) [122], the increase in operating pressure resulted in decreasing adsorption rate due to the lower residence of estrone onto the membrane surface, to decrease its rejection and increase its concentration in the permeate side [89].

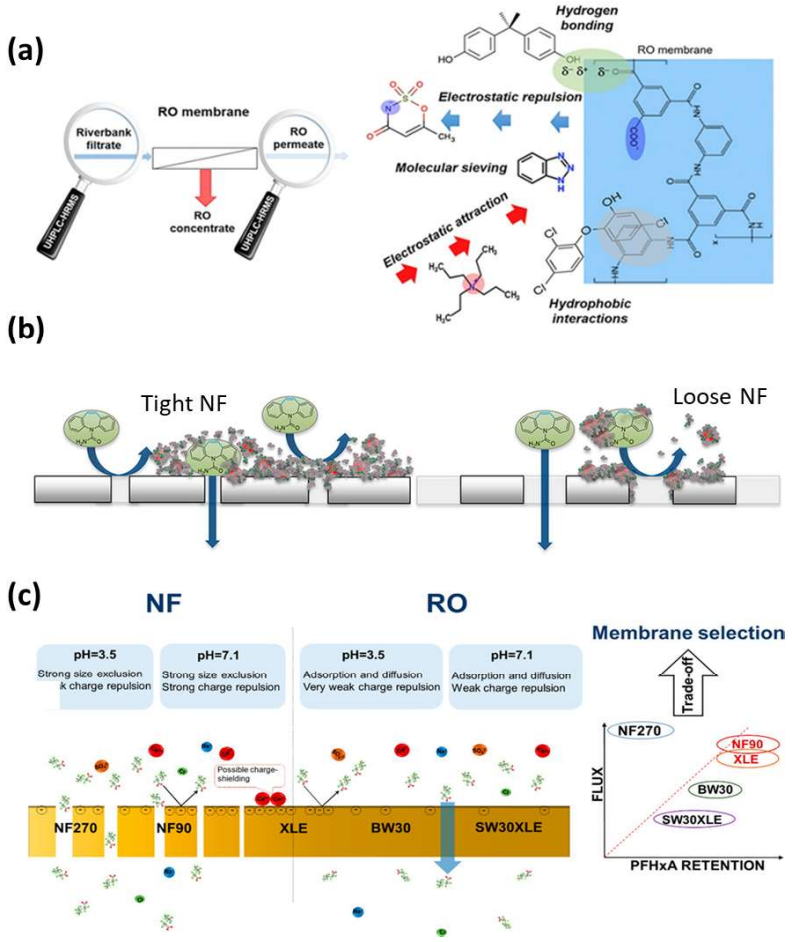

Figure 3. Effect of MPs characteristics and feed solution chemistry on their removal from NF/RO membranes: (a) Hydrogen-bonding/non-polar interaction of MPs with the membrane [84]; (b) Effect of fouling layer formation on the removal of MPs [105]; (c) Effect of the feed solution pH on removal performance (left) and removal efficiency of different membranes as a function of flux (right) [119]. Reprinted with the copyright permission.

3.1.3. Effects of membrane properties on MPs rejection

Polyamide thin film composite (TFC) membranes with a typical three-layer structure are considered as the most successful and commercialized membranes. It is comprised of a thin composite active layer (in the order of 100 nm thickness for RO and <100 nm for NF) attached with a more open intermediate layer (about 40 μm) and an even more open support layer. The possible reason for rejection performance deterioration by the membrane might be the strong interaction of organic pollutants with the membrane polymer [122,123]. Molecular docking was

performed by Lie et al. [124] between the PA layer and seven PhACs (propranolol, sulfamethoxazole, primidone, carbamazepine, atenolol, metoprolol, and trimethoprim) to investigate the effect of membrane charge characteristics on solute-membrane interaction (i.e., л-л stacking interaction, hydrogen bonding, л-cation interaction, and ionic bridge binding) by employing protonated/deprotonated states. They concluded that various specific and non-specific interactions were found to exist between the PA layer and propranolol at a neutral pH (Figure 4).

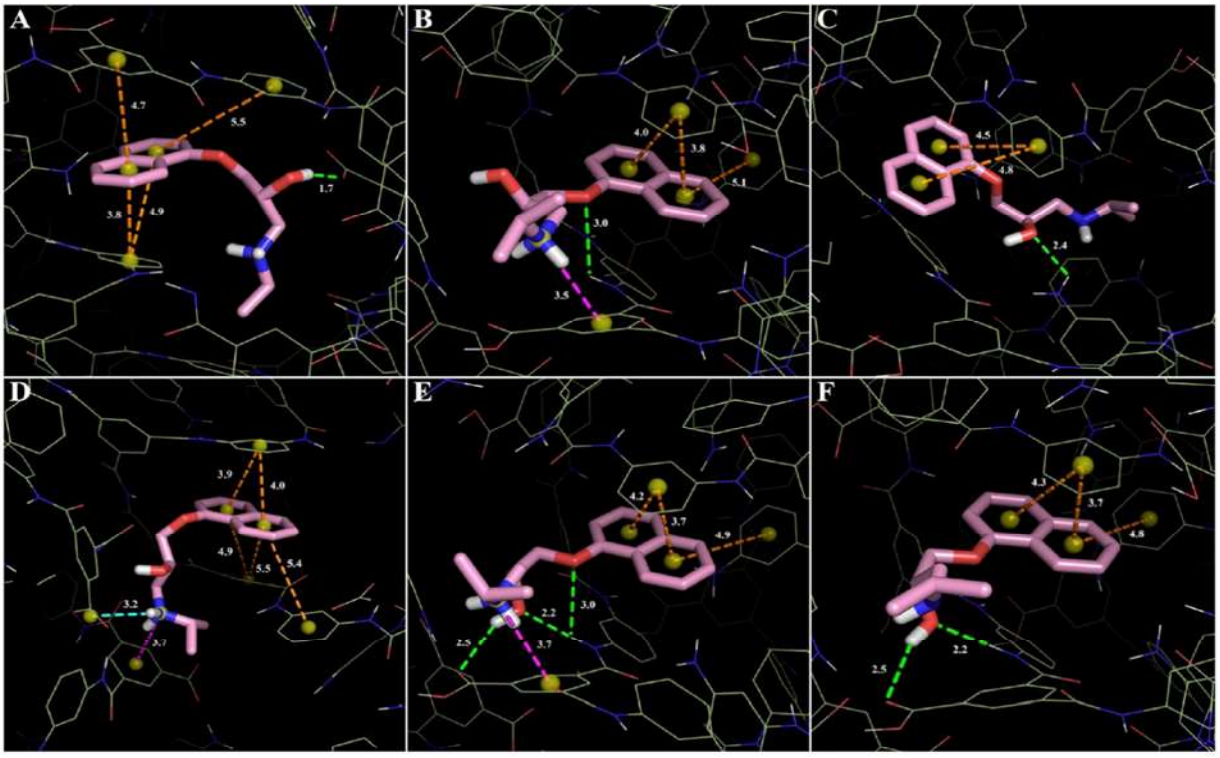

Figure 4. Binding mode between PA layer and propranolol (Pr): (A) PA^{-}-Pr^{+} (r), (B) PA^{o}-Pr^{+} (r), (C) PA^{o}-Pr (r), (D) PA^{-}-Pr^{+} (s), (E) PA^{o}-Pr^{+} (s), (F) PA^{o}-Pr (s). Hydrogen, nitrogen, and oxygen atoms are represented by white, blue, and red, while the carbon atoms of PA and Pr are represented by light green and pink colours, respectively. The dashed lines with different colours represent the solute-membrane interactions (hydrogen bonding by green, л-л stacking by orange, л-cation interaction by rose colour, and ionic bridge binding by cyan colour, respectively) [124]. Reprinted with the copyright permission

Yoon et al. [125] investigated the removal of 17b-estradiol, fluoranthene, and parachlorobenzoic acid by NF membranes in both the presence and absence of natural organic matter (NOM). Their findings revealed that hydrophobic adsorption was the main mechanism for the transport/removal of hydrophobic compounds and that the adsorption was positively correlated with hydrophobicity (log(Kow); fluoranthene (5.2), 17b-estradiol (4.0). parachlorobenzoic acid (2.7)). More precisely, the high removal of 17b-estradiol and fluoranthene during the initial filtration operation was governed by hydrophobic adsorption; however, once steady-state operation was achieved, size exclusion was the dominant removal mechanism for the tested compounds. For parachlorobenzoic

391 acid, the adsorption was insignificant due to its relatively lower hydrophobicity, however its
392 removal was attributed to electrostatic exclusion mechanism. The authors also reported that the
393 adsorption of 17b-estradiol and fluoranthene slightly decreased in the presence of NOM due to
394 competition for adsorption sites and pore blockage by NOM; however, the removal of
395 parachlorobenzoic acid showed no significant change in the presence of NOM as its removal was
396 mainly due to electrostatic exclusion rather than adsorption. The rejection behavior of estrone, 17-
397 β-estradiol, progesterone, and testosterone by NF membranes solely based on size exclusion was
398 below the estimated value. The removal of natural hormones by both NF membranes (i.e., NF90
399 and NF270) was similar, despite having different membrane pore size structures based upon their
400 MWCO values. This could be explained by a comparable polyamide active layer thickness (15-
401 40nm), which uniquely inhibited the diffusion process of natural hormones through both
402 membranes [126]. The sparsely soluble nature of water in the polymer led to the diffusion of
403 natural hormones through the polymer matrix, which was saturated with a small amount of water.
404 Although the convective flow made a small contribution to the transport of hormones across the
405 polymeric membrane, the presence of water was thought to have played an important role in
406 encouraging diffusion processes [127]. The diffusion of hormones through a dense polymeric
407 phase was accomplished after a series of successive attempts in forming and breaking the
408 secondary bond transformation between two bond sites (i.e., hydrophobic-bond to a substrate and
409 a hydrogen-bond to water) [128]. These findings suggested that the commercial TFC membranes
410 cannot serve as an absolute barrier for MPs.

411 Since the phenomena of MPs adsorption and subsequent diffusion occur on the active surface layer
412 of the membrane, the modification of the active layer through the incorporation of nanomaterials
413 and their deposition over the active layer surface have become a primary interest in the efforts to
414 improve membrane performance (Figure 5 and 6). The improved rejection of EDC and PhACs
415 (bisphenol A, ibuprofen, and salicylic acid) was observed from a chemically-modified NF
416 membrane via graft polymerization and cross-linking method. In contrast with a pristine membrane
417 (raw NF membrane = 74.1%), the chemically-modified membrane exhibited an improved
418 bisphenol A rejection (96.9%) [129]. Since bisphenol A showed a non-ionic behavior for the
419 selected pH condition (pH=7.2), it was presumed that steric hindrance associated with the
420 polymeric chain was the reason behind the improved bisphenol A removal. In addition, the
421 polymerized membrane reflected a stabilized performance against bisphenol A in contrast to the
422 pristine membrane, which was seemingly due to the improved bisphenol A adsorption allied with
423 the somewhat hydrophilic polymerized membrane. Moreover, the removal of negatively-charged
424 ibuprofen and salicylic acid by the polymerized membrane was slightly improved from 98.1% to
425 99.7% and 97.0% to 99.1%, respectively, clearly illustrating that the negatively-charged
426 membrane surface and polymer steric hindrance were directly responsible for this improved
427 behavior.

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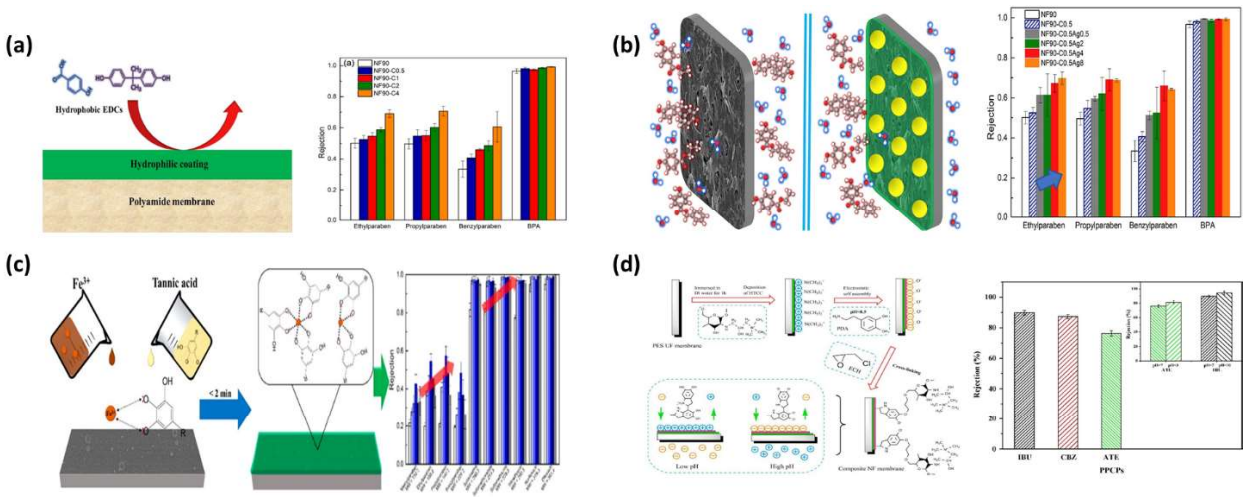

Figure 5. Effect of modified membrane surface (TFC) on MPs removal: (a) Polydopamine-coated membrane (left) and its rejection performance at different coating time intervals (right) [130]; (b) Silver nanoparticles immobilized on the commercial membrane surface using polydopamine (left) and its rejection performance for EDCs at different nanoparticle loadings (right) [131]; (c) Tannic acid and ferric ion-modified membrane (left) and its rejection performance for different MPs [132]; (d) Dual charged membrane surface (left) with improved MPs rejection upon varying feed solution pH (right) [9]. Reprinted with the copyright permission.

Similarly, the steric hindrance of a membrane which was polymerized for an extended duration (i.e., 60 minutes), compared to a membrane with a short polymerization period (i.e., 15 minutes), was found to be greater, since the longer polymerization period created a longer polymer chain [129]. Guo et al. [130] enhanced membrane performance against hydrophobic EDCs (i.e., ethyl-paraben, propyl-paraben, benzyl-paraben, and bisphenol A) through a hydrophilic coating utilizing polydopamine (PDA) as the coating agent (Figure 5a). The improved hydrophilic membrane exhibited high rejection and minimized the bisphenol A passage up to 75% as compared with the non-coated membrane. The performance of the modified membrane was also evaluated against neutral hydrophilic ethylene glycol, which exhibited no systematic change in the rejection, suggesting that the mechanism behind the improved performance was the weakened hydrophobic interactions between the EDCs and the membrane [130]. Another study reported better selectivity of membrane against EDCs via surface modification by PDA coating followed by in-situ immobilization of silver nanoparticles over the membrane surface (Figure 5b). Here, the higher detainment of EDCs was ascribed to the combination of steric impediment and weak hydrophobic interactions [131].

In a separate study, an improved rejection against the hydrophobic neutral EDCs (from 21.6% to 42.6% for methylparaben, 19.9% to 54.4% for ethylparaben, 21.3% to 57.3% for propylparaben, and 19.6% to 48.3% for benzylparaben) and six antibiotics (consistently above 95%) was obtained by Guo et al. [132] using the coordination complex of tannic acid and ferric ions, suggesting a green, fast, and simple surface modification approach for real applications in contrast with slow

PDA polymerization (Figure 5c). Ouyang et al. [9] reported a dually charged polyelectrolyte NF membrane as an effective approach for higher rejection (Figure 5d). The dually charged membrane surface exhibited an improved rejection against atenolol (76.22 to 81.67%) and ibuprofen (from 89.85 to 94.5%) when the pH of the feed was adjusted from neutral to acidic (pH = 3) and neutral to alkaline (pH = 10) conditions, respectively. The authors attributed this enhancement to the changes in the MPs and membrane surface charge properties.

Parallel to the functionalization of nanomaterials, their incorporation into the polyamide layer has also been found promising for minimizing the permeability-selectivity tradeoff of NF/RO. Paseta et al. reported improved performance of an NF membrane by controlling the positioning of nanofillers (i.e., metal organic framework (MOF) bi-layered TFC) (Figure 6a). The addition of ZIF93 (Zn(4-methyl-5-imidazolecarboxaldehyde)$_2$) and HKUST-1 (Cu_3(1,3,5-bencenetricarboxylate)$_2$($H2O)_3$) MOFs did not present any significant difference, showing >99% rejection for diclofenac and naproxen. However, it exhibited four times higher flux when compared with in-house fabricated TFC membrane [133]. A similar trend was observed by Dong et al. [134] from a TFN NF membrane prepared on a support with in-situ embedded zeolite nanoparticles (Figure 6d). Meanwhile, a simultaneous improvement of water flux and rejection against tris(2-chloroethyl) phosphate, tris(1-chloro-2-propyl) phosphate, and tris(1,3-dichloro-2-propyl) phosphate molecules from the TFN hollow-fiber NF membrane containing nanoporous SAPO-34 nanoparticles when compared with an NF90 membrane was reported by Liu et al. [135] (Figure 6b). Also, a TFN membrane with an optimum amount of silica nanoparticles (modified with oleic acid (OA)) in a trimesoyl solution (0 – 0.3 w/v%) resulted in an improved rejection of propazine (7%) and atrazine (4%) when compared with a pristine membrane (Figure 6c). The authors attributed this improved performance to the smaller pore size (0.35 to 0.32 nm) of the TFN membrane [136]. The strong interaction of the OA tails of the nanoparticles and polymer chain resulted in structural compactness, hence lowering the solute permeation through the membrane. However, water flux was enhanced due to the increased hydrophilicity of the TFN membrane [135,136]. Likewise, other studies focusing on the removal of MPs using commercial TFC, surface-modified, and nanocomposite-incorporated membranes are summarized in Table 3.

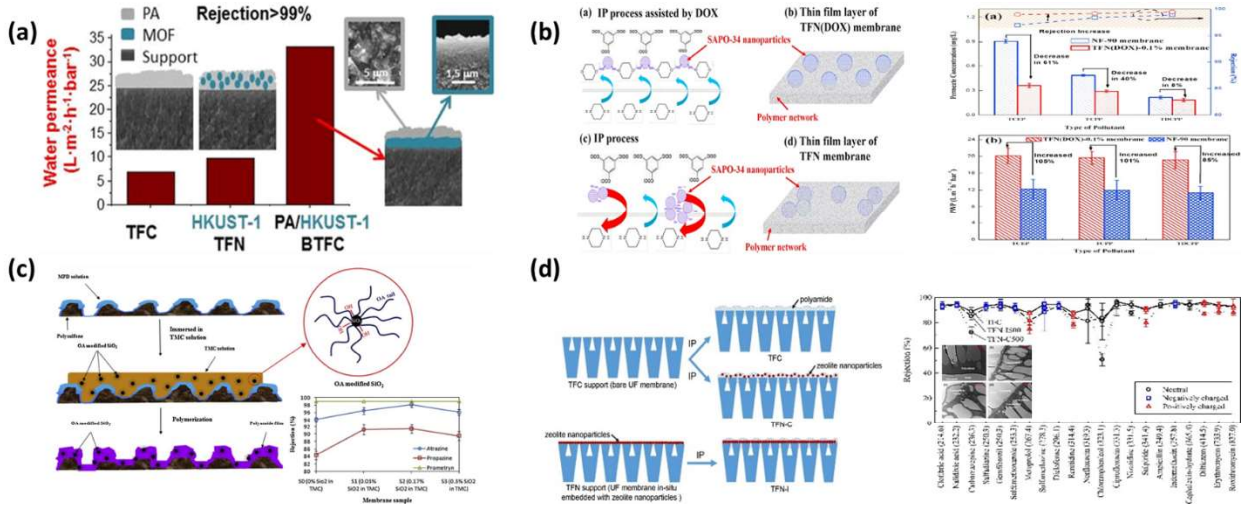

Figure 6. Effect of membrane morphology on MPs rejection: (a) Thin film nanocomposite membrane with metal organic frameworks [133]; (b) Thin film nanocomposite membrane with SAPO-34 nanoparticles (left) and its removal performance (right) [135]; (c) Thin film nanocomposite membrane containing oleic acid-modified silica nanoparticles (left) and its rejection performance at different loading conditions (right) [136]; (d) Thin film nanocomposite membrane embedded with zeolite nanoparticles (left) and its removal performance against positive, negative, and neutrally charged MPs [134]. Reprinted with the copyright permission.

498 **Table 3.** Removal of MPs by RO/NF membranes

Membrane Type	Membrane Material	Configuration	Rejection	Micropollutants	Feed	Capacity	Removal Mechanism	Ref
RO-BW30XLE Filmtec	Polyamide Thin Film Composite	Spiral Wound	45-98%	Gemfibrozil, Ketoprofen, Carbamazepine, Diclofenac, Mefenamic acid, Acetaminophen, Sulfamethoxazole, Propyphenazone, Hydrochlorothiazide, Metoprolol, Sotalol, Glibenclamide,	Ground Water, NE-Spain	Full-Scale	Size exclusion for neutral MPs & electrostatic interaction for negative charge MPs	[137]
RO-TriSep, X201-TSF	Polyamide Thin Film Composite	Flat Sheet	82-100%	Acetaminophen, Alachlor (Lasso), Atraton, Bisphenol A, Caffeine, Carbadox, Carbamazepine, DEET, Diethylstilbestero, Equilin, 17_-Estradiol, 17_-Estradiol, Estriol, Estrone, 17-Ethynyl Estradiol, Gemfibrozil, Metolachlor, Oxybenzone, Sulfachloropyridazine, Sulfamerazine, Sulfamethizole, Sulfamethoxazole	Lake Ontario water, membrane bioreactor effluent, and laboratory-grade water (Milli-Q)	Bench-Scale	Size exclusion	[138]
RO	Cellulose Triacetate Thin Film Composite	Hollow Fibre	25-95%	N-nitrosodimethylamine (NDMA), N-nitrosomethylethylamine (NMEA), N-nitrosopyrrolidine (NPYR), N-nitrosodiethylamine (NDEA), N-nitrosopiperidine (NPIR), N-nitrosomorpholine (NMOR), N-nitrosodipropylamine (NDPA), Caffeine, Simazine, Atrazine, Primidone, Meprobamate, Triamterene, Tris(2-chloroethyl)phosphate (TCEP), Trimethoprim, N-nitrosodi-n-butylamine (NDBA), N,N-Diethyl-meta-toluamide (DEET), Bisphenol A, Diuron, Carbamazepine, Linuron, Diazepam, Triclocarban, Clozapine, Omeprazole, Hydroxyzine, Paracetamo, buprofen, Naproxen, Gemfibrozil, Dilantin, Sulfamethoxazole, Ketoprofen. Triclosan, Diclofenac, Enalapril, Simvastatin hydroxy acid, Atenolol, Amitriptyline, Fluoxetine, Verapamil	Synthetic wastewater	Bench-Scale	Size exclusion & hydrophobic interaction	[139]
RO-BW30-400 Filmtec	Polyamide Thin Film Composite	Flat Sheet	87-99%	Acetaminophen, Bisphenol A, Caffeine, Carbamazepine, Cotinine, Ethinyl Estradiol-17α,	Synthetic water in line with the quality of North	Bench Scale	Size exclusion	[140]

				Gemfibrozil, Ibuprofen, Progesterone, Sulfamethoxazole, Triclosan, Trimethoprim	Bay Regional Water Treatment Plant, California, USA			
RO-XLE Filmtec RO-SC-3100 Toray	Polyamide Thin Film Composite Cellulose Acetate Thin Film Composite	Flat Sheet	57-91% 0-85%	2-Naphthol, 4-Phenylphenol, Phenacetine, Caffeine, NAC standard, Primidone, Bisphenol A, Isopropylantipyrine, Carbamazepine, Sulfamethoxazole, 17-Estradiol	Laboratory grade water (Milli-Q)	Bench Scale	Size exclusion for PA RO membrane & electrostatic interaction for cellulose acetate RO membrane	[141]
RO-SW30HRLE Lenntech	Polyamide Thin Film Composite		73-99%	NDMA (i.e. N-nitrosodimethylamine, N-nitrosomethylethylamine, N-nitrosopyrrolidine, N-nitrosodiethylamine, N-nitrosopiperidine, N-nitrosomorpholine, N-nitrosodi-n-propylamine, N-nitroso-n-dibutylamine) and PhACs (Atenolol, Bezafibraat, Carbamazepine, Clenbutanol, Clofibrinezuur, Diclofenac, Gemifibrozil, Ketoprofen, Metformine, Naproxen, Paracetamol, Pentoxifylline, Pindolol, Propranolol, Salbutamol, Sotalol, Sulfamethoxazool, tTrbutaline, Trimethoprim)	Tap Water	Small-Scale Pilot	Size exclusion	[123]
RO-SW30HR	Polyamide Thin Film Composite	Flat Sheet	76.5	N-nitrosodimethylamine	Synthetic Water	Lab-Scale	Size exclusion	[142]
RO-ESPA1-2521 Hydranautics	Polyamide Thin Film Composite	Spiral Wound	30-67%	Bisphenol-A, Carbamazepine, and Acetaminophen	Synthetic Water	Pilot-Scale	Steric effect	[143]
RO-ESPA2-LD Hydranautics.	Polyamide Thin Film Composite	Spiral Wound	75-99%	2,6-Dichlorobenzamide, 2-Hydroxyquinoline, Atrazine, Bisphenol A, Carbamazepine, DEET, Diuron, Triclosan, 1H-benzotriazole, 4-Hydroxyquinoline, Barbital, Caffeine, Chloridazon, Paracetamol, Phenazone, Phenylurea, Tolyltriazole, Triethyl Phosphate, Acesulfame, Bentazon, Diclofenac, Ibuprofen, PFBA, PFBS, Perfluorohexanoic acid, Sulfamethazine, Sulfamethoxazole,	Anaerobic riverbank filtrate	Pilot-Scale	Size exclusion, adsorption, and electrostatic repulsion	[84]

RO-XLE Filmtec RO-BW30 Filmtec RO-SW30XLE Filmtec	Polyamide Thin Film Composite	Flat Sheet	97-99% 96-99% 96-98%	Perfluorohexanoic acid	Synthetic wastewater	Lab-Scale	Size exclusion and electrostatic interaction	[119]
TFC-ROBW30 Filmtec GE Osmonics SG	Polyamide Thin Film Composite	Flat Sheet	less than Minimum Dedection Limit (8ng/L) less than Minimum Dedection Limit (8ng/L)	Betamethasone, Fluconazole, Phenylbutazone, Prednisone and Metformin	Doce river (geographical coordinates 18°51′50.45″S and 41°56′46.86″ W)	Lab-Scale	Size exclusion	[144]
NF 90-Filmtec NF270 - Filmtec NF-PMPF 34 (Koch Membrane) NF-DK GE Osmonics DK NF GE Osmonics Duracid	Polyamide Thin Film Composite	Flat Sheet	<Minimum Dedection Limit (MDL) (8ng/L) <MDL (8ng/L) <MDL (8ng/L) 44-99% 70 - 100% <MDL	Betamethasone, Fluconazole, Phenylbutazone, Prednisone and Metformin	Doce river (geographical coordinates 18°51′50.45″S and41°56′46.86″ W)	Lab-Scale	Size exclusion and hydrophobic interaction	[144]

NF90-Filmtec NF270-Filmtec	Polyamide Thin Film Composite	Flat Sheet	98-99% 82-96%	Perfluorohexanoic acid	Synthetic wastewater	Lab-Scale	Size exclusion and electrostatic interaction	[119]
NF90 - Filmtec	Polyamide Thin Film Composite	Spiral Wound	30-98%	Gemfibrozil, Ketoprofen, Carbamazepine, Diclofenac, Mefenamic acid, Acetaminophen, Sulfamethoxazole, Propyphenazone, Hydrochlorothiazide, Metoprolol, Sotalol, Glibenclamide,	Ground Water, NE-Spain	Full-Scale	Size exclusion for neutral MPs & electrostatic interaction for negative charge MPs	[137]
NF-270-Filmtec NF-TS80 TriSep, Goleta	Polyamide Thin Film Composite	Flat Sheet	0-93% 0-100%	Acetaminophen, Alachlor (Lasso), Atraton, Bisphenol A (BPA), Caffeine, Carbadox, Carbamazepine, DEET, Diethylstilbestero, Equilin, 17-Estradiol, 17-Estradiol (E2), Estriol (E3), Estrone (E1), 17-Ethynyl Estradiol (EE2), Gemfibrozil, Metolachlor, Oxybenzone, Sulfachloropyridazine, Sulfamerazine, Sulfamethizole, Sulfamethoxazole	Lake Ontario water, membrane bioreactor effluent, and laboratory-grade water (Milli-Q)	Bench-Scale	Hydrophobic interaction and cake-enhanced concentration polarization	[138]
NF270 - Filmtec	Polyamide Thin Film Composite	Flat Sheet	19.6-95%	Methylparaben, Ethylparaben, Propylparaben, Benzylparaben, Sulfadiazine, Sulfamethoxazole, Sulfamethazine, Trimethoprim, Norploxcin, Ofloxacin	Synthetic Water	Lab-Scale	Size exclusion	[132]
RO - SW75ES NanoH2O	Thin Film Nanocomposite		61-99%	NDMA (i.e. N-nitrosodimethylamine, N-nitrosomethylethylamine, N-nitrosopyrrolidine, N-nitrosodiethylamine, N-nitrosopiperidine, N-nitrosomorpholine, N-nitrosodi-n-propylamine, N-nitroso-n-dibutylamine) and PhACs (Atenolol, Bezafibraat, Carbamazepine, Clenbutanol, Clofibrinezuur, Diclofenac, Gemifibrozil, Ketoprofen, Metformine, Naproxen, Paracetamol, Pentoxifylline, Pindolol, Propranolol, Salbutamol, Sotalol, Sulfamethoxazool, Terbutaline, Trimethoprim)	Tap Water	Small-Scale Pilot	Size exclusion	[123]

NF	Thin Film Nanocomposite Dually Charged (polydopamine and quaternate chitosan)	Flat Sheet	76-89%	Atenolol, Carbamazepine, and Ibuprofen	Synthetic Water	Lab-Scale	Donnan exclusion and steric hindrance	[9]
NF-Dox	Thin Film Nanocomposite	Hollow Fibre	98-99%	tris (2-Chloroethyl) phosphate, tris(1-Chloro-2-propyl) phosphate, and tris (1,3-dichloro-2-propyl) phosphate	Wastewater	Lab-Scale	Size exclusion and electrostatic repulsion	[135]
NF	Thin Film Nanocomposite (Oleic acid modified silica nanoparticles)	Flat Sheet	90-98%	Atrazine, Propazine, Prometryn		Lab-Scale	Size exclusion	[136]
NF	Thin Film Nanocomposite (Embedded zeolite nanoparticles)	Flat Sheet	84- ≈95	Ampicillin, Carbamazepine, Cephalexinhydrate, Chloramphenicol, Ciprofloxacin, Clofibric acid, Diclofenac, Diltiazem, Erythromycin, Gemfibrozil, Indomethacin, Metoprolol, Nalidixic acid, Nizatidine, Norfloxacin, Ranitidine, Roxithromycin, Sulfadiazine, Sulfamethazine, Sulfamethoxazole, Sulpiride		Lab-Scale	Size exclusion and electrostatic interaction	[134]
NF (PA/HKUST -1 BTFC) NF (PA/ZIF-93 BTFC)	Thin Film Nanocomposite	Flat Sheet	98-99%	Naproxen, Diclofenac		Lab-Scale	Size exclusion	[133]

NF	Thin Film Nanocomposite with 0.4 Montmorillonite	Flat Sheet	72-93%	Cephalexin, Amoxicillin, Ibuprofen		Lab-Scale	Size exclusion, charge exclusion, physicochemical interaction	[145]
	0.6 Montmorillonite		55-85%					
	0.4 Modified Montmorillonite		85-97%					
	0.6 Modified Montmorillonite		83-96%					
RO-SW30HR, Filmtec	Thin Film Composite GO Modified	Flat Sheet	82.70%	N-nitrosodimethylamine	Synthetic Water	Lab-Scale	Size exclusion	[142]
RO-ESPA1-2521-Hydranautics	Thin Film Composite poly(glycidyl methacrylate) modified	Spiral Wound	59-100%	Bisphenol-A, Carbamazepine, and Acetaminophen	Synthetic Water	Pilot-Scale	Steric effect	[143]
NF270-Filmtec	Thin Film Composite with Fe 0.5	Flat Sheet	26-98%	Methylparaben, Ethylparaben, Propylparaben, Benzylparaben, Sulfadiazine, Sulfamethoxazole, Sulfamethazine, Trimethoprim, Norploxcin, Ofloxacin	Synthetic	Lab-Scale	Size exclusion	[132]
	Fe 1		33-98%					
	Fe 3		42.6-97%					
	Fe 6		33-99%					
	C0.5		26-98.5%					

3.2. UF for MPs Removal

3.2.1. Influence of MPs characteristics on their rejection

Unlike NF and RO, the removal of organic MPs by UF (particularly by size exclusion) is often considered negligible owing to the large MWCO of UF membranes (1-100 kDa), which is generally larger than the molecular weight of most MPs (< 1 kDa) [13,16]. Since UF membranes are not effective in retaining MPs based on size exclusion, adsorption is thus considered the mainstream mechanism contributing to the removal of MPs by UF membranes. This can be attributed to the fact that the adsorption of MPs in membrane filtration is not only restricted to the membrane surface but can also occur in the membrane's porous structure and is often directly related to pore radius [146]. Generally, membranes with larger pore sizes (UF membranes) allow MPs to access the membrane's internal porous structure (more adsorption sites), whereas the access of these pollutants to the internal sites may be limited in dense membranes (NF/RO). Hence, the more porous the membrane, the more MPs the membrane may allow to adsorb within the membrane pores in addition to its surface as a function of their physicochemical characteristics.

Secondes et al. [147] evaluated the removal of MPs (diclofenac, carbamazepine, and amoxicillin) by a UF membrane using a single hollow fiber membrane unit (A/G Technology Corporation, USA) with an active membrane area of 6.6 cm^2. The polysulfone (Psf) UF membrane (MWCO 100 kDa) exhibited low rejection (<30%) for all contaminants. The highest rejection was observed for diclofenac followed by carbamazepine and amoxicillin. This rejection trend was in correlation with their hydrophobic characteristics. Since the adsorption of MPs on the membrane surface is mainly derived by their hydrophobicity, adsorption was considered as the key rejection mechanism for MPs removal in this study. Similarly, Chon et al. [148] reported MW, log D, and charge characteristics (at neutral pH) as the major driving factors affecting the detainment of selected MPs (atenolol, carbamazepine, diclofenac, sulfamethoxazole, caffeine, dilatin, and florfenicol) by UF membranes. With the exception of diclofenac and sulfamethoxazole (>33% and >28% respectively), most of the targeted MPs were not effectively eliminated (<17%). Nevertheless, there was no clear relationship between the rejection of target contaminants and their properties (i.e., MW, log D, charge characteristics).

Wray et al. [149] reported a consistently low removal (<5%) of MPs from Milli-Q water spiked with 1000 ngL^{-1} using a UF membrane. This low rejection could be attributed to the dominance of the adsorption mechanism for MPs removal in UF processes. Since the size of the compounds (MW <300 $gmol^{-1}$) was smaller relative to the pore size of the membrane (0.04 µm), it was unlikely that the observed removal was due to size exclusion. Similar findings were reported by Pramanik et al. [150], who investigated the efficiency of a PVDF hollow-fiber UF membrane (Asahi Kasei Chemicals, Japan, pore size 0.1 µm) for the removal of perfluorooctanesulfonic acid and perfluorooctanoic acid contaminants from lake water. Their results revealed that the UF membrane had low removal efficiency for both perfluorooctanesulfonic acid and perfluorooctanoic acid

compounds (~20 and ~28 %, respectively), which was attributed to the bigger pore size of the UF membrane failing to act as a physical barrier for retaining the MPs.

Yoon et al. [151] tested the rejection of 27 MPs (i.e., PhACs and EDCs) having MWs < 0.4 kDa using a commercial UF membrane (GM, Desal-Osmonics, USA: MWCO -100 kDa). A low rejection (<30%) for all contaminants were observed, except for triclosan (>80%), oxybenzone (>70%), erythromycin (>60%), progesterone (>50%), and estrone (>40%). Their reported findings highlighted that the general separation trend was the hydrophobic adsorption of MPs as a function of K_{ow}. Since the adsorption of MPs over the membrane surface was the function of their hydrophobic value, it was believed that MPs with high hydrophilic properties (less hydrophobic; log K_{ow}<3) were improbable to be adsorbed over the membrane surface. However, MPs with high hydrophobicity (log K_{ow}>3) reflected the opposite behavior. Several other studies also reported similar trends for the removal of MPs using commercial UF membranes [146,149]

3.2.2. Effects of operating conditions on MPs rejection

Along with the characteristics of pollutants, the removal of MPs by UF is also largely dependent on the process operating conditions (either chemical and/or physical operating parameters). Irrespective of type, these operating conditions play a vital role in the removal of MPs in the UF process. Acero et al. [152] investigated the influence of important operating variables, such as membrane MWCO and pH on the rejection of 11 MPs, including acetaminophen, metoprolol, antipyrine, caffeine, sulfamethoxazole, flumequine, ketorolac, atrazine, isoproturon, hydroxybiphenyl, and diclofenac from municipal secondary effluents using UF membranes. According to their results, lower removal coefficients (<50%) were obtained for all of the tested compounds except for hydroxybiphenyl, with adsorption being the main mechanism for rejection of MPs by UF membranes.

The highest rejection coefficient for hydroxybiphenyl was attributed to its highest value of log D (3.27) at pH 7, which validates its high adsorption capacity. The remaining 10 compounds were poorly rejected by the UF membranes as they present log D values below 0.5 at pH 7, thus possessing lower adsorption capacities. The authors further reported that the removal (adsorption) of all tested compounds by the UF membranes was higher at pH 5 than at pH 9, in particular for compounds with a negative charge at high pH (sulfamethoxazole, flume-quine, ketorolac, and diclofenac). This observed behavior was attributed to the phenomenon that, as the pH increases, the concentration of negatively-charged species also increases, decreasing the hydrophobicity of the compounds (log D decreases versus log K_{ow}), thus hindering their adsorption on the membrane surface. Moreover, the authors found the contribution of the size exclusion mechanism by UF membranes to be insignificant, since the MWCOs of the membranes were much higher than the MW of the compounds.

In addition to pH speciation, the effect of turbidity on the MPs removal performance of a UF polyvinylidene fluoride membrane was investigated by Chen et al. [153] (Figure 7a). Kaolin clay was used to adjust the turbidity of a carbamazepine-spiked working water sample. In their findings,

577 they reported the unsatisfactory removal of carbamazepine (5%) from the feed when the turbidity
578 value is below 1 NTU. In contrast, an improved rejection was observed (15%) in an increasing
579 turbid feed environment (60 NTU) in the first UF circulation, which was attributed to the
580 enhancement of the sieving effect as a result of the cake layer (formed by the deposition of
581 particulate matters on the membrane surface) which intercepted the fraction of carbamazepine into
582 the UF membrane [153].

583 Likewise, Wray et al. [149] investigated the influence of shear stress on the removal of MPs from
584 three natural surface water (i.e., two lake and one river) by employing four different shear stress
585 regimes: 1) no shear stress; 2) low peak shear stress (representative of continuous coarse bubble
586 sparging); 3) sustained peak shear stress (representative of intermittent coarse bubble sparging);
587 and 4) high peak shear stress (representative of large pulse bubble sparging) (Figure 7b). The
588 addition/formation of continuous coarse, intermittent course, and large pulse bubble sparging
589 contributed positively and resulted in 18%, 22%, and 34% rejection of MPs, respectively.
590 However, no significant difference was observed from the controlled process (no shear stress;
591 32%). The authors attributed this low influence to the water matrix composition and compound
592 properties, since the high removal of MPs under no shear stress was likely due to a heavy fouling
593 layer which altered the membrane selectivity and was able to entrap MPs of larger molecular
594 weights.

595 Due to the high MWCO value of the UF membrane in comparison to the size of the MPs, it was
596 reported that an enhanced fouling layer, by reducing the membrane pore size, over the membrane
597 surface improved the size exclusion mechanism [154]. The fouled membrane demonstrated
598 different electronegativity as compared with the clean membrane and offered adsorption sites on
599 the cake as well as on the membrane, which ultimately contributed towards the rejection of the
600 contaminants. Furthermore, the fouling layer significantly modified membrane
601 properties/characteristics such as hydrophobicity and porosity, and the fouled membrane with high
602 hydrophilicity and low porosity favored MPs rejection [155]. Since the cake, with its low porosity,
603 endured relentlessly and possessed a large number of narrow pores, it provided more hindrance,
604 which kept the MPs from penetrating the membrane. In addition, hydrophobic contaminants were
605 found to be more repulsive to the hydrophilic cake [151]. Moreover, MPs might adsorb on the
606 humic substance to form a matrix which became co-rejected by the membrane [156].

607

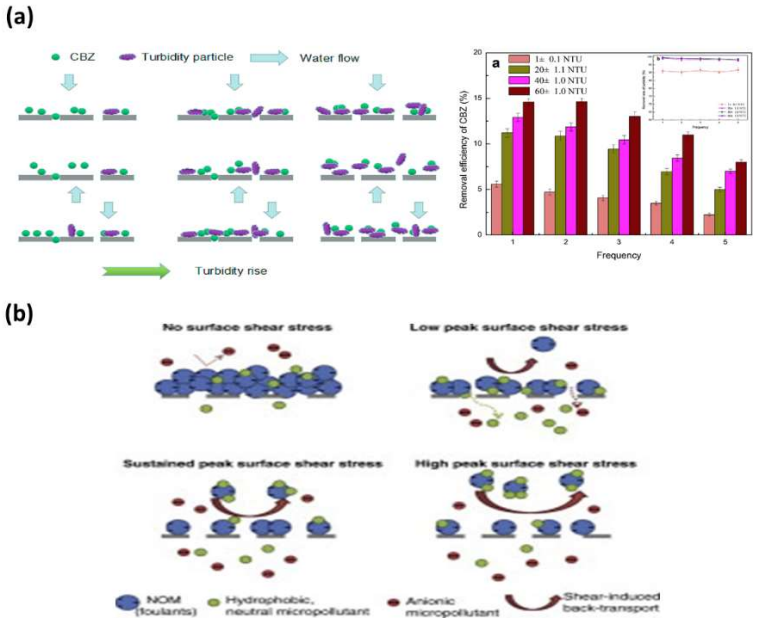

Figure 7. Effect of MPs characteristics and feed solution chemistry on their rejection by UF membranes: (a) Effect of feed solution turbidity on MPs removal: mechanism (left) and results (right) [153]; (b) Effect of NOM presence on MPs removal [149]. Reprinted with the copyright permission.

As aforementioned, along with feed water characteristics, process configuration/type in UF is another critical parameter for MPs removal. Electrochemical ultrafiltration processes for MPs removal have been recently explored as these methods have proven to be efficient and versatile for handling broad spectrum pollutants in the wastewater. Chen et al. [157] used an electro-ultrafiltration process to remove benzophenone-3 from water by applying an electric field across the membrane. Their results revealed that electro-ultrafiltration significantly increased benzophenone-3 rejection, which was later attributed to electrophoretic migration and electro-osmosis. Other electrochemical reactions (such as electrolysis, oxidation, and reduction) which may have occurred at the electrode presumably changed the chemical structure and/or mineralized the pollutants. These findings were in accordance with another study by Chen et al. [158] that focused on the electro-ultrafiltration of 4-methylbenzylidene camphor. In a separate study, Bakr et al. [159] used an electrochemical filtration system with a CNT-based Bucky paper as a flat sheet membrane electrode for the removal of ibuprofen and bisphenol A. Their tested crossflow configuration was highly efficient in retaining both pollutants from salt electrolyte as well as from synthetic wastewater effluents at an applied DC potential of 3V. They further revealed that the delayed stay time of 18.3 s for the two pollutants in the membrane was sufficient enough for a near-complete degradation of both the contaminants.

Some of the MPs (if present in high concentration) can also be degraded using the photocatalytic process. Recently, efforts have been made to employ hybrid photocatalytic UF processes for the photodegradation of MPs as well as for the recovery of used photocatalyst materials. In

continuation of these efforts, Singh et al. [160] fabricated Cu_2O photocatalyst modified Psf mixed matrix ultrafiltration membrane using the phase inversion method for visible light-driven photocatalytic removal of PhACs. The authors reported that the Cu_2O-Psf membrane exhibited superior flux, improved porosity, increased hydrophilicity, high protein adsorption, and successful removal of ibuprofen at 86% under visible light conditions. In another study, Chakraborty et al. [161] studied the degradation of a PhAC compound (chlorhexidine di-gluconate) by heterogeneous photocatalysis using TiO_2 nanoparticles immobilized on polymeric commercial hollow fiber ultrafiltration membranes. A 40% degradation of the chlorhexidine was achieved under simultaneous filtration and simulated solar light radiation. In another study, Plakas et al. [162] tested a fully automated photocatalytic membrane reactor (PMR) pilot unit and evaluated the unit for the degradation of diclofenac. The PMR-pilot system had a maximum system capacity of 1.2 m^3/d of treated water with two combined processes: heterogeneous photocatalysis (dispersed TiO_2 nanoparticles with UV-C irradiation) and membrane separation (submerged ultrafiltration hollow fibers). Pilot test results with tap and surface water under stable and continuous operation demonstrated an excellent steady state performance (~96%) for diclofenac degradation under UV-C irradiation.

It is worthwhile to mention here that there is little or no information contained in the reviewed literature on the formation or removal of the byproducts of degraded MPs which may form during the hybrid UF-photocatalytic/advanced oxidation/electrochemical treatments. Considering the MWCOs of most UF membranes, it is assumed that the potential byproducts may not be removed by the UF process and can be released into the water bodies. Thus, it is highly desirable to track the potential degradation of MPs during the process and ensure their adequate removal. Table 4 presents a summary of studies focusing on the removal of MPs using UF or the combination of UF with other treatment processes.

658 **Table 4**. Removal of MPs by UF membrane

Membrane Type	Membrane Material	MWCO /pore size	Conf.	Feed waters	Removal Efficiency	Removal Mechanism	Ref
UF	1) PES 2) Regenerated Cellulose 3) Cattle intestine	1 kDa	Flat sheet	Synthetic MPs feed	Sulfamethoxaz 20-50 % Carbamazepin 20-30 % Diclofenac 75-85 % Ibuprofen 70-80 %	Hydrophobic interactions Size exclusion after MPs binds with humic substances	[163]
UF	PES	2 kDa	Flat sheet	Synthetic feed Ground water Surface water Secondary wastewater	Amoxicillin 18 % Naproxen 75 % Metoprolol 55 % Phenacetin 10 %	Adsorption	[164]
UF	Cellulose/PP	1-100 kDa	Flat sheet	Synthetic MPs feed	Estrone 11-42 % Estradiol 25-45% Progesterone 35-65 % Testosterone 4-26 %	Solute-solute interactions	[165]
UF	PVDF	100 nm	Hollow fiber	Lake water Spiked pollutant	Perfluorooctanoic acid 39 % Perfluorooctanesulfonic acid 25 %	Hydrophobic and electrostatic interactions	[150]
MEUF	Cellulose	3 kDa	Flat sheet	Secondary wastewater effluent Spiked pollutant CTAB	UF 15-20% MEUF 40-74 %	Adsorption through surface complexation	[166]
UF	PVDF	100 kDa	Flat sheet	Synthetic secondary effluent	Bisphenol A 64%–76%, 17α-ethynyl 42%–53% Estrone 28%–46% 17β-estradiol 24%–63% Estriol 10%–17%	Adsorption and size exclusion	[155]
UF	PVDF	40 nm	Hollow fiber	Synthetic MPs feed Lake water 1 Lake water 2 River water	Average Removal (3 % Synthetic feed) 14 % Lake water 1 30 % Lake water 2 36 % River water	Entrapment of MPs in the fouling layer via adsorption or sieving mechanism	[149]
MEUF	PES	5 kDa	Flat sheet	Synthetic MPs feed Secondary wastewater effluent CPC surfactant	Acetaminophen 17% Metoprolol 8 % Caffeine 12 % Antipyrine 65 % Sulfamethoxazole 94 % Flumequine 65 % Ketorolac 99 %	Hydrophobic and electrostatic interactions	[167]

UF (Pilot)	Alumina/TiO_2	1 kDa	Tubular	Synthetic MPs feed	Atrazine 9 % Isoproturon 70 % 2-Hydroxybiphenyl 98 % Diclofenac 98 % Vitamin B12 57 % Lysozyme 85 % L-tyrosine 5 % L-phenylalanine16 %	Adsorption	[168]
UF	Alumina/TiO_2	1-8 kDa	Tubular	Wastewater effluent	Acetaminophen 20-45% Caffeine 10-20 % Diazepam 45-70 % Diclofenac 32-75 % Erythromycin 50-85 % Ibuprofen 35-62 % Naproxen 28-60 % Sulfamethoxazole 18-54 % Triclosan 20-75 % Trimethoprim 18-50 %	Electrostatic interactions Hydrophobic/hydrophilic interactions	[169]
UF	Alumina/TiO_2/GO	1 kDa	Flat sheet	Synthetic MPs feed	Ibuprofen 60 % Sulfamethoxazole 47 %	Electrostatic repulsion Size exclusion	[170]
UF	TiO_2 and ZrO_2	150 kDa	Tubular	Secondary wastewater effluent	Bisphenol A 48-100 %	Sieve effect after BPA-OM complex formation	[171]
UF	PES/Silica dioxide	-	Hollow fiber	Synthetic MPs feed	Bisphenol A 99.61 %	Adsorption and size exclusion	[172]
UF	PES / SWCNT	6.48 nm	Flat sheet	Synthetic MPs feed	Bisphenol A 80 % 4-Nonylphenol 84 %	Adsorption	[173]
UF	PES/N-doped CNTs	37 nm	Flat sheet	Drinking water Wastewater sources	Carbamazepine 88.97 % Galaxolide 99.92 % Caffeine 87.21 % Tonalide 98.85 % 4-nonylphenol 99.15 % Bisphenol A 98.59 %	Hydrophobic interactions	[174]
UF	PVC/ MWCNTs/Fe_3O_4	33-36 nm	Flat sheet	Synthetic MPs feed	Bisphenol A 65 % Norfloxacin 23 %	Charge effect and/or adsorption	[175]

UF	PES/Silica/Germanium dioxide nanoparticles	-	Flat sheet	Surface water	Bisphenol A 97 % Technical 4-Nonylphenol 98.7 % Tonalide 92 % Carbamazepine 90 % Caffeine 87.42 % Galaxolide 99 %	Hydrophobic interactions	[176]
UF	Psf/ PVP/GO	70 nm	Flat sheet	Synthetic MPs feed	Bisphenol A 59 %	Electrostatic repulsion	[7]
UF	Polysulfone/GO	-	Flat sheet	Synthetic MPs feed Tap water	Ofloxacin Benzophenone-3 Rhodamine b, Diclofenac, Triton X > 90 % for all contaminants	Adsorption	[177]
UF	PVDF	80 nm	Flat sheet	Synthetic MPs feed Humic acid	Benzophenone-3 99.9 %	Hydrophobic adsorption Steric exclusion after BP-3-OM complex formation Electrophoretic migration and electroosmosis	[157]
UF	PVDF	80 nm	Flat sheet	Synthetic MPs feed Humic acid	4-methylbenzylidene camphor 99.9 %	Hydrophobic adsorption Steric exclusion after BP-3-OM complex formation Electrophoretic migration and electroosmosis	[158]
UF	MWCNT	-	Flat sheet (Bucky Paper)	Synthetic secondary wastewater effluent	Ibuprofen, Bisphenol A > 90 % for both contaminants at 3V	Electrochemical degradation	[159]
UF	TiO_2/ZrO_2	100 nm	Tubular	Synthetic MPs feed Humic acid	Perfluorooctanoic acid 80 % Perfluorooctanesulfonic acid 84 %	Electrochemical degradation	[178]
UF (Pilot)	PVDF/TiO_2	30 nm	Hollow fiber	Tap water Surface water	Diclofenac ~ 96 %	Photocatalytic degradation	[162]
UF	Fe-doped TiO2/PSF	-	Flat sheet	Synthetic MPs feed	Bisphenol A ~ 90 %	Electrostatic adsorption Photocatalytic degradation	[179]
UF	CA/MWNT–TiO_2	-	Flat sheet	Synthetic MPs feed	Carbamazepine 80 % Ibuprofen 45 %, Acetaminophen 24%	Hydrophobic adsorption	[180]
UF	PVC-PAN/ TiO_2	100 kDa	Hollow fiber	Synthetic MPs feed	Chlorhexidine Digluconate ~ 40 %	Photocatalytic degradation	[161]

UF	Psf/PEG /Cu_2O	75 nm	Flat sheet	Synthetic MPs feed	Ibuprofen ~ 86 %		Photocatalytic degradation	[160]
UF-MIEX	Cellulose/PP	1-100 kDa	Flat sheet	Synthetic feed Humic acid	Estradiol	Only UF 12 % Hybrid 80 %	Ion exchange interactions	[181]
COA-Oz-UF (Pilot)	Al_2O_3	60 nm	Flat sheet	Surface water	Bisphenol A, Nonyl phenol, 4-tert-octylphenol, Estrone, Estradiol, 17a-estradiol, Estriol, 17a-ethinylestradiol, Erythromycin, Lincomycin, Roxithromycin, Sulfamethoxazole, Sulfamerazine, Sulfapyridine, Griseofulvin, Trimethoprim, Indomethacin, Benzafibrate	Only UF ~50 % COA ~10 % UF-Oz ~90 %	Adsorption Coagulation-sedimentation Hydrophilic/hydrophobic interactions Oxidation	[182]
COA-PAC-UF	-	100 kDa	Cylindrical	Wastewater		Only UF / Hybrid		[183]
					Acetaminophen	0 % / 85 %	Adsorption Coagulation-sedimentation Hydrophobic interactions Sieving effect	
					Caffeine	0 % / 100 %		
					Carbamazepine	65 % / 85 %		
					Cotinine	45 % / 88 %		
					Diclofenac	25 % / 100 %		
					Gemfibrozil	47 % / 100 %		
					Ibuprofen	1 % / I 92 %		
					Metoprolol	38 % / 97 %		
					Naproxen	0 % / 100		
					Sulfadimethoxine	18 % / 93 %		
					Triclosan	100 % / 100 %		
					Trimethoprim	4 % / 100 %		
AB-UF	PA	1 nm	Flat sheet	Synthetic MPs feed		Only UF / Hybrid		[184]
					Ibuprofen	41 % / 60 %	Hydrophobic adsorption	
					17 α- estradiol	22 % / 62 %		
					Carbamazepine	8 % / 42 %		
MIEX-UF	PVDF	40 nm	Flat sheet	Synthetic MPs feed	Carbamazepine	Only UF 13 % / Hybrid 74 %	Adsorption rejection Ion exchange interactions	[153]
ACA-UF-Ultrasonic	Psf	100 kDa	Flat sheet	Synthetic MPs feed		Only UF / Hybrid		[147]
					Diclofenac	22 % / 99 %	Adsorption Sonolytic degradation	
					Carbamazepine	20 % / 99.4 %		
					Amoxicillin	30 % / 99.8%		

3.2.3. Effects of membrane properties on MPs rejection

The UF membrane's type (polymeric, ceramic, nanocomposite) and properties (MWCO, charge, hydrophilicity, etc.) are also critical for the removal of MPs. For instance, Comerton et al. [146], studied the removal of 22 MPs (i.e., EDCs and PhACs) using a commercial PSf UF membrane (TriSep, UE10, Goleta, CA, MWCO- 10 kDa) and reported that the removal of selected contaminants by the UF membrane was lower as compared to tested NF and RO membranes. They concluded that adsorption, rather than size exclusion, was the mainstream removal mechanism for MPs using a UF membrane. Alongside commercial polymeric UF membranes, many research groups have also investigated the efficiency of ceramic UF membranes for the efficient removal of organic MPs. For instance, Garcia-Ivars et al. [169] investigated the rejection of ten selected PhACs in secondary wastewater effluent using ceramic ultrafiltration membranes (INSIDE CéRAMTM, TAMI Industries, France, MWCO 1-8 kDa). The results of the study revealed that during filtration, a foulant layer was formed on the ceramic membrane surface, which eventually benefited the rejection of the selected contaminants by providing a secondary barrier with different hydrophobicity and charge. Similarly, Zielińska et al. [171] investigated the removal of bisphenol A using a ceramic UF membrane (INSIDE CéRAMTM, TAMI Industries, MWCO 150 kDa) during the post-treatment of wastewater effluent. The reported total removal efficiency of the tested membrane for bisphenol A was above 98%. The authors attributed the high bisphenol A removal by the ceramic UF membranes to the sorption of bisphenol A on the particulate organic matter present in the wastewater effluent as well as to the direct adsorption of bisphenol A on the membrane surface.

It should be noted that in both the abovementioned studies, adsorption of MPs on the ceramic UF membranes was the predominant rejection mechanism, which depended significantly on the feed solution chemistry as well as the presence of organic and inorganic compounds (foulants). The pristine ceramic membranes were not capable of retaining most of the MPs by the sieving/seize exclusion effect in the absence of organic foulants in the feed water. As discussed earlier, in the presence of organic matter in the feed water, a foulant layer is formed on the ceramic UF membrane by the adsorbed organic compounds. This fouling layer is generally hydrophobic and negatively-charged and reduces the pore size and the porosity of the ceramic membrane due to complete or intermediate pore blocking during the initial stages of the filtration. As a result, the rejection of certain MPs (hydrophobic and negatively-charged) may increase significantly compared to clean ceramic UF membranes mainly due to 1) the repulsion between the negative charge of the additional foulant layer and negatively-charged MPs; 2) the hydrophobic interactions between the foulant layer and hydrophobic MPs; and 3) the formation of organic macromolecules-MPs complexes which can be retained by size exclusion or charge repulsion effects.

As UF membranes are incapable of rejecting MPs based on the size exclusion mechanism, lately, efforts have been devoted to modifying UF membrane properties to enhance MPs removal and overall membrane performance (Figure 8). In this regard, the use of hydrophilic nanomaterials

with high adsorption capacity is considered a new viable approach for tailoring UF membrane surface properties. Among various nanoparticles, carbon nanotubes (CNTs) and graphene oxide (GO), owing to their unique properties (high adsorption capacity, presence of oxygen-containing functionalities, low fouling potential, and high aqueous stability), are the two most widely used nanomaterials to change the characteristics of UF membrane for MPs removal. Zambianchi et al. [177] fabricated PSf-GO-based UF membranes by the phase inversion method for the removal of PhACs and PCPs from water (Figure 8a). Their findings revealed that the PSf-GO membrane showed a high affinity for organic pollutants (>90% removal) with GO-driven preferential adsorption of hydrophilic and polar molecules. Kaminska et al. [173] reported that the addition of single-walled carbon nanotubes (SWCNT) to PES UF membranes improved the removal of two endocrine disrupters, including bisphenol A and nonylphenol. They further reported that increasing the nanotube concentration in the PES membranes made them slightly more hydrophobic, empowering the adsorption of bisphenol A and nonylphenol, which are also hydrophobic, to make adsorption an underlying mechanism in the removal of the selected pollutants.

In another study, Singh et al. [160] also reported improved ibuprofen removal from a Cu_2O-modified mixed matrix membrane when operated under different pH conditions (Figure 8b). The high rejection of ibuprofen (24%) in an acidic condition in contrast with neutral (11.7%) and basic (7.9%) feed pH was mainly attributed to the enhanced adsorption effect. Owing to its pKa value of 4.52-4.9, ibuprofen demonstrated neutral behavior under an acidic pH range (pH = 2.7-4.9), and the positive charge of the modified membrane facilitated the adsorption mechanism. Whereas, when the feed pH was neutral or alkaline, both the composite membrane and ibuprofen possessed negative charges and thus repelled each other. Similarly, various other studies reported that the addition of GO and CNTs in UF membranes significantly improved the overall rejection of various MPs [7,175,185].

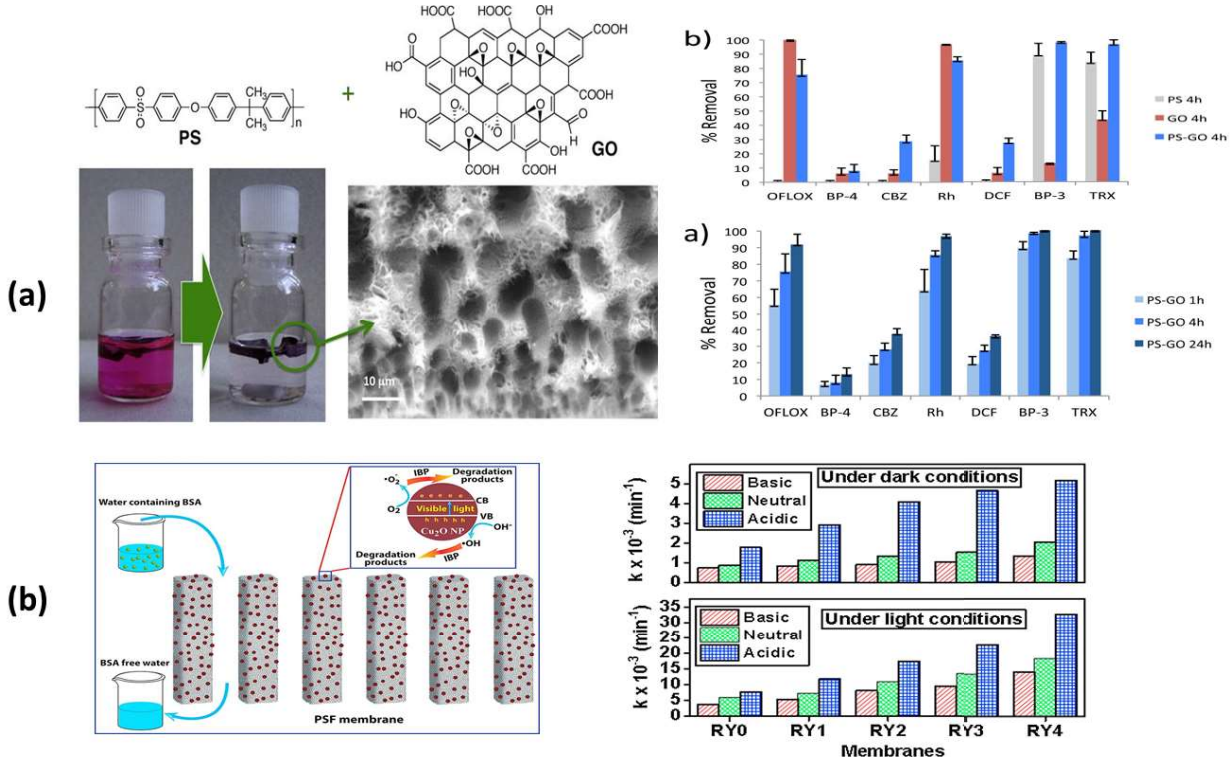

Figure 8. Effect of membrane properties and operating conditions on MPs rejection: (a) Morphology of graphene oxide-doped PSf UF membrane (left) and its removal performance (right) [177]; (b) Cu_2O modified UF membrane (left) and its removal against ibuprofen at different loadings and pH conditions [160]. Reprinted with the copyright permission.

3.3. FO for MPs Removal

3.3.1. Influence of MPs characteristics on their rejection

Until the 1990s, the concept of FO was only applied as an experimental method for determining the properties of RO membranes. However, since its first use for water treatment, FO has been gaining much interest, as can be seen from the number of papers published each year. Especially from 2008, the number of publications on FO started to increase rapidly, and in particular, the number of published papers dealing with the treatment of MPs using FO appeared in greater frequency from 2012. This interest in FO from the scientific community comes from the idea that FO may be able to replace pressure-driven membrane processes. FO uses a semi-permeable membrane that has many advantages to offer over pressure-driven processes, such as high recovery, lower energy consumption, and low fouling. These advantages result from using naturally occurring osmotic pressure as its driving force. Unlike other membrane processes (i.e., RO, NF, and UF), FO requires a natural osmotic gradient (caused by a concentration gradient between the feed solution and draw solution), rather than high pressure applications, for water molecules to pass through the membrane [186]. In FO, driven by the osmotic gradient, water molecules are diffused through a dense, semi-permeable membrane from the feed solution towards

744 the draw solution. As such, FO is considered as a simple and economical process, enabling FO to
745 emerge as a low-cost, high performance, and low energy requirement membrane-based treatment
746 method and drew the attention of researchers and other stakeholders towards its commercialization
747 [71]. However, although FO has some very promising potential for MPs removal, at least one
748 additional process is required to produce clean water using FO and regenerate the draw solution.
749 Hence, FO is often coupled with another membrane process, such as UF, MD, NF, or RO, for draw
750 solution regeneration.

751 Similar to other membrane-based processes, one of the factors that affects the removal of MPs in
752 the FO process is the characteristics of MPs. Linares et al. [187] investigated the removal
753 mechanism of FO membranes (Hydration Technology Innovation) against the hydrophilic neutral
754 (1,4-dioxane, acetaminophen, metronidazole, phenazone, caffeine), hydrophobic neutral
755 (bisphenol A, carbamazepine, 17α-ethynyl estradiol), and hydrophilic ionic (ibuprofen, naproxen,
756 fenoprofen, gemfibrozil, ketoprofen) MPs at neutral pH (pH=7). For the selected MPs, size
757 exclusion was found to be the dominant mechanism for their removal. In addition, the removal of
758 MPs was observed in the following order: ionic MPs (92.9-96.5) > hydrophobic MPs (40-87.5) >
759 hydrophilic neutral MPs (48.6-84.7). The high rejection of hydrophobic and ionic compounds was
760 attributed to adsorption and electrostatic repulsion, respectively [187]. Hancock et al. [188]
761 investigated the performance of bench-scale FO for a wide range of ionic (positive and negative),
762 non-ionic, and hydrophobic non-ionic MPs. The bench-scale FO demonstrated a 40-98% rejection.
763 In contrast with the non-ionic MPs (40-90%), a high rejection was observed for charged MPs (80-
764 98%). The high rejection value for charged compounds was subsequent to their electrostatic
765 interaction and repulsion with the negatively-charged FO membrane [189]. The improved rejection
766 of four MPs (carbamazepine, ibuprofen, diclofenac, and naproxen) was observed for FO
767 membranes with increased hydrophobic characteristics [190], which indicated that the short-term
768 rejection of MPs was influenced by the hydrophobic interaction between the cellulose triacetate
769 (CTA) FO membrane surface and selected MPs [191]. Despite having similar hydrophobic
770 properties (at pH 6; Log D for carbamazepine = 2.45, for ibuprofen = 2.43), the high removal of
771 carbamazepine (MW: 236 $gmol^{-1}$) in contrast with ibuprofen (MW: 206 $gmol^{-1}$) elucidated the
772 dominance of the size exclusion effect in the rejection of MPs by FO membranes.

773 Similarly, the average rejection for MPs by a FO membrane was observed in the following manner:
774 sulfamethoxazole (MW=253.3 $gmole^{-1}$; 67-90%) > carbamazepine (MW=236.3 $gmole^{-1}$; 68-
775 83%) > atrazine (MW=215.7 $gmole^{-1}$; 34-49%) > 4-chloraphenol (MW=128.6 $gmole^{-1}$; 28-39%)
776 > phenol (MW=94.1 $gmole^{-1}$; 21-22%) [87]. This descending order clearly illustrated the
777 correlation between the FO membrane's impounding tendency and the molecular size of MPs. The
778 observed rejection for sulfamethoxazole and carbamazepine was relatively high, which could be
779 attributed to the large MW and the dominant charge effect (from the negative charge of
780 sulfamethoxazole at pH=7). The cumulative effects of the small MW and low hydrophobic values
781 was considered the reason behind the FO membrane's significantly low removal of 4-chlorophenol
782 and phenol.

3.3.2. Effects of operating conditions on MPs rejection

Although the characteristics of MPs greatly influence their rejection in the FO process, feed solution chemistry and operating conditions, such as draw solution type, fouling layer thickness, membrane orientation, and CFV, also play significant roles in the overall rejection performance. The effect of varying pH conditions (pH 3, 5, 7, and 9) on selected PhACs (metoprolol, sulfamethoxazole, and triclosan) removal by both modified (impregnated with TiO_2-PDA) and pristine membranes showed interesting results for each selected compound [192]. The removal of sulfamethoxazole was enhanced from ~85% to >96% with the pristine membrane and 91% to >97% with the modified membrane by changing the solution pH value (pH=3-9), which varied the charge property of sulfamethoxazole from neutral at pKa1<pH<pKa2 to negatively-charged at pH>pKa2). In addition, the increase in pH resulted in decreased zeta potential values of the pristine (−2 to −35 mV) and modified membranes (−6 to −45mV) due to the increased dissociation of the carboxyl functional group (COO^-) on the active layer. The speciation of sulfamethoxazole from neutral to negatively-charged at high pH promoted an electrostatic repulsion between the negatively-charged membrane surface and sulfamethoxazole, hence resulting in a higher rejection. In contrast, triclosan, which is neutral at pH 7, was found to be relatively independent of pH for the modified membrane, showing above 95% removal for all pH conditions (95-98% removal); whereas, for the pristine membrane, the rejection of triclosan was improved from 90% to 97%. This was presumably due to the change in the hydrophilic characteristic of the pristine membrane (contact angle ~ 38°) and modified membrane (contact angle ~ 26°), which consequently led to low adsorption of neutrally charged triclosan over the surface. However, for metoprolol, which is positively charged with pKa=9.49, no significant influence of pH variations was observed for the selected pH range. The dominant mechanism for the removal of metoprolol was revealed to be electrostatic interaction and steric impediment [192].

Similarly, the removal of hormones as a function of water recovery ranging from 20-70% showed promising results (>95% rejection). The performance of the negatively-charged FO membrane against estrone and 17-β-estradiol (uncharged) hormones was improved by the application of an anionic surfactant (sodium cocoyl N-methyl taurate) [186]. As illustrated in Figure 9a, it is supposed that the hydrophobic interaction between the membrane and surfactant tail causes the deposition of individual surfactant molecules over the membrane surface [97]. The removal of hormones in the presence of surfactant by a relatively hydrophilic membrane (contact angle = 61°) was likely to be improved by following two proposed mechanisms: (i) the formation of micelles due to hydrophobic interactions between the hormones and the anionic surfactant, which provides a platform for the hormones to be adsorbed on the hydrocarbon chain, thereby avoiding hormone-membrane interaction; and (ii) the adsorption of surfactant molecules over the membrane surface, which halts the transfer of hormones by avoiding the hydrophobic interaction of hormones over the membrane surface [186].

Numerous studies have suggested that the formation of a fouling layer on the membrane surface increases the removal efficiency of MPs. Membrane fouling in FO also has been found to play a

significant role in terms of MPs removal by becoming a hindrance to the hydrophobic interactions of ionic and non-ionic compounds [193]. Primarily, the type of foulant and its formation pattern over the membrane surface determined the effect of fouling on MPs removal. Hancock et al. [188] observed that the removal of MPs increased substantially with the presence of a fouling layer; Valladares Linares et al. [187] determined that with the formation of fouling layer, the charge and hydrophobicity of the membrane surface were altered, leading to an enhanced rejection of ionic and neutral MPs. Xie et al. [194] found that the rejection of carbamazepine and sulfamethoxazole was enhanced with the growth of humic acid (HA) deposition on the membrane surface (Figure 9b). Additionally, the formation of an alginate fouling layer on an FO membrane declined the removal of sulfamethoxazole and naproxen; however, no significant variation was observed for the remaining 18 MPs rejection [195]. In this study, the lower rejection of sulfamethoxazole and naproxen appeared to be independent of MW or the charge of the compounds. Also, the author did not find any clear link for the dominant transport mechanism (convection and/or diffusion) and, therefore, attributed that the solute-membrane and solute-foulant specific interactions were responsible for the poor rejection [195].

The formation of a more porous cake layer structure of alginate seemed to promote what is called concentration polarization by providing a hindrance to MPs in the back diffusion toward bulk feed [103]. In another study [192], an improved rejection of sulfamethoxazole was observed in the presence of HA by both pristine and modified (TiO_2) FO membranes due to the formation of an HA shield layer on charged membrane surfaces. In contrast, a negligible impact was observed for triclosan (neutral) rejection, which was attributed to the permeation in the absence of electrostatic interactions for the selected pH condition. The addition of HA led to a negative impact on metoprolol rejection by both pristine and modified FO membranes, resulting in a high concentration in the permeate since the positively-charged metoprolol (pH=7) was deposited over the HA layer followed by their diffusion through the membrane [192].

Other than feed solution chemistry, changes in other operating conditions may also significantly influence MPs rejection. Alturki et al. [196] found that the rejection of charged and small molecular weight MPs was higher for 0.5 M NaCl, compared to 2 M NaCl, in the active layer facing the draw solution (AL-DS mode), which was due to the high reverse solute flux (RSF) in 2 M NaCl. The high RSF resulted in a higher ionic strength inside the support layer, leading to a reduced solute rejection by electrostatic interaction. Compared to the AL-DS mode, the rejection of charged and small MW MPs in the active layer facing the feed solution (AL-FS mode) was higher because of the position of the active layer/support layer. For example, in AL-DS mode, the water permeates through the support layer first, which causes the internal concentration polarization (ICP) of the MPs to be more severe, thus resulting in lower rejection.

Likewise, during a bench-scale FO experiment, >99% removal for estrone and 17-β-estradiol was observed at 20% recovery, after which the rejection behavior declined for both estrone and 17-β-estradiol with increasing recovery till 45% (95-96% during 20-45% recovery), then slightly improved by the end of experiment (96-97% during 45-70% recovery) [186]. Another study

demonstrated improved sulfamethoxazole removal with increasing CFV condition (i.e., 9.8 cms^{-1} to 58.8 cms^{-1}), indicating the significance of CFV over the diffusive movement, which was attributed to the decreasing concentration polarization effect [87,188,192]. Another study found that the reverse solute transport of DS in an osmotically-driven process had a positive influence, as it provided a hindrance to the organic pollutants and inhibited their forward diffusion phenomena [197]. FO membranes also showed different performance behaviors when operated at different capacity levels (i.e., pilot-scale and lab-scale) in order to check the removal of 23 EDCs, PhACs, and PCPs. A significantly high rejection value (80 ≥ 99%) was observed during the pilot-scale experiment, however, for the lab-scale arrangement, a declining behavior (40-98%) in terms of EDC, PhACs, and PCPs removal was observed [188]. Although the reason for this wide variation is unclear, membrane compaction, high hydrodynamic conditions, and fouling layer formation were considered as the proximate aspects. The studies focusing on the removal of MPs from FO membranes are summarized in Table 5.

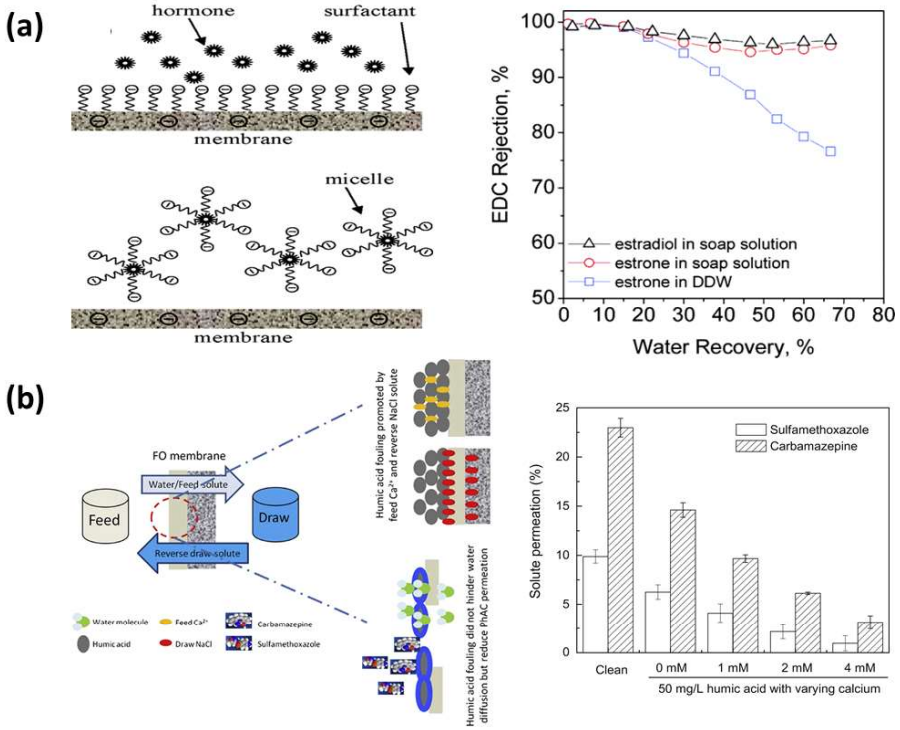

Figure 9. Effect of MPs characteristics and feed solution chemistry on MPs rejection by FO membranes: (a) Deposition of surfactant and on the membrane (left) and its effect on MPs removal (right) [186]; (b) Formation of an HA fouling layer on the membrane and its effect on MPs rejection at different HA concentrations [194]. Reprinted with the copyright permission.

881 **Table 5**. Removal of MPs by FO membrane

Membrane type	Membrane material	Membrane Properties	Capacity	Config.	DS	Feed	Contaminant & rejection (%)	Mechanism	Ref.
FO	CTA	Pore radius: 0.37nm Contact angle: 62.8 ± 3.9º	Lab-scale	Flat sheet	1M NaCl	5µM of each contaminant	PHN (21.9) 4CP (38.6) Atrazine (48.7) Carbamazepine (82.6) SMT (89.7)	Size exclusion and RSF	[197]
FO	CTA, TFC	Pure water permeability: 0.65±0.03 (CTA), 4.70±0.16 (TFC) Salt (NaCl) permeability coefficient: 0.25±0.07 (CTA), 0.16±0.03 (TFC) Membrane structural parameter: 0.67±0.13 (CTA), 0.52±0.11 (TFC) Pore radius: 0.33 – 0.40 nm (CTA), 0.41 – 0.44 (TFC)	Lab-scale	Flat sheet	0.5M NaCl	2 µg/L of contaminants + 20 mM NaCl and 1 mM NaHCO3	12 TrOCs CTA (30~92) TFC (65~98)	Size exclusion and charge effect	[198]
FO	CTA	Contact angle: 61º Surface charge: negative	Lab-scale	Flat sheet	20~ 70 g/L NaCl	Estrone (330 ng/L) Estradiol (290 ng/L)	Estrone (>96)) 17β-Estradiol (>96))	Size exclusion and charge effect	[186]
FO	TiO2 modified membrane	Contact angle: 27.04±2.99º Surface roughness (Sa): 45.50nm Surface charge: negative	Lab-scale	Flat sheet	0.5M NaCl	MTP, SMX and TCS, total of 500 µg/L	Triclosan (95~99) Metoprolol (89~93) Sulfamethoxazole (90~99)	Size exclusion	[192]
FO	CTA	N/A	Lab-scale	Flat sheet	3M NaCl	2 µg/L	10 Negative (>95) 6 Neutral (50~85) 5 Positive (89~98)	Size exclusion and charge effect	[195]
FO	CTA	N/A	Lab-scale Pilot-scale	Flat sheet Spiral wound	30g/L Synthetic seasalt	SMBR permeate	>30 compounds Positive (70~95) Negative (60-95) Hydrophobic nonionic (40~95) Nonionic (40~95)	Size exclusion	[188]

FO	CTA-ES, CTA-NW	N/A	Lab-scale	CTA-ES (12.45) CTA-NW (6.48)	0.1~3.0 M NaCl	Mixture of 24 PhACs (100 µg/L)	CTA- ES: Nalidixic acid (82.5) Gemfibrozil (83.0) Carbamazepine (84.7) Sulfamethoxazole (88.2) Naproxen (90.2) Diclofenac (92.9) Propranolol (95.0) Sulfamethazine (97.2) Clofibric acid (97.4) Chloramphenicol (97.8) Diltiazem (98.0) Metoprolol (98.1) Indomethacin (98.4) Ranitidine (98.8) Nizatidine (99.0) Ciprofloxacin (99.0) Sulpiride (99.5) Erythromycin (99.5) Sulfa- diazine (99.7) Norfloxacin (99.7) Roxithromycin (99.8) Ampicillin (~100) Cephalexin-hydrate (~100%) CTA- NW: Gemfibrozil (87.1) Nalidixic acid (91.7) Sulfamethoxazole (93.9) Carbamazepine (94.4) Diclofenac (96.0) Propranolol (97.8) Naproxen (97.9) Indomethacin (98.1) Clofibric acid (98.5) Chloramphenicol (98.6) Metoprolol (98.7) Sulfamethazine (98.7) Ciprofloxacin (98.7) Sulpiride (98.8) Diltiazem (98.9) Sulfa- diazine (99.1) Ranitidine (99.3) Nizatidine (99.6) Norfloxacin (99.7)	Size exclusion	[199]

							Roxithromycin (99.8) Erythromycin (99.8) Ampicillin (~100) Cephalexin-hydrate (~100%)		
FO	CTA, TFC	Contact angle: 43 - 45º Surface charge: negative	Lab-scale	Not specifie d	1M NaCl	10 mM NaCl 250 µg/L (each PhACs)	CTA: Carbamazepine (94~95) Diclofenac (94~96) Ibuprofen (82~83) Naproxen (65~73) PA-TFC: Carbamazepine (>95) Diclofenac (>95) Ibuprofen (>95) Naproxen (>95) Sulfamethoxazole (>95) Trimethoprim (>95) Norfloxacin (>95) Roxithromycin (>95)	Charge effect, size exclusion, adsorption	[190]
FO	CTA	N/A	Lab-scale	~13 LMH	2M NaCl	200 µg/L of each antibiotic +model SWWE		Electrochemical oxidation, size exclusion, charge effect	[200]
FO	CTA, TFC	Contact angle: 76.6 (CTA), 45 (TFC) Zeta potential (at pH6): -2.1 mV(CTA), 86 mV (TFC)	Lab-scale	Varied dependi ng on the DS concentr ation.	117 ~ 194.5 g/L NaCl	100 mg/L	>96 (CTA ≥ TFC)	Size exclusion and charge effect	[201]
FO	CTA	Surface charge: negative (zeta potential between 04 - -8mV) Contact angle: 62±7.2º	Lab-scale	10~12	1M NaCl	5 µM	Sulfamethoxazole (90) Carbamazepine (83) Atrazine (49) 4-Chlorophenol (39) Phenol (22)	Size exclusion	[87]
FO	CTA	Surface charge: negative Pure water permeability (A): 1.08 Salt permeability (B): 0.245 Contact angle: 64±3º Pore diameter: 0.74 nm	Lab-scale	0.5M DS(5~6) 2.0M DS (11~12)	0.5M NaCl 2.0M NaCl	750 ng/L	40 TrOCs 0.5M DS (20~99) 2.0M DS (30~99) (larger MW= higher rejection)	Size exclusion and charge effect	[196]

FO	Aquaporin	Pure water permeability coefficient (A): 2.09±0.02 Salt (NaCl) permeability coefficient (B): 0.07±0.01 Structural parameter (S): 301±36 Pore radus: 0.30 (average)	Lab-scale	0.5M DS (68~98) 1.0M DS (72~99) 2.0M DS (88~99)	0.5~2M NaCl	Synthetic wastewater + 2 µg/L TrOCs	30 TrOCs 0.5M DS: Negative (80~98) Non-ionic hydrophobic (72~98) Non-ionic hydrophilic (68~99) 1.0M DS: Negative (83~98) Non-ionic hydrophobic (84~99) Non-ionic hydrophilic (72~99)	Size exclusion and solution diffusion	[202]
FO	CTA, Aquaporin	Pore radius: 1.04 nm(CTA) Surface charge: negative (CTA)	Lab-scale	CTA (~5) Aquaporin (~10)	1M NaCl	1mg/L	2.0M DS: Negative (>98) Non-ionic hydrophobic (>90) Non-ionic hydrophilic (>90) Aquaporin: Atrazine (>98) 2,6-dichlorobenzamide (>98) desethyl-desisopropyl-atrazine (>98) CTA: Atrazine (~70) 2,6-dichlorobenzamide (~56) desethyl-desisopropyl-atrazine (~20)	Size exclusion and charge effect	[203]
FO	TFC	Pore radius: 0.39 – 0.41nm Pure water permeability (A): 0.68 – 3.94	Lab-scale	15~20	1M NaCl	500 mg/L	Nitrobenzene (74~81) Aniline (>90) Phenol (70~80)	Size exclusion	[204]
FO	Not specified	Surface charge: negative Contact angle: 58.8±0.3°	Lab-scale	~6	Seawater (from Red Sea)	0.005~0.5 µg/L (within SWWE)	13 MPs 48~96	Size exclusion, charge effect, membrane swelling	[187]

3.3.3. Effects of membrane properties on MPs rejection

Membrane properties, such as polymer/material, pore size, hydrophobicity, and charge, are also reported to have a considerable impact on MPs rejection in FO. Kong et al. [199] studied the removal of 23 MPs using two different types of CTA membranes, one with an embedded polyester screen mesh (CTA-ES) and the other one with a non-woven backing consisting of polyester fibers individually coated with polyethylene (CTA-NW). The removal of contaminants from the CTA-ES and CTA-NW membranes ranged from 82.5% to 100% and 87.1% to 100%, respectively. This difference in rejection resulted from the differences in the permeability coefficient for water and salt, where the CTA-ES had higher permeability, resulting in a lower rejection (Figure 10a). In comparison with the CTA membranes, a TFC polyamide membrane showed better performance against selected PhACs (carbamazepine, diclofenac, naproxen, and ibuprofen) with 94%-97% rejection and offered high flux values (4.53 and 8.15 µm/s). The CTA membranes rejected the selected PhACs in the following order: carbamazepine, 95-96% > diclofenac, 92-95% > ibuprofen, 82-83% > naproxen, 64-73%, with declining flux values of 3.29 and 3.64 µm/s. The higher removal efficiency of the TFC membrane could be attributed to a combination of various aspects: (i) better size exclusion property, verified by the high glucose rejection of the TFC polyamide membrane; (ii) the electrostatic repulsion between the negatively-charged membrane and deprotonated (negatively-charged) PhACs; and (iii) the adsorption of PhACs over the membrane surface [190]. Thus, TFC membranes offered great MPs rejection in contrast to the CTA membranes [205]. The possible justification for this behavior could be: (i) a considerably different active layer structure; (ii) a relatively high-charged surface; and (iii) significantly high pore hydration characteristics (Figure 10b) [198,206,207]. These factors indicated prospects for improved FO membrane performance by modifying surface properties [198].

Similarly, Madsen et al. [203] studied and compared the performance of a CTA membrane and a biomimetic TFC aquaporin membrane in rejecting three organic MPs (atrazine, 2,6-dichlorobenzamide, and desethyl-desisopropyl-atrazine). The rejection of all compounds was significantly higher for the aquaporin membrane, which rejected over 97% of all three compounds. In the case of the CTA membrane, rejection varied for each compound: the rejection for desethyl-desisopropyl-atrazine, 2,6-dichlorobenzamide, and atrazine were approximately 22%, 56%, and 70%, respectively [203]. Although numerous studies have been performed to determine the effects of the membrane properties on the rejection of MPs in FO processes, not many studies discuss the relationship between the membrane module type (flat sheet, hollow fiber, spiral-wound, etc.) and MPs rejection. This may be because FO is not a mature process, and the number of studies done on hollow fiber FO membranes is limited. Further research is required on hollow fiber FO membranes for a better understanding of the rejection mechanism, especially concerning MPs removal by FO processes.

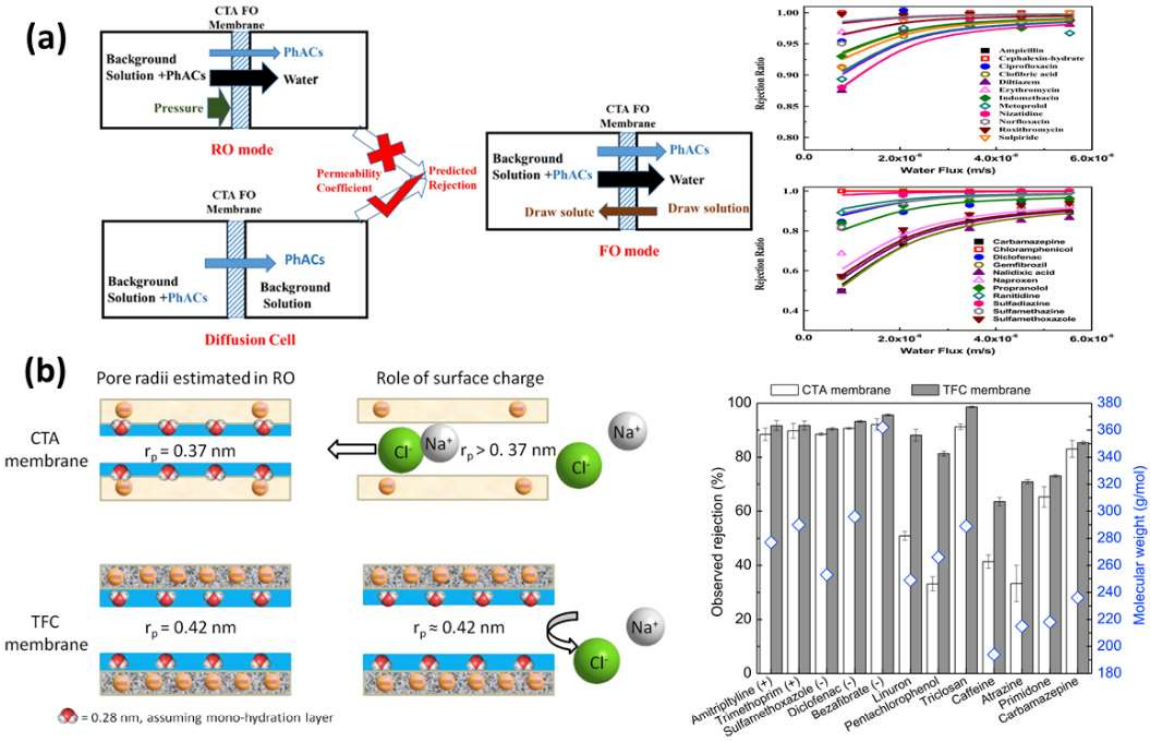

Figure 10. Effect of membrane properties and operation conditions on MPs rejection: (a) Effect of the diffusion coefficient: Mechanism (left) and results (right) [199]; (b) Effect of membrane properties: Mechanism (left) and results (right) [198]. Reprinted with the copyright permission.

3.4. MD for MPs Removal

3.4.1. Influence of MPs characteristics on their rejection

MD has emerged as an attractive method for desalination and wastewater treatment [208]. MD's low operating temperature, near-zero hydrostatic pressure, low mechanical strength requirement, and high rejection performance are taking MD toward its commercial application in multiple domains including RO brine management [209], crystallization [210], wastewater reclamation [211,212], food industries (juices and milk processing) [213–215], petrochemical industries [216], and pharmaceutical region [217]. In addition, the low temperature requirement of the process has been explored for the integration with the existing processes by utilizing low-grade/waste-heat and/or renewable energy resources for heating the feed [218,219]. Unlike other membrane processes (i.e., RO, NF, UF, and FO), MD is a non-isothermal membrane-based treatment process that uses a hydrophobic membrane as a separating medium between the hot feed and cold permeate [220]. The hydrophobic membrane inhibits direct feed permeation and allows only water vapors to pass through the membrane. Therefore, the pore size of the MD membrane is bigger than other (NF/RO/UF) membranes, thereby excluding the size exclusion mechanism as an option for separating pollutants. Instead of the concentration difference, electric potential, and hydrostatic pressure gradient utilized by other membrane-based treatment systems, the driving mechanism in the MD process is the vapor pressure gradient caused by a temperature difference between the feed

and permeate [221] which forces the volatile compounds to move through membrane pores. Accordingly, volatility plays an important role in the MD separation of MPs; non-volatile pollutants are retained in the feed, while those with high volatility could easily pass through the hydrophobic membrane and pollute the permeate. Moreover, membrane wetting in MD is still one of the major problems which could result in a process failure. Amphiphilic contaminants found in challenging feeds such as oil or shale gas wastewaters reduce the surface tension of the feed water and/or become adsorbed on the membrane surface and, consequently, induce partial or complete membrane wetting [222]. Membrane fouling in MD has also shown to induce partial membrane wetting [223]. Once wetting occurs, the separation mechanism is no longer sustained, and direct passage of feedwater to the permeate side will easily occur, yielding to a deteriorated rejection and failure of the MD process.

Guo et al. [8] investigated the potential of MD as an alternative treatment method in the biological and chemical treatment systems for removing antibiotics from wastewater. In this study, the negatively-charged commercial polyvinylidene fluoride (PVDF) membrane exhibited a high rejection performance (100%) against ten negatively-charged antibiotics. However, less removal was observed for the positively-charged antibiotics (i.e., Gentamicin sulfate, 86%; and tobramycin, 78%) with a declining flux (Figure 11), presumably due to the electrostatic interactions and deposition of positively-charged PhACs over the membrane surface leading to membrane pore blockage and their permeation through the membrane. This hypothesis was also validated by scanning electron microscopy (SEM), high-performance liquid chromatography (HPLC), and Fourier transform infrared spectroscopy (FTIR) results [8]. A separate study conducted a long-term experiment (500 hrs) using a direct contact membrane distillation (DCMD) arrangement for the removal of MPs (diclofenac, azithromycin, clarithromycin and erythromycin) from distilled water and synthetic seawater, followed by three different real water solution matrices (river water (RW-R), seawater (SW-R), and secondary treated municipal WWTP (MW-R) effluent). The process resulted in a higher rejection of the anti-inflammatory diclofenac, however, no antibiotics (azithromycin, clarithromycin, and erythromycin) were detected in the permeate and concentrate. Thermal degradation was found to be a possible reason for this phenomenon [224]. In agreement with this finding, Llorca et al. [225] also reported on the low stability of azithromycin, clarithromycin, and erythromycin when kept in the water for one week.

The volatility and hydrophobicity of MPs are reported as important physicochemical properties affecting MD performance. A commercial PTFE membrane was used to assess the removal of three antibiotics (azithromycin, clarithromycin, and erythromycin) and an anti-inflammatory drug (diclofenac) present in a real seawater feed. All three antibiotics were removed during the MD operation. In fact, both the permeate and retentate streams did not contain any detectable concentration of those MPs, which was attributed to the thermal degradation of the said antibiotics. Diclofenac, on the other hand, was successfully removed but was found in the retentate stream with 4 times more intense signal than in the feed. The removal of diclofenac was attributed to the physical and chemical characteristics of low volatility, negative surface charge, and hydrophilicity

[224]. In a separate study, a set of 29 MPs consisting of pharmaceuticals, industrial chemicals, pesticides, phytoestrogens, steroid hormones, and UV filters was selected to examine the feasibility of the MD process for their removal during water and wastewater treatment. The MPs with $pK_H > 9$ (low volatility) exhibited a high rejection (>90%), while moderate volatile ($pK_H < 9$) and hydrophobic (log D=5.18, 3.37, 3.21) contaminants (4-tert-octylphenol, benzophenone, and 4-tert-butylphenol) showed lower rejections (54%, 66%, and 73%, respectively). The volatile characteristics of MPs resulted in a phase conversion followed by their permeation across the membrane [226]. The same fate/transport of MPs was reported by Wijekoon et al. [226], who concluded that hydrophilic compounds with low volatility were mainly concentrated in the retentate stream, while those with hydrophobic nature and moderate volatility were lost by either thermal degradation or adsorption.

DCMD configuration has been applied to assess the MD performance for the treatment of wastewater reverse osmosis concentrate (WWROC) to achieve a zero liquid discharge approach. To evaluate the water reuse potential, a WWROC sample was collected from Sydney Olympic Park Authority (SOPA) containing 20 MPs with different classifications (household and industrial chemicals, antibiotics/prescription drugs, fire retardant, hormones, and pesticides/herbicides). It is worth highlighting that DCMD showed considerably good rejection (96-99%) for most of the MPs with the exception of propyl-paraben, salicylic acid, benzophenone, triclosan, bisphenol A, and atrazine, which were detected in the permeate and exhibited low rejections (50, 86, 62, 83, 84, and 88%, respectively). This low rejection could be associated with many factors, such as high hydrophobicity, high volatility, and electrostatic interactions [211]. In a separate study, a pilot-scale air gap membrane distillation (AGMD) plant revealed a high rejection (below the detection limit) of 37 PhACs during the treatment of wastewater effluent. However, sertraline exhibited a different trend in both the feed and permeate during the concentration process by AGMD and was detected in the permeate in trail 1, 2, and 4. This unique trend could be related to sertraline's highly hydrophobic characteristic (log K_{ow}=5.76) which resulted in the adsorption and permeation of sertraline through organic membrane [77]. In a separate study, the removal of estrone and 17-β-estradiol from wastewater using a capillary micro-porous hydrophobic membrane (MD020-CP-2N, Microdyn, Germany) in a DCMD arrangement was investigated to explore the application of MD in a space shuttle. The nonvolatile estrone and 17-β-estradiol was showed high rejection (>99.5%) in the DCMD process [186].

3.4.2. Effects of operating conditions on MPs rejection

Despite the fact that separation in MD is not based on the size exclusion mechanism but rather a phase change mechanism, like other membrane-based treatment processes, operating parameters play a significant role in MD systems. Operating parameters in MD could be classified into chemical conditions, such as feed and compounds chemistry (composition, concentration, pH, charge), and physical conditions, such as flowrates, module configuration, and temperatures. Both chemical and physical operating conditions have shown to affect MPs removal in MD processes.

During the inspection of the electro-kinetics interactions between PhACs and a hydrophobic PVDF membrane, differently charged antibiotics such as ciprofloxacin (neutral), tobramycin (positively-charged), and cefotaxime (negatively-charged) were used. At varying feed pH conditions (pH=1-11.82), the removal of positively charged tobramycin was improved with a declining fouling trend and enhanced flux values. However, no significant variation was observed for the negatively-charged cefotaxime. The zeta potential value of tobramycin decreased from +42 to +3mV with increased pH value (pH=1-11.8), whereas the positively-charged tobramycin exhibited neutral characteristics at pH=11.8, consequently weakening the opposite charge interaction with the negatively-charged PVDF membrane. Similarly, ciprofloxacin showed a slightly positive ZP value for pH < 4, possessed the iso-electric point (IEP) at pH 5, and was slightly negatively-charged at pH > 6 [8]. To evaluate the influence of MPs concentration (x 1000) over MD performance, a short experiment was performed with diphenhydramine as a representative MP using a DCMD arrangement (W-cell; stream flow perpendicular to the membrane surface). 30 mg of diphenhydramine was detected in the permeate (1.3 wt%) when the distilled water was spiked with 2.3 gL^{-1} as the feed. This was presumably due to the higher affinity of the positively-charged diphenhydramine towards the negatively-charged polytetrafluoroethylene (PTFE) membrane, facilitating its permeation [224]. The removal efficiency of MPs by MD subjective to its varied concentration in the feed was also reported in a previous study [211]. The removal of non-steroidal anti-inflammatory drugs (i.e., diclofenac, ibuprofen, naproxen) from ultrapure-water, tap-water, and primary and secondary effluents (collected from a wastewater treatment plant, Szczecin Poland) using a photocatalytic membrane reactor (PMR) in a DCMD arrangement was investigated. A capillary hydrophobic polypropylene membrane (Accurel PP S6/2; Membrana GmbH, Wuppertal, Germany) with an effective area of 0.014 m^2 and a pore size of 0.2 µm was utilized for the photolysis and photo-catalysis degradation. The removal of contaminated drugs from the photolysis/photo-catalysis-MD was depicted in the following order: ultra-pure water > tap-water > secondary effluent > primary effluent. The presence of organic compounds, inorganic ions, and effluent turbidity reflected the detrimental effects on the degradation of the drugs, which could be attributed the decrease in the amount of ultraviolet (UV) irradiation for the photolysis/photo-catalysis degradation [227]. In a separate study, MD system performance for surface and groundwater treatment was investigated to check the robustness of the system and explore its potential applications. Different feed streams (distilled water to synthetic feed mimicking surface water and WWRCO) spiked with 5 mgL^{-1} ibuprofen were utilized to achieve the set target. The performance of the membrane was also observed in the presence of HA (HA=100 and 160 mgL^{-1}) at variable pH conditions (pH=2.6, 7.2, and 11) to understand the contaminant rejection behavior in the presence of NOM (i.e., carboxylic group due to the dissociation of HA at high pH). The results demonstrated that the PVDF membrane (Durapore GVHP; Merck-Millipore) exhibited a 88-92% ibuprofen rejection when operated with the synthetic feed matrix. Unlike other membrane-based treatment processes, no significant influence was observed by HA over the rejection of ibuprofen at variable pH conditions. However, the complete rejection of ibuprofen was observed by MD when operated with WWROC, though the

spiked amount of ibuprofen was lower (i.e. 0.2 mgL^{-1}) in order to mimic more realistic condition [228].

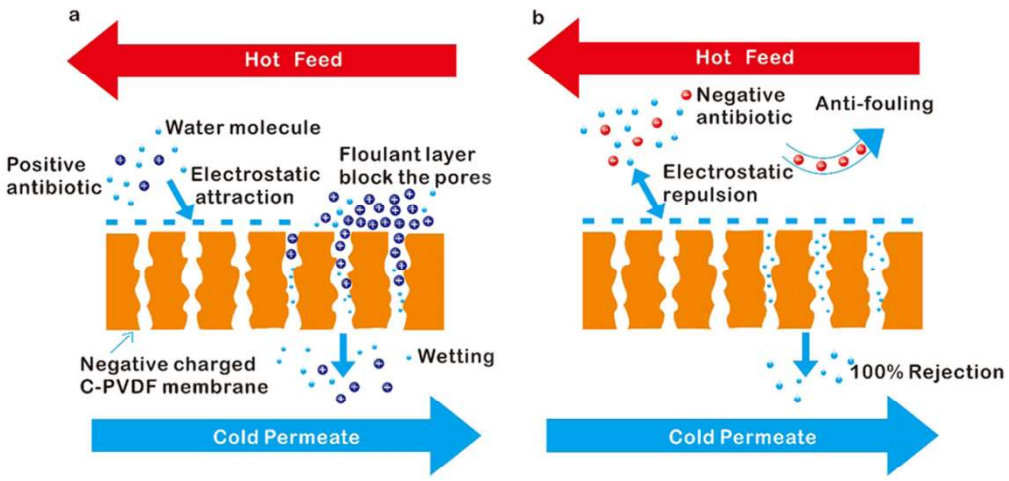

Figure 11. Effect of MPs characteristics and feed solution chemistry on MPs rejection in MD [8]. Reprinted with the copyright permission.

Temperature, flow rate, membrane surface area, and membrane configurations are also noted as important parameters in MD. Gethard et al. utilized MD (X-50 hollow fiber membrane, Membrana, USA) as an online concentration technique for the determination of pharmaceutical residues (i.e., ibuprofen, diphenhydramine, acetaminophen, and dibucaine) in natural water [229]. With different membrane modules having different surface areas, the experimental results using a 5 mg/L ibuprofen in pure water solution at 90 °C feed temperature and 0.5 mL/min flowrate showed a high correlation between trace residues concentration and the membrane surface area. A linear trend was found in the increased solvent reduction (SR) and enrichment factor (EF) for higher membrane surface area. The feed temperature showed a similar trend where both SR and EF were enhanced at higher feed temperature under constant feed concentration and flowrate when increasing the feed temperature up to 90 °C. Upon raising the feed temperature to 100 °C, both EF and SR were decreased. These observations were attributed to the possibility that, because the permeating vapor is not dry at higher temperatures but rather carry small water droplets, they would block some of the membrane pores to result in an overall reduction in permeability. The feed flowrate, on the other hand, showed an opposite but linear trend where higher flowrates resulted in decreasing EF and SR. At a higher flowrate, the solution residence time is lower and thus, less time is available for vapor permeation [229]. Also, Woldemariam et al. reported that the pilot trail of AGMD consisting of five cascades and ten membrane modules (i.e., two membrane modules in each cascade, denoted by a and b) showed a similar rejection performance for all PhACs. However, a slight variation was observed for metoprolol and citalopram by modules 4a and 5a which was detected in the permeate during trail 3. The reason was unclear, though, as all modules were equipped with PTFE membranes with polypropylene supports and similar characteristics in terms of active membrane area, average pore size, porosity, and thickness [77].

Likewise, Silva et al. [224] studied the performance of two commercial PTFE membrane (FGLP Fluoropore®, Millipore) module designs for the removal of diphenhydramine, which was used as a model organic micropollutant. Two different feeds of DI water and synthetic seawater containing diphenhydramine were tested in a DCMD configuration, with the difference in the two module designs being the direction of feed stream entering the MD cell: perpendicular (W-cell) or parallel (H-cell) to the membrane. The results showed that the removal of diphenhydramine, assessed by its detection limit concentration in the permeate, was achieved for both feed types regardless of the module design used. The H-cell achieved better solute rejection in the case of diphenhydramine-containing synthetic seawater feed, however, the W-cell showed a 2-fold higher permeate flux and a 7-fold higher diphenhydramine concentration in the retentate than the H-cell. Because of the arrangement of the inlet streams, the authors reported that the better performance of the W-cell might be attributed to the lower temperature and concentration polarization effects due to a decrease in the thickness of both the temperature and concentration boundary layers adjacent to the membrane surface. However, it was found that the arrangement of the W-cell led to a higher possibility of membrane wetting. The aforementioned results were for a 1 h experiment that showed a better performance and higher concentration of diphenhydramine in the retentate for the W-cell; however, when both configurations were compared for the same obtained permeate volume and a synthetic seawater feed containing only salts (no addition of diphenhydramine), the W-cell showed a lower performance with 12 times higher salt passage to the permeate than the H-cell.

3.4.3. Effects of membrane properties on MPs rejection

In addition to the aforementioned characteristics and conditions, membrane properties, such as material/polymer, pore size distribution, thickness, porosity, charge, and hydrophobicity, are also expected to affect MD performance. In consistence with the above findings, the use of different membrane materials (PTFE and PVDF) exhibited different rejection (100% and 90%, respectively) for ibuprofen, which has low volatility. The transport of ibuprofen through the membrane could be governed by the hydrophobic interactions between the organic ibuprofen and the membrane polymer [226]. During the treatment of diclofenac, ibuprofen, and naproxen from primary and secondary wastewater treatment effluents in a PMR-DCMD arrangement, the removal efficiency of photolysis-DCMD was greater than photo-catalysis-MD with different TiO_2 loading conditions (i.e., 0.5, 1, 1.5 gdm^{-3}) for the primary effluent. The removal efficiency was increased with increasing TiO_2 loading when the secondary effluent was treated during the first hour of operation. Overall, photolysis-MD exhibited effective drugs removal from both effluents during the first hour interval. However, equal performance of photolysis-MD and photo-catalysis-MD was observed for the secondary effluent after 5 hrs of operation [227]. Similarly, slight variations were depicted in the photo-catalysis degradation of ibuprofen from tap water using a PMR-MD arrangement when the operating mode (batch and continuous process) was changed, presumably due to the change in the feed volume (i.e., volume of feed decreased with the time in a batch process, but remained constant in a continuous process) [230].

Compared to other mature membrane separation technologies (RO, NF, and UF), MD technique is relatively new, despite being known and applied in different domains for over forty years since its first discovery. MD is still going under further research and development to achieve better performance and efficiency, which ultimately aims for its commercialization at a larger scale. Despite the growing interest in MD application for different domains, there still lacks sufficient understanding of MD's applicability for MPs removal, and very few studies have been performed on the applicability of MD for MPs removal so far. Although the amount of research in this area is steadily growing, more studies are necessary to understand the mechanisms involved in MD processes and the interactions between the MD membrane and emerging contaminants. Different membrane materials (PVDF and PTFE), configurations (flat-sheet and hollow fiber), and properties (pore size, porosity) were experimented and resulted in different removal efficiencies, however, no studies were performed to evaluate the specific effects of membrane properties on MPs removal. Moreover, all the studies examining MD for MPs removal were performed using commercially available microporous membranes which are commonly used for most MD domains. The performance of in-house membranes made with properties specifically designed for the removal of such emerging pollutants has not been studied yet. This opens a clear room for future research towards membrane development for MPs removal by MD. Lastly, contradicting results were reported on membrane fouling caused by humic acid [231,232], thus, further investigations of membrane fouling by NOM in MD and the interaction mechanism of MPs with deposit membrane fouling (i.e., cake layer) are also required. The studies focusing on the removal of MPs from MD membrane are summarized in Table 6.

Table 6. Removal of MPs by MD

MD Config.	Membrane Config. & Pore size	Membrane material	Capacity	Thickness & Porosity (%)	Feed	Contaminants & Rejection	Removal Mechanism	Ref
DCMD	Flat-sheet 0.22 µm	PTFE supported (commercial Millipore Fluoropore®)	Lab scale	150 63 ± 2	Synthetic seawater for DP, and seawater for the other Pharmaceuticals	Diclofenac (99.8%), Azithromycin (99.9%), Clarithromycin (99.6%), Erythromycin (99.8%), DP below detection limit of 0.03 ng/L	Volatility and charge Thermal degradation Thermal degradation Thermal degradation Charge	[224]
AGMD	Flat-sheet 0.2 µm	PTFE supported (commercial)	Pilot scale	200 µm 80	Municipal wastewater treatment plant effluent after standard biological treatment	Diclofenac (99%), Atenolol (99%), Carbamazepine (99–100%), Ciprofloxacin (37-99%), Estradiol (70–98%), Estriol (76–87%), Estrone (66–86%), Ethinylestradiol (72–90%), Hydrochlorothiazide (99–100%), Ibuprofen (95–98%), Ketoprofen (92–98%), Metoprolol (100%), Naproxen (62–95%), Norfloxacin (60–98%), Progesterone (67–83%), Propranolol (96–100%), Ranitidine (89–100%), Sulfamethoxazole (92–99%), Trimetoprim (80–99%).	Volatility and hydrophobicity	[77]
DCMD	Flat-sheet 0.2 µm	PTFE supported (commercial General Electric)	Lab scale	179 µm 70-80	Wastewater reverse osmosis concentrate	Propylparaben (50%), Salicylic acid (86%), Benzophenone (62%), Triclosan (83%), Bisphenol A (84%), Atrazine (88%). Other 14 MPs (96-99%)	Detected in permeate because of high hydrophobicity, high volatility, and electrostatic interactions *	[211]

Hybrid photocatalysis-DCMD	Capillary (9 membranes module) 0.2 µm	PP (commercial Membrana GmbH, Wuppertal, Germany)	Lab scale	d_{out}/d_{in} = 2.6/1.8 mm	Tap water contaminated with ibuprofen sodium salt (IBU)	Ibuprofen sodium salt (IBU) 100%	Photo-catalysis degradation	[230]
DCMD	Flat-sheet 0.22 µm	PVDF unsupported (commercial Durapore® GVHP, Merck Millipore Ltd)	Lab scale	104 µm *	Antibiotic-containing wastewater	Cefazolin -Negative charge- 100%, Cefotaxime -Negative charge- 100%, Amoxicillin trihydrate -Negative charge- 100%, Cephalothin -Negative charge- 100%, Ceftazidime hydrate -Negative charge- 100%, Piperacillin -Negative charge- 100%, Cloxacillin monohydrate - Negative charge- 100%, Antimony (III) isopropoxide - Negative charge - 100%, Gentamicin sulfate -Positive charge - 86%, Tobramycin -Positive charge - 78%, Ciprofloxacin -Neutral- 100%, Enrofloxacin -Neutral- 100%	Electrostatic interaction/Charge	[8]
DCMD (1) And hybrid MBR-MD (2)	Flat-sheet 0.22 µm	PTFE (commercial GE, Minnetonka, MN)	Lab scale	* 70	Water for DCMD and Municipal wastewater (MBR effluent with TrOCs introduced to the MBR feed) for MBR-MD	DCMD: 4-tert-octylphenol (54%), Benzophenone (66%), 4-tert-butylphenol (73%), Oxybenzone (81%), Octocrylene (81%), Other 25 TrOCs (>90%) MBR-MD: All 29 TrOCs had complete or near complete removal (>95%)	Volatility	[226]
Lab scale vacuum MD	Tubular (3 membranes module) 0.2 µm	PP (commercial Enka Microdyn, North Carolina)	Lab scale	1.5 mm *	Water contaminated with Benzene	Low benzene rejection	Detected in permeate due to evaporation and diffusion (volatility)	[233]

Lab scale DCMD	Capillary (40 membranes module) 0.2 µm	PP (commercial MD020-CP-2N, Microdyn, Germany)	Lab scale	*	Simulant wastewater (mixture of humidity condensate and urine) (1) or doubly deionized water DDW (2), spiked with both estrone and estradiol	Urea and ammonia (99.9%) for feed (1), Estrone and Estradiol (>99.5%) for feed (2).	Volatility	[186]
Lab scale PMR consisting of hybrid photocatalysis-DCMD	Capillary (9 membranes module) 0.2 µm	PP (commercial Membrana GmbH, Wuppertal, Germany)	Lab scale	*	Primary effluents, PE (1) and secondary effluents, SE (2) of municipal wastewater treatment plant, spiked with 100 µg dm^{-3} of each sodium salt	Diclofenac (100%), Ibuprofen (73%), and Naproxen (90%) for feed (3). Diclofenac (100%), Ibuprofen (93%), and Naproxen NAP (94%) for feed (4).	Photo-catalysis degradation	[227]
Lab scale DCMD	Flat-sheet 0.22 µm	PVDF (commercial Durapore GVHP; Merck-Millipore)	Lab scale	125 µm 75	Synthetic surface water in a pH range of 2.6-11 (1), and NEWater brine (i.e., reverse osmosis concentrate from a local wastewater treatment plant in Singapore) (2).	Ibuprofen (100%) for feed (1) with and without presence of HA. Ibuprofen (100%) for feed (2).	Volatility and hydrophobic interaction	[228]
Solar thermal liquid-gap MD	Spiral wound 0.2 µm	PTFE (commercial)		70 µm 80	Demineralized water for the experiments with Escherichia coli and Fusarium solani. Municipal wastewater treatment plant effluent for the experiment with Clostridium sp. spores.	Fusarium solani (100%), Clostridium sp. Spores (100%).	*	[234]
DCMD unit (Convergence,	Flat-sheet 0.2 µm	PTFE (commercial, General	Bench scale	179 µm 70-80	Four synthetic feed solutions (A- with DI water, B- with humic acid, C- with	Atrazine (>97%), Clofibric acid (97-99%) Dichlorvos (10-60%), Phorate (10-50%),	Volatility and hydrophobic interaction	[235]

The Netherlands)		Electric, USA)			inorganic salts and D- with humic acid and inorganic salts), containing 200 µg/L of each pesticide	Parathion-methyl (60-80%).				
DCMD	Flat-sheet 0.22 µm	PTFE (commercial, Sterlitech)	Lab scale	* 70	Synthetic feed solution composed by DI water, 1 µg/L of 25 PhACs, and different concentrations of humic acid	Betamethasone (>99%), All other 24 PhACs (below detection limit)			(1) Volatility and hydrophobic interaction (2) Adsorption and electrostatic interaction	[236]
			Lab scale			TrOC	After bioreactor	After MD		
DCMD–thermophilic bioreactor (MDBR) system	Flat-sheet 0.22 µm	PTFE (commercial, GE, Minnetonka, MN)		175 µm 70	MD feed was the mixed liquor of the bioreactor. The bioreactor inoculated with activated sludge from the Wollongong Wastewater Treatment Plant (Wollongong, Australia) and used a synthetic wastewater to simulate medium strength domestic wastewater.	Clofibric acid	30%	99%	Biodegradation governed the removal of most TrOCs by the bioreactor Physical separation by MD (volatility) governed the removal of recalcitrant TrOCs	[237]
						Salicylic acid	95%	95%		
						Ketoprofen	96%	99%		
						Fenoprop	32%	98%		
						Naproxen	97%	100%		
						Ibuprofen	85%	100%		
						Primidone	62%	100%		
						Diclofenac	21%	90%		
						Gemfibrozil	63%	99%		
						Propoxur	90%	100%		
						Carbamazepine	5%	93%		
						Pentachlorophenol	80%	95%		
						Estriol	98%	99%		
						Atrazine	30%	98%		
						Ametryn	86%	99%		
						Benzophenone	80%	98%		
						Amitriptyline	98%	99%		
						4-Tert-butyphenol	97%	98%		
						Oxybenzone	98%	100%		
						Estrone	99%	99%		
						17α-Ethinylestradiol	97%	100%		
						17β -Estradiol	99%	100%		
						Triclosan	50%	98%		

					17β-Estrodiol-17-Acetate	99%	100%	
					Octocrylene	96%	97%	
Hybrid Photolysis – DCMD	Capillary (9 membranes module, Accurel PP S6/2,) 0.2 µm	PP (commercial Membrana GmbH, Wuppertal, Germany)	d_{out}/d_{in} = 2.6/1.8 mm *	Synthetic feeds containing Ibuprofen sodium salt as the model Compound with initial concentration of 0.05, 0.1, 0.2 or 0.4 mmol/dm3.	Ibuprofen by MD alone (99.95-99.89%), Ibuprofen by Photolysis-MD (99.97-99.99%)		Volatility Photodegradation	[238]
					TrOC	MD alone	AnMBR-DCMD	
Hybrid AnMBR -DCMD	Flat-sheet (Plate and frame module) 0.2 µm	PTFE (commercial, Porous Membrane Technology, Ningbo, China)	60 µm 80	MD feed was the permeate of the bioreactor. The bioreactor inoculated with digested sludge from a full-scale wastewater treatment plant and used a synthetic wastewater to simulate high strength domestic wastewater.	Caffeine	98%	100%	Volatility [239]
					Sulfamethoxazole	85%	90%	
					Ketoprofen	98%	100%	
					Trimethoprim	97%	100%	
					Paracetamol	98%	100%	
					Naproxen	98%	100%	
					Primidone	98%	98%	
					Ibuprofen	98%	100%	
					Triamterene	98%	99%	
					Carazolol	95%	98%	
					Tris(2-chloroethyl) phosphate	97%	97%	
					Diclofenac	75%	75%	
					Carbamazepine	90%	90%	
					Gemfibrozil	98%	100%	
					Simazine	95%	90%	
					Amtriptyline	75%	100%	
					Atrazine	80%	75%	
					Diuron	98%	98%	
					Propylparaben	90%	90%	
					Linuron	90%	95%	
					Clozapine	95%	98%	
					Phenylphenol	51%	80%	
					Bisphenol A	90%	90%	
					Diazinon	52%	98%	
					Triclosan	85%	85%	
					Triclocarban	75%	97%	

DCMD-EMBR (enzymatic membrane bioreactor)	Flat-sheet 0.2 µm	PTFE supported (commercial, GE, Minnetonka, MN)	Lab scale	175 µm (active layer 5 µm) 70	MD feed was the media of the bioreactor. The bioreactor used synthetic wastewater containing a mixture of 30 phenolic and non-phenolic TrOCs.	4-tert-octylphenol, Octocrylene, 4-tert-butylphenol, Benzophenone, and Oxybenzone by MD alone (54-70%), 4-tert-octylphenol, Octocrylene, 4-tert-butylphenol, Benzophenone, and Oxybenzone by MD-EMBR (>99%), All other 25 TrOCs by MD-EMBR (94-98%)		Rejection by MD membrane (volatility), biocatalytic degradation, and molecular properties of MPs	[240]
DCMD-EMBR	Flat-sheet 0.2 µm	PTFE supported (commercial, GE, Minnetonka, MN)	Lab scale	175 µm (active layer 5 µm) 70	MD feed was the media of the bioreactor. The bioreactor used a mixture of 30 phenolic and non-phenolic TrOCs (each at 20 µg/L) in Milli-Q water.	TrOC	After Enzymatic degradation	After MD	Volatility, enzymatic degradation, and molecular properties of MPs	[241]
						Ketoprofen	40%	99%		
						Diclofenac	45%	98%		
						Naproxen	46%	99%		
						Atrazine	48%	93%		
						Fenoprop	50%	97%		
						Carbamazepine	52%	98%		
						Ibuprofen	54%	99%		
						Clofibric acid	55%	99%		
						Primidone	57%	99%		
						Propoxur	57%	98%		
						Gemifibrozil	58%	99%		
						DEET	68%	97%		
						Ametryn	72%	99%		
						Metronidazole	94%	97%		
						Benzophenone	97%	99%		
						Amitriptyline	97%	98%		
						Octocrylene	98%	99%		
						Salicylic acid	55%	99%		
						Pentachlorophenol	62%	99%		
						Enterolactone	72%	99%		
						4-tert-Butylphenol	82%	90%		
						Bisphenol A	83%	98%		
						Oxybenzone	83%	97%		
						Estriol	92%	99%		
						Estrone	93%	99%		
						17ß – Estradiol	93%	99%		

						17 – Ethinylestradiol Triclosan 4-tert-Octylphenol 17ß–Estradiol-17-acetate	95% 98% 99% 99%	99% 99% 99% 99%		
DCMD-EMBR	Flat-sheet 0.22 µm	PTFE supported (commercial, GE, Minnetonka, MN)	Lab scale	175 µm (active layer 5 µm) 70	MD feed was the permeate of the enzymatic bioreactor. The enzyme solutions for EMBR contained a mixture of 1 mL and 0.1 g of A. oryzae and T. versicolor laccases, 5 TrOCs (each at 1 mg/L), HBT and VA mediators (at 1mM concentration) in 1.5L Milli-Q water.	Carbamazepine, Oxybenzone, Diclofenac, Atrazine, Sulfamethoxazole	43-53% 90-99% 82-90% 54-67% 40-46%	>90% >90% >90% >90% >90%	Enzymatic degradation	[242]
DCMD	Hollow fiber (shell and tubular format X-50, membrana, USA)	*	Lab scale	The “shell” portion of the module was a 1/4 ID×3 in. long brass threaded pipe fitting	Pure water with 1 mg/L of each of four pharmaceutical active ingredients (at 90 °C)	Enrichment factor (EF): Ibuprofen (5.6), Dibucaine (3.6), Acetaminophen (3.6), Diphenhydramin (3.1) Solvent reduction (SR): Ibuprofen (48%), Dibucaine (40%), Acetaminophen (35%), Diphenhydramin (47%)		Rejection by MD membrane (volatility)		[229]

Cells with * sign correspond to a lack of data

4. Conclusion and Future Research Trends

The extensive use of PhACs, PCPs, and EDCs, leading to the widespread occurrence of MPs from ngL^{-1} to μgL^{-1} in water, is a menace to the global terrestrial and aquatic environment. The inadequacy of conventional water/wastewater treatment systems for removing MPs from aquatic bodies has presented a great challenge for water managing authorities and has led to the consideration of the most appropriate technologies to deal with these emerging concerns. The overview of existing studies show that it is difficult to draw a general conclusion based on a comparison of the MPs removal performances of different membrane technologies due to their different working principles (i.e., pressure gradient in RO/NF/UF, osmotic gradient in FO, and thermal gradient in MD), the wide range of MPs and their characteristics (i.e., MW, pKa value, dipole moment, volatility, and hydrophobicity), and diverse operational parameters (i.e., feed type, feed pH, temperature, pressure, draw solution concentration, etc.). However, the following conclusions can be drawn from the thorough review provided herein:

- In general, RO and NF are able to remove a wide spectrum of MPs based on the size exclusion mechanism. Compounds with relatively large MW can usually be well removed by RO and tight NF membranes. Additional mechanisms, such as charge interaction, dipole interaction, and hydrophobic interaction, can also play important roles. In this respect, small molecules with the opposite charge to the membrane surface, large dipole moments, or high log K_{ow} values tend to show low rejection.
- The rejection mechanisms involved in FO are mostly similar to those of RO and tight NF. Additionally, the RSF phenomena in FO also contributed positively toward MPs rejection. Further trends can be developed to investigate the effect of RSF in different configurations.
- The formation of a fouling layer resulted in an improved sieving effect in FO, RO and tight NF. However, for loose NF membranes, their performance deteriorated due to the concentration polarization effect. Since size exclusion was not the dominant rejection mechanism for loose NF, the formation of a fouling layer resulted in membrane charge neutralization, which affected/minimized the electrostatic contribution arising from the negatively-charged membrane surface.
- In UF, adsorption was the predominating removal mechanism, since the MWCO value of UF was larger than the molecular size of MPs. Therefore, MPs with high hydrophobic value were more effectively rejected. However, this phenomenon was also affected by saturation.
- Non-volatile MPs were effectively removed by MD (100% in most cases), while declined rejection was observed for volatile MPs. The effect of operating temperature on the degradation/by-product formations needs to be further investigated.
- The functionalization of nanomaterials over the active layer of the membrane led to better rejection performances in contrast with their incorporation in the polymeric surface. Since the diffusion of MPs occurs due to the affinity between hydrophobic moieties and the membrane's active layer, the incorporation of nanomaterials could improve flux more so than the rejection of MPs.

- Although the physicochemical characteristics of MPs, operating conditions, and membrane properties had a significant influence on MPs rejection, complete and near-complete removal were explicated for RO, FO, MD, NF, and UF membranes. For MPs with high volatile characteristics, the rejection was depicted as RO > MD~FO~NF > UF.

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