

Removal of emerging organic micropollutants via modified-reverse osmosis/nanofiltration membranes: A review

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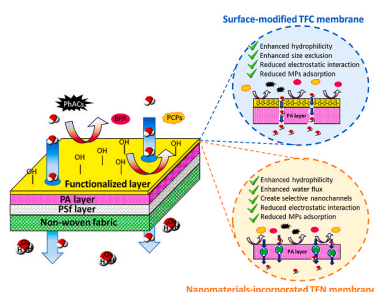
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HIGHLIGHTS

- Occurrence, risks and type of emerging micropollutants (MPs) have been identified.
- Removal of emerging MPs via RO and NF membranes have been summarized.
- TFN and surface-modified TFC membranes showed remarkable MPs removal rate.
- The MPs rejection mechanisms of polyamide RO/NF membranes were elucidated.

GRAPHICAL ABSTRACT



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ABSTRACT

Hazardous micropollutants (MPs) such as pharmaceutically active compounds (PhACs), pesticides and personal care products (PCPs) have emerged as a critical concern nowadays for acquiring clean and safe water resources. In the last few decades, innumerable water treatment methods involving biodegradation, adsorption and advanced oxidation process have been utilized for the removal of MPs. Of these methods, membrane technology has proven to be a promising technique for the removal of MPs due to its sustainability, high efficiency and cost-effectiveness. Herein, the aim of this article is to provide a comprehensive review regarding the MPs rejection mechanisms of reverse osmosis (RO) and nanofiltration (NF) membranes after incorporation of nanomaterials and also surface modification atop the PA layer. Size exclusion, adsorption and electrostatic charge interaction mechanisms play important roles in governing the MP removal rate. In addition, this review also discusses the state-of-the-art research on the surface modification of thin film composite (TFC) membrane and nanomaterials-incorporated thin film nanocomposite (TFN) membrane in enhancing MPs removal performance. It is hoped that this review can provide insights in modifying the physicochemical properties of NF and RO membranes to

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achieve better performance in water treatment process, particularly for the removal of emerging hazardous substances.

1. Introduction

Over the past two decades, the tremendous growth of global population and industrial expansion has significantly increased the production of emerging contaminants (ECs). ECs (also known as micropollutants (MPs)) are primarily synthetic organic/inorganic chemicals that possess deleterious effects not only to human life, but also to the entire ecosystem (Ahmed et al., 2017; Olmez-Hanci et al., 2020). The incessant existence of these organic micropollutants can be mostly found in the wastewater, landfills, daily household products, hospital, municipal sewage, pharmaceutical production plants and natural aquatic environment (Klamerth et al., 2012; Prieto-Rodriguez et al., 2012). Typically, MPs are present in water bodies at trace concentration ranging from $\mu\text{g/L}$ (equivalent to ppb) to ng/L (equivalent to ppt) (Tran et al., 2013). As most of the MPs are not able to be eliminated by conventional wastewater treatment process, the presence of them in treated water is likely to accumulate in plants and soil which eventually intrudes into human endocrine system (Belhaj et al., 2015; Klamerth et al., 2013; Luo et al., 2014b). Stumm-Zollinger and Fair (1965) and Tabak and Bunch (1970) first raised the awareness regarding the adverse effects of non-biodegradable pharmaceutical active compounds (PhACs) in municipal wastewater in the 1960s–1970s.

Currently, the complexities of MPs have gained significant attention from researchers/governments worldwide in terms of their potential adverse effect on the environmental and human health (Hashim et al., 2021). For instance, government and non-government agencies including the World Health Organization (WHO), European Union (EU), International Program of Chemical Safety (IPCS) and North American Environmental Protection Agency (EPA) are taking initiatives by setting up legal frameworks to enhance and protect the quality of freshwater resources (Esplugas et al., 2007). Numerous advanced technologies including activated carbon (AC), ozonation, adsorption biodegradation and ultraviolet (UV) irradiation have been studied to remove the MPs from water (Deegan et al., 2011; Han et al., 2012; Jung et al., 2013; Rathi and Kumar, 2021; Suman Raj and Anjaneyulu, 2005). However, removing MPs using these approaches are still challenging due to several limitations, such as high energy consumption and huge maintenance and operation cost (Ahmed et al., 2017; Katibi et al., 2021). It is also not easy to access the removal rates of wide range of MPs in wastewater treatment plants since the removal mechanism is strongly dependent on many physicochemical properties of MPs including hydrophobicity, pKa, size, shape and charge (Kim et al., 2018).

The effective removal of MPs has been a major area of NF and RO membrane for producing portable water. NF and RO are pressure driven process where the membranes retain certain ions, particles or molecules depending upon membrane surface charge, pore size and chemical affinity. Moreover, NF possesses molecular weight cut-off (MWCO) of 200–1000 Da with pore size of 1 nm, while RO membrane with MWCO less than 200 Da and pore size less than 1 nm, are effective in removing dissolved ions and small organic molecules (Gohil and Ray, 2017). Due to these properties, the demand of NF and RO membranes are gradually elevating in different separation applications (liquid/molecular separation) especially for wastewater and water treatment. The retention of MPs by NF membrane mainly depends on the physicochemical properties of MPs, which can be affected by solution chemistry (etc., pH of solution). While for the permeation of MPs through RO membranes, it could be affected by the adsorption of MPs onto the membrane surfaces and the dissolution of MPs molecules into membrane, and subsequently diffuse through the membrane matrix (Kim et al., 2018).

Membrane technology has been used to remove MPs from water/wastewater and this research topic is receiving great attention in recent

years (Nasseri et al., 2018; Wang et al., 2018). Several dedicated review papers have reported about the use of membrane technology for the MPs removal (Dharupaneedi et al., 2019; Khanzada et al., 2020; Kim et al., 2018). Nevertheless, the content of these reviews were mostly focused on the membrane technologies such as ultrafiltration (UF), NF, RO and forward osmosis (FO) (Dharupaneedi et al., 2019; Khanzada et al., 2020; Kim et al., 2018). Although the removal of MPs from water using commercial TFC membrane have been commonly reported, yet there is limited dedicated review on the recent trend in removing MPs using surface-modified TFC membrane and nanomaterials-incorporated TFN membrane. Hence, the MPs rejection mechanism and rejection governing solute parameters for TFC and TFN membranes is of paramount importance for the accurate selection of materials and membrane process design. Therefore, this article intends to provide an overview on the removal of MPs through TFN and surface-modified TFC membranes by correlating their physicochemical properties with the removal mechanism of MPs. The sources of MPs as contaminants, classification of MPs and their adverse impacts on the environment are also reviewed and discussed.

1.1. Sources of MPs

In general, MPs can exist in the environment via numerous pathways, such as household domestic water, wastewater treatment effluent discharged from manufacturing industries and hospitals, agricultural activities and landfill runoff as depicted in Fig. 1 (Price et al., 2010). These pollutants can intrude directly towards the aquatic from the effluent outflow and indirectly as run-off. Among these, the major point sources that distribute the MPs into the environment are wastewater from factories and hospitals (Liu and Wong, 2013). Reports have indicated that MPs are hardly removed by conventional treatment methods (Gulkowska et al., 2008, 2007; Hua et al., 2008; Leung et al., 2012; Yang et al., 2010). Such treated/untreated effluents are possibly discharged into the rivers and coastal with the concentrations of MPs ranging from ng/L to $\mu\text{g/L}$ (Liu et al., 2005; Mulla et al., 2018; Pojana et al., 2007). In certain situations, the treated portable-water resources might be polluted by surface outflow and deep-well injection of the effluent (Mompelat et al., 2009). This indicates that drinking water is not free from these MPs. In fact, esteroidal hormones and plasticizers were previously detected in portable water, groundwater and surface water (Colin et al., 2014; Ferreira et al., 2020; Katibi et al., 2021; Lee et al., 2015; Sornalingam et al., 2016).

1.2. Classification of MPs

There are several classifications to differentiate MPs. These include PhACs, industrial chemicals, pesticides and personal care products (PCPs). All of these are bioaccumulative and bioactive and have a pervasive perseverance. Antibiotics, hormones, anti-epileptic drug, blood pressure regulator, lipid regulator, painkillers, steroids, beta-adrenergic blocking agents and anti-inflammatory drugs can be categorized as PhACs products (Shahid et al., 2021). Meanwhile, examples of PCPs products are anti-microbial agents, preservatives, sunscreens and disinfectants. Triclosan is one of the anti-microbial agents that can be commonly found and identified from toothpaste, washing soaps, detergents and shampoos. Also, triclosan can be used in utensils, toys and cutting boards (Li et al., 2020). In addition, plasticizers and fire retardants are classified as industrial chemical products while insecticides, herbicides, fungicides are fall under the category of pesticides. [28]. A summary of several categories and sub-categories of MPs is presented in Table 1.

1.3. Effects of MPs on human health and wildlife

The occurrence of MPs in fresh water is a rising concern attributed to their multitude adverse effects on the ecosystem and human health. The human especially children and infants suffer the highest threats when exposed to MPs. Children would experience the most critical physiological systems development. The exposure of MPs towards the children can cause hormonal imbalance, inducing a lot of side effects in the future such as prostate and breast cancer, thyroid, fertility and pregnancy problems (Vieira et al., 2021).

According to a study conducted on post-mortem brain material acquired from 24 individuals (12 normal-weight and 12 obese with body mass index lower than 25 kg/m² and higher than 30 kg/m², respectively), seven types of endocrine disrupting chemicals (EDCs) such as BPA, triclosan, triclocarban and methyl-, ethyl-, n-propyl- and benzyl paraben were detected in the hypothalamus brain tissues (Van Der Meer et al., 2017). Likewise, an environmental working group from Oakland, California found out that about 16 toxic MPs including fragrance, preservatives and antimicrobial compounds were detected in the urine and blood samples of 20 teenage girls, aged between 14 and 19 (EWG, n.d.). Meanwhile, plasticizers such as BPA and 4-n-nonylphenol (4-NP) are often notable in drinking water, surface sediment, aquatic organism and treated wastewater effluent as reported elsewhere (Křesinová et al., 2018).

The existing of EDCs can alter the behavior and obesity of children and further diminish the gonadal development. It must also be pointed out that these persistent pollutants not only affect adult individual, but also cause offspring via the possible mechanism of transgenerational epigenetic inheritance (Skinner et al., 2011). Obesity is currently one of the significant diseases that growing continuously and become a worldwide health concern issue, especially in adolescents and children. This is likely due to the fat-soluble nature of EDCs that accumulates in adipose tissue, interrupting the metabolism process of human (Magueresse-Battistoni et al., 2017; Petrakis et al., 2017).

Due to the epigenetic heritage, mothers who have been exposed to dichlorodiphenyltrichloroethane (DDT) pesticide may also cause their children suffering from obesity disease (Petrakis et al., 2017; Skinner et al., 2013). In addition to DDT, MPs such as polychlorinated biphenyls (PCBs) and BPA are also able to inhibit the insulin function, interrupting

body enzyme and triggering diseases (Arsa et al., 2009). Diabetes is one of the exponentially growing diseases caused by obesity. On the other hand, Alzheimer's disease (AD) can be generated from the Type 2 diabetes owing to the impaired insulin levels (Hoscheidt et al., 2016; Stanley et al., 2016). The exposure to mercury (Hg) and lead (Pb) can also stimulate pathological proteins related to AD.

Research studies also reported the MPs may be associated with the hormonal and health issue towards living organism by inhibiting the regular action of endocrine system, mimicking the actions of endogenous estrogen and binding to estrogen receptors of organism (Cipolletti et al., 2018; Varticovski et al., 2022; Wang et al., 2021; Yoon et al., 2014). All these adverse effects could cause feminization, reproductive disorders and carcinogenesis problem in wildlife (Komesli et al., 2015; Zacharakis et al., 2013). Furthermore, the bioaccumulation and biomagnification of MPs in the marine environment also endangered the aquatic ecosystem such as reduction in fish fertility due to skewed sex ratios, alteration of gonadal development (intersex and imposex) and abnormal blood hormone levels (Katibi et al., 2021). For instance, the reproductive issue and feminization of male fishes can be correlated to the estrogens E1, E2, phthalates and BPA (Matthiessen, 2013). In the 1990s, the EDCs were reported to cause a significant impact on the alligators. The female alligators were found to show changes in their ovaries, with plasma concentration beyond the normal level. The young male alligators suffered from depressed plasma testosterone concentration and reduced phallus size. These anomalies can be attributed to the exposure to the DDT (Jr et al., 1994; Ternes et al., 2002).

1.4. Endocrine disrupting compounds and its removal techniques

Owing to the cumulative effect of MPs in the environment, many techniques have been studied to separate it from the water sources (Vieira et al., 2021). Although conventional wastewater treatment has been widely used in the industries, this technique which is based on biological process is not efficient enough in removing MPs (Luo et al., 2014b). Fig. 2 presents the publications related to various types of treatment techniques that have been studied for MPs removal. Obviously, the membrane technology received the greatest attention compared to other techniques over the past 5 years. Moreover, the advantages and challenges of different processes for the removal of ECs are

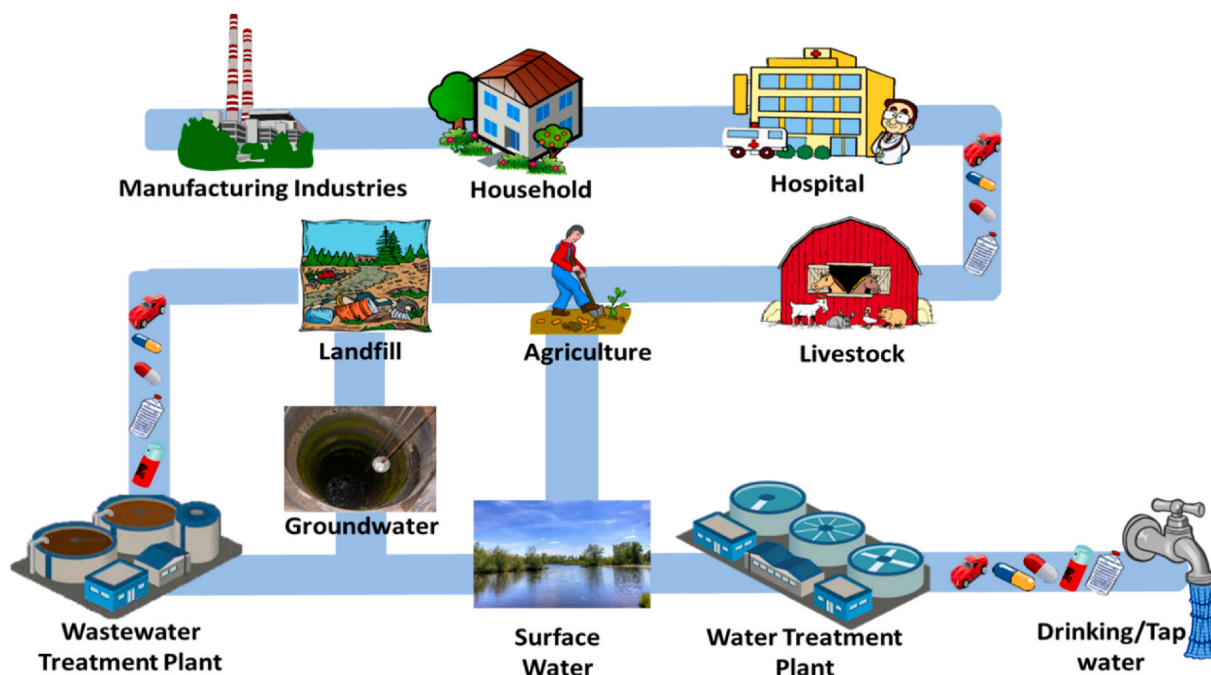
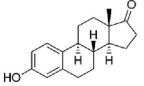
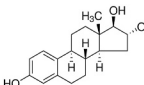
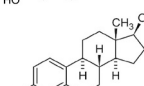
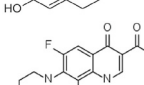
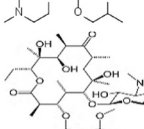
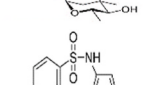
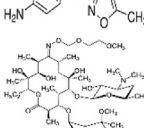
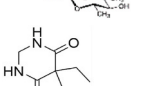
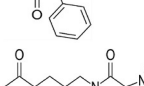
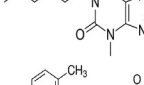
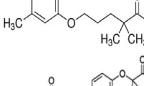
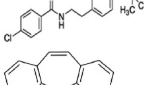
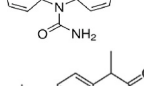
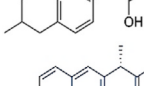
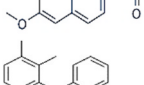
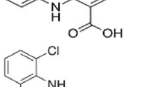
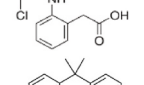
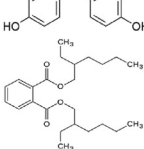


Fig. 1. Typical sources and routes of MPs. Source: Modified from Refs (Katibi et al., 2021; Khanzada et al., 2020).

Table 1

Classification of MPs. The physicochemical properties of MPs were concluded from references (Khanzada et al., 2020; Liu et al., 2009; Taheran et al., 2016).

Groups	Uses	MPs	<i>pka</i>	^a log K _{ow}	MW (g/mol)	Charge at pH 7	Structure
PhACs	Hormones	Estrone	10.5	3.13	270.4	Neutral	
		Estriol	10.4	2.45	288.4	Neutral	
		17 β -estradiol	10.7	4.01	272.4	Neutral	
	Antibiotics	Ofloxacin	5.8	-2	361.4	-	
		Erythromycin	8.9	3.06	733.9	Neutral	
		Sulfamethoxazole	1.7; 5.6	0.89	253.3	-	
	Anti-epileptic	Roxithromycin	9	2.75	837.05	Neutral	
		Primidone	-1; 12.2	0.91	218	-	
		Pentoxifylline	0.97	0.29	278.3	Neutral	
	Lipid regulator	Gemfibrozil	4.45	4.77	250.34	-	
		Bezafibrate	3.44	4.25	361.8	-	
		Carbamazepine	13.0	2.45	236.3	Neutral	
	Anti-inflammatory	Ibuprofen	4.47	3.97	206.3	-	
		Naproxen	4.20	3.18	230.0	-	
		Mefenamic acid	3.80	5.12	241.3	-	
		Diclofenac	4.08	4.51	296.2	-	
Industrial Chemicals	Plasticizers	Bisphenol A	9.6	3.32	228.3	-	
		Di (2-ethylhexyl) phthalate	-	-	390.6	-	

(continued on next page)

also tabulated in Table 2.

1.5. Conventional water treatment process

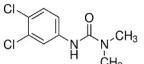
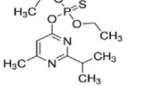
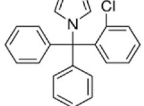
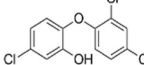
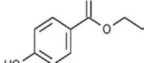
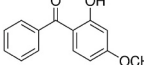
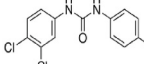
Conventional water treatment plants typically consist of coagulation-flocculation, sedimentation, filtration and lastly disinfection as depicted in Fig. 3. Generally, screening process can be used to remove floating matters such as dried leaves, fallen twigs or other large-sized suspended particles from the water influent. After the screening process, lime is added to soften the water by removing magnesium and calcium ions, followed by the addition of coagulants to neutralize the particle charges and to allow these particles to coagulate to form flocs. Subsequently, the flocs settle to the bottom of sediment basin, while the clean water on top flows through the filtration tank that consists of sand, gravel and charcoal to eliminate fine particles. Lastly, the filtered clean water is disinfected prior to deliver to distribution system (Abu Shmeis, 2018). Hydrophobic MPs and PCPs with colloidal material associated with high organic carbon content are expected to be removed via coagulation process (Westerhoff et al., 2005). However, several findings reported that conventional water treatment plants were unable to remove EDCs and PCPs effectively (Benotti et al., 2009; Ternes et al., 2002). According to Benotti et al. (2009), the removal rate of atrazine, an insecticide, was only <50% through conventional water treatment process. Westerhoff et al. (2005) on the other hand reported that the removal rates of EDCs and pharmaceutical products were lower than 20% during coagulation and lime softening process. Using rapid sand filtration as the treatment method, Petrovic et al. (2003) found that only 7% of MPs such as nonylphenol, nonylphenol monoethoxylates, nonylphenol diethoxylates and nonylphenol were separated from drinking water treatment plants.

1.6. Advanced oxidation process

Advanced oxidation process (AOP) is used in the wastewater treatment plants by introducing ozone into the water bodies to remove the MPs. AOPs such as photolysis mainly focus on using ultraviolet radiation as energy source to degrade the MP compounds. AOP process utilizes highly reactive oxygen species to attack organic compounds and

microbes present in the water bodies (Lu and Astruc, 2020). During AOP process, the hydroxyl radicals (OH) with an oxidation potential of 2.80 V mineralize the organic molecules to CO₂ and H₂O or reduce the MPs into less harmful intermediate products (Gadupudi et al., 2021; Miklos et al., 2018). In general, AOP is efficient in removing the MPs that cannot be treated through biological and conventional physicochemical methods (Vieira et al., 2021). For instance, Liu et al. (2017) revealed that 21.2% of 17β-Estradiol (E2) hormone was removed by photodegradation. The authors claimed that a variety of active oxygen species such as hydroxyl radicals and singlet oxygen generated under natural light in surface waters can degrade E2 and other estrogenic compounds. Nevertheless, there are some disadvantages for traditional applications related to AOP, such as high operating costs, requires expensive catalysts or co-catalysts and form highly toxic secondary products which could lead to environmental pollution. Photocatalysis, which also one kind of promising solar-driven AOP technology, has attracted interest in recent years. Photocatalysts can be activated by radiation, producing highly reactive photo-induced charge carriers to react with MP compounds (Liu et al., 2020). Vela et al. (2019) demonstrated the photocatalytic degradation of wastewater effluent using hydroxyl radical- and sulphate radical-based AOP. Zinc oxide (ZnO) was used as photocatalyst and sodium persulfate (Na₂S₂O₈) as oxidant. The integration of ZnO/Na₂S₂O₈ strongly enhanced the degradation rate of malathion and fenarimol pesticides compounds up to 90%. However, there are several drawbacks for single ozonation process including selective reaction between ozone and organic compounds, lower solubility and stability of ozone in water (Wang et al., 2019a). In order to enhance the efficient removal of MPs from the effluent, homogeneous or heterogeneous catalysts could be added during the ozonation process (Shahid et al., 2021; Wang et al., 2019a). The presence of catalysts is able to enhance the decomposition of ozone and generate higher amount of hydroxyl radicals (Faria et al., 2009). Numerous catalysts such as activated carbon (Faria et al., 2008), metal oxides (Hu et al., 2008; Radhakrishnan and Oyama, 2001; Wang and Bai, 2017), alumina (He et al., 2010) and natural minerals (Wu et al., 2016) were applied in the catalytic ozonation process. However, the role of different catalysts highly depends on their mechanism involved in reaction, reaction conditions and nature of contaminants.

Table 1 (continued)

Groups	Uses	MPs	pKa	^a log Kow	MW (g/mol)	Charge at pH 7	Structure
Pesticides	Herbicides	Diuron	2.68	–	233.1	Neutral	
	Insecticides	Diazinon	2.6	3.8	304.4	–	
	Fungicides	Clotrimazole	–	–	344.8	–	
Personal care products (PCPs)	Anti-microbial agents	Triclosan	7.8	5.17	289.5	Neutral	
	Preservatives	Propyl-paraben	8.5	3.04	180.2	–	
	Sunscreens	Oxybenzone	–	3.79	228.0	–	
	Disinfectants	Triclocarban	11.4	–	315.6	Neutral	

^a compound is hydrophobic when log Kow >2.

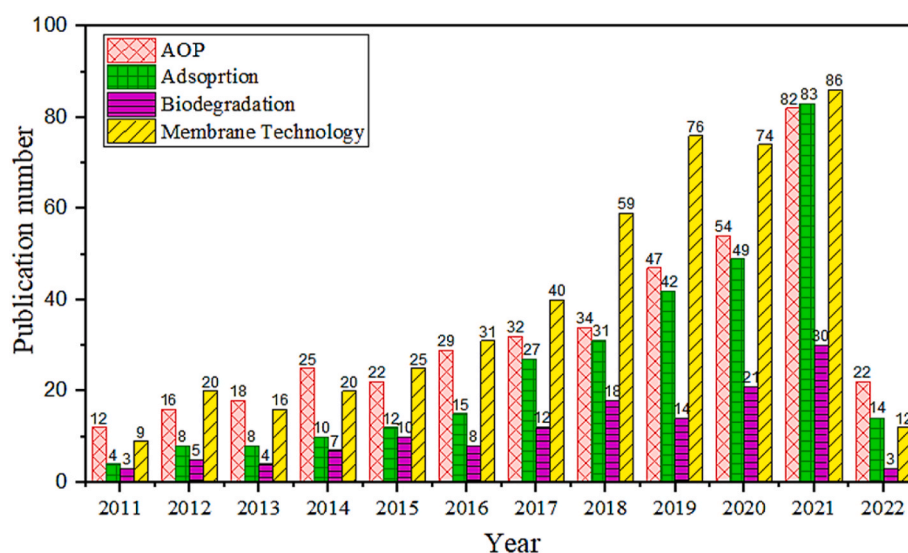


Fig. 2. Publications related to different techniques used for the MPs removal. The data were obtained from Scopus between 2011 and 2022, with further manual screening to differentiate the techniques used, Date: Jan 7, 2022).

For example, the catalytic activity of alumina is significantly reduced in the presence of phosphate, humic acid and 1,2,4-trichlorobenzene. It is found that phosphate species would attack hydroxyl groups on the alumina surface during ozonation process, which serve for the ozone decomposition (Ikhlaiq et al., 2012; Qi et al., 2009).

1.7. Adsorption

Adsorption is considered as one of the mainstream methods applied for removing both organic and inorganic contaminants from water (Shahid et al., 2020; Umpierrez et al., 2017). This process offers several advantages such as eco-friendly operation, relatively low operation cost and simple operational scheme (Shahid et al., 2021, 2019; Zhang et al., 2021). Adsorption is a surface phenomenon whereby adsorbents are used to adsorb the MPs (adsorbate). The adsorbate can be absorbed via mass transfer mechanism between two phases such as gas-liquid, liquid-liquid, gas-solid and liquid-solid interface (Shahid et al., 2021). Generally, MPs can be adsorbed by adsorbents via $\pi-\pi$ interaction, intermolecular forces, electrostatic attraction, hydrophobic interfaces and van der Waals forces. The commonly used adsorbents for MPs removal are silica gel (Qu et al., 2018; Wu et al., 2019), activated carbon (Esmaeeli et al., 2017; Leite et al., 2018; Wong et al., 2018), alumina (Zhao et al., 2015), zeolite (Jiang et al., 2020), polyacrylamide (Peng et al., 2019), biochar (Tomul et al., 2020) and absorbent resin (Wang et al., 2019b). Each material shows different adsorption properties and mechanisms due to the differences in the physicochemical properties. The discrepancy has led to different removal efficiency of pollutants. For instance, Ahsan et al. (2018) reported that the adsorption capacity towards BPA can reach up to 236.80 mg/g using sulfonic acid functionalized carbonaceous adsorbent (TW-SO₃H) extracted from tea leaves. On the other hand, Lu et al. (2017) reported BPA adsorption capacity of 9.68 mg/g using *Ulva prolifera* based biochar. Although activated carbon consists of strong adsorption capacity and large specific surface area, the production cost is relatively high and environmental unfriendly (Menya et al., 2018). Adsorbents such as alumina, silica gel and adsorption resin show narrow adsorption surface.

1.8. Removal of EDCs via membrane technology

Membrane bioreactor (MBR) technology has been increasingly used to remove the MPs from municipal and industrial wastewater treatment owing to its high effluent removal efficiency (Abegglen et al., 2009; Luo

et al., 2014a, 2014b). MBR is a hybrid technique that integrates biodegradation of a conventional activated sludge process with a membrane separation process. The enhanced adsorption through improved sludge characteristic, better biodegradation with high sludge age and high removal efficiency via membrane process have enabled MBR to become one of the promising MPs removal technologies. Typically, microfiltration (MF) membranes with the pore size of 0.4 μ m are utilized as part of the MBR process to filter out microorganism flocs ranged from 10 to 100 μ m (Taheran et al., 2016). In term of biodegradation, the sorption of MPs against sludge flocs and microbial products (etc., algae, fungi and bacteria) is vital in dictating the removal of MPs during wastewater treatment process (Radjenović et al., 2009). Particularly, the MPs removal mainly depends on the sorbents mass concentration and also physicochemical properties of MPs which include hydrophobicity, electrostatic interactions and hydrogen bond. The higher the hydrophobicity of MPs, the significant the sorption propensity towards the activated sludge, which leading to higher MPs removal efficiency (Radjenović et al., 2009). Despite its higher MPs removal rate compared to other conventional aerobic biological treatment methods, MBR is not effective in removing compounds with highly branched side and long chains, polycyclic/saturated compounds, and compounds consisting electron receptive functional groups (halogen and sulphate) such as diclofenac, carbamazepine and trimethoprim (Bui et al., 2016). Moreover, the microbial community is also subject to toxicity of some recalcitrant substances (Cazes et al., 2014). Some pharmaceuticals were found to be poorly degraded by MBR while some pharmaceuticals compounds can be well removed. For instance, the removal efficiency of anti-inflammatory drug (acetaminophen), antibiotics (azithromycin, clarithromycin, erythromycin, ofloxacin and sulfamethaxazole) and analgesics (carbamazepine, citalopram, ibuprofen, lorazepam, metronidazole, preimidone and trazodone) was reported in the range of 75–95% (Banihashemi and Droste, 2014; Dolar et al., 2012; Maeng et al., 2013; Nguyen et al., 2013; Trinh et al., 2012).

Compared to MBR, membrane technologies such as NF and RO involve less complex operating conditions and parameters. Most NF and RO membranes available in the market are with pore sizes suitable for water and wastewater treatment process (Al-Obaidi et al., 2017; Corzo et al., 2017; Lee et al., 2017; Soriano et al., 2017). To treat the water containing MPs, NF and RO membrane can offer high removal efficiency against pollutants than that of MBR, even those substances unable to be removed by MBRs such as diclofenac, carbamazepine, fenoprop, metronidazole and trimethoprim (Hai et al., 2011). According to Y. Liu

Table 2

Summary of the advantages and disadvantages of different treatment techniques.

Treatment techniques	Advantages	Disadvantages
Chemical and coagulation process	- Settling the fine colloidal and inorganic particles (Nicholas, 2020) - Remove insoluble contaminants (pigment, etc.) rapidly (Crini and Lichtfouse, 2019)	- Produce large volume of hazardous sludge (Nicholas, 2020) - Ineffective in MPs removal (Luo et al., 2014b) - Requires non-reusable chemicals (coagulants, flocculants, aid chemicals) (Crini and Lichtfouse, 2019)
Biodegradation process	- Environmentally friendly method since no need require chemicals (Aneyo et al., 2016) - Low cost (Sreekanth et al., 2009)	- Necessary to create an optimally favorable environment (Crini and Lichtfouse, 2019) - Uncontrolled degradation degree of products (Crini and Lichtfouse, 2019; Luo et al., 2014b)
Constructed wetland	- Lower operational costs compared to conventional treatment (Stefanakis, 2020) - Low energy consumption and maintenance-free (Sasaki et al., 2003)	- Consume large amount of space (Makopondo et al., 2020) - Function may be altered in different climatic condition (Stefanakis, 2020)
Membrane bioreactor (MBR)	- Higher volumetric loading rates (Iorhemen et al., 2016) - Less sludge production (Iorhemen et al., 2016) - High separation efficiency and can remove most of the bacteria and virus (Kowalik et al., 2021)	- Higher energy costs (Iorhemen et al., 2016; Kowalik et al., 2021) - Susceptible to fouling issues (Iorhemen et al., 2016; Kowalik et al., 2021)
Adsorption	- Low cost and easy maintenance (Vieira et al., 2020) - Suitable treatment for inorganic effluent (heavy metal, etc.) (Barakat, 2011)	- Low selectivity (Barakat, 2011) - Hard separation of the adsorbent from dye (Moosavi et al., 2020)
Advanced oxidation process (AOP)	- Could degrade majority of the organic compounds into biodegradable or harmless products (Zhou et al., 2018) - High reactivity and rapid process (Cuerda-Correa et al., 2020)	- Not cost-effective to mineralize toxic and recalcitrant compounds (Cesaro et al., 2013) - Additional by-products (Vieira et al., 2020) - High cost and chemicals needed (Kraft et al., 2003) - Require subsequent process to treat residual oxidant (Wang et al., 2019a)
Photocatalysis	- Environmentally friendly (Zhang et al., 2019) - Complete degradation of pollutants (Zhang et al., 2019)	- Poor chemical and photochemical corrosion resistance for 2D photocatalysts (Yang and Wang, 2018) - Difficult to treat large scale of wastewater (Prieto-Rodriguez et al., 2012) - Cost associated with artificial UV lamps and electricity (Prieto-Rodriguez et al., 2012)
Membrane technology (TFC membrane)	Good chemical and mechanical stabilities (Khoo et al., 2021c)	Membrane fouling issues (Khoo et al., 2021c)

Table 2 (continued)

Treatment techniques	Advantages	Disadvantages
	- Able to remove dye and PhACs, EDCs and other MPs (Katibi et al., 2021) - Suitable for treating saline water and waste water treatment plant influents (Khoo et al., 2020)	Membrane disposal problem after used (Khoo et al., 2021a)

et al. (2022) and Ma et al. (2018), the porous microfiltration and ultrafiltration membrane with large membrane pore size is not suitable for treating micropollutants. This is due to the fact that MPs molecular sizes are generally smaller (about 0.8 nm) (Tagliavini et al., 2017) than the pore size of MF/UF membranes (MF pore size varying between 0.1 and 5 µm while NF pore size ranged from 10 to 100 nm) (Mohammad and Ang, 2021). Hence, the inefficiency of MF/UF membrane can be overcome by using fine-pore membranes such as NF and RO membranes for MPs removal. During the early TFC NF and RO membranes development, Cadotte and his colleagues back in the 1970s successfully developed TFC membrane with high flux by crosslinking using piperazine (PIP) with trimesoyl chloride (TMC) via interfacial polymerization (IP) method (Cadotte, 1981). Generally, TFC membrane composed of an ultrathin polyamide (PA) selective layer (<200 nm) supported by a microporous substrate (~40 µm), has been commonly applied for NF and RO application. It is well acknowledged that by employing IP technique, the properties of membrane surface film and bottom substrate can be tailored individually to obtain better solute rejection and water permeation. Despite RO and NF require higher pressure requirement compared to UF, both systems are still widely used in water/wastewater treatment particularly due to their promising features such as excellent compression resistance and mechanical strength, easy to scale up, no chemical additives required and simple in operation (Alzahrani and Mohammad, 2014; Baker, 2012; Lau et al., 2012; Shannon et al., 2008).

The installation of RO and NF membranes for tertiary water treatments has been significantly increased in recent years for the production of high purity water (Silva et al., 2017). The key parameters that affect the rejection of both RO and NF membranes against MPs are dependent not only on the physicochemical properties of MPs (i.e., charge, size, diffusivity, hydrophobicity and solubility) but also the separation mechanisms of membranes (i.e., size exclusion, electrostatic interaction and hydrophobic interaction) (Bellona et al., 2004; Ojajuni et al., 2015). The main objective of this review is to elaborate the important findings of some exemplary RO and NF membranes applied for the removal of MPs from water. Particularly, this review aims to reveal the important of physicochemical properties of TFC, surface-modified TFC and nanomaterials-embedded TFN membranes on the MPs rejection via size exclusion, hydrophobicity interaction, electrostatic charge and adsorption. Tables 3 and 4 summarize some of the relevant studies over the past decade regarding the MPs removal using TFC and TFN membranes, respectively.

The NF and RO membranes in TFC configuration are the most favored membrane structure for water separation process primarily due to thin selective layer coupled with good water permeability compared to the asymmetric dense membranes. Numerous studies have evaluated the fabrication of membranes with improved water flux and MPs rejection and sought to investigate their underlying MPs rejection mechanisms. For example, the physicochemical properties of membrane PA layer have been altered through surface coating and chemical grafting methods using hydrophilic monomers to enhance the MPs rejection by controlling the adsorption mechanisms and size exclusion effect (Ben-David et al., 2010; Guo et al., 2016). Besides surface modification on membrane PA layer, the membrane MPs rejection can be improved via the introduction of nanomaterials to form TFN membrane. Many kinds of inorganic nanomaterials such as graphene oxide (GO),

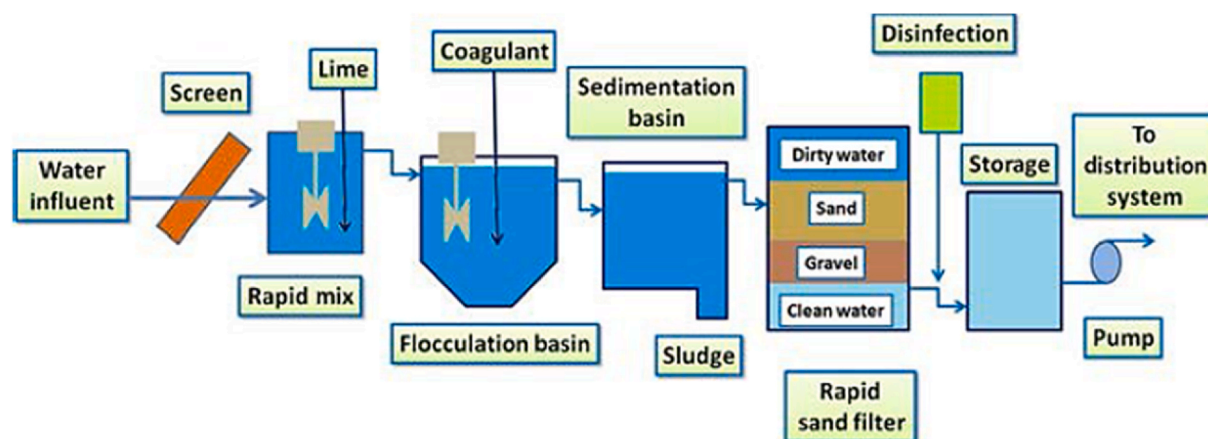


Fig. 3. Conventional water treatment process (Abu Shmeis, 2018).

metal-organic framework (MOF), silica dioxide (SiO_2) and titania nanotubes (TNTs) have been used for TFN membrane fabrication (Basu and Balakrishnan, 2017; Chong et al., 2019; Lai et al., 2018; Rakhshan and Pakizeh, 2015a). Recently, growing interests have been focused on the deposition of nanomaterials on the substrate before forming the PA layer. The fabrication of TFN membrane with an interlayer is found to enhance the solute rejection significantly and reduce the nanomaterials aggregation compared to the nanomaterials that deposited atop the PA layer or embedded within the PA layer (Yang et al., 2020). Most importantly, TFN membrane can effectively remove MPs without compromising membrane water flux when compared to the surface-modified membrane using monomer (Dong et al., 2016). Although membrane technology exhibits excellent performance in MPs solute rejection, membrane technology is challenged with the fouling issue which result in significant performance degradation such as flux deterioration, frequent replacement of membranes and eventually higher operating costs (Qi et al., 2020; Rao et al., 2022; Said et al., 2020; Saqib and Aljundi, 2016). Currently, considerable efforts have been attempted to overcome membrane fouling issues (Huang et al., 2021; Li et al., 2022; Y. W. Liu et al., 2022). Detailed discussion on the membrane water flux performance as a result of surface modification and incorporation of nanomaterials will be further provided in following sections.

1.8.1. EDCs removal using TFC and surface-modified TFC membranes

It was reported that both NF and RO membranes are able to remove numerous types of MPs not only through size exclusion, but also with the aid of charge effects and adsorption (Vieira et al., 2021). Guo et al. (2019) investigated the removal of methylparaben, ethylparaben, propylparaben and benzylparaben with concentration of 200 $\mu\text{g/L}$ using NF90 and XLE RO commercial membranes. Their findings revealed that size exclusion was the main mechanism for the MPs removal. Compared to NF90 membrane with pore size of 0.82 nm, XLE RO membrane with smaller pore size (0.62 nm) showed 75%, 27.2%, 34% and 43.5% higher rejection for methylparaben, ethylparaben, propylparaben, and benzylparaben, respectively.

Yüksel et al. (2013) on the other hand utilized NF90 and NF270 commercial membranes to remove BPA from water. Since the NF270 possesses larger molecular weight cut-off (MWCO: 200–300 Da) and higher permeate flux (121 $\text{L/m}^2 \cdot \text{h}$ measured at 10 bar) than that of NF90 (MWCO: 150–200 Da; 61 $\text{L/m}^2 \cdot \text{h}$), it allowed BPA to pass through its structure easier and thus lower BPA rejection was recorded (80% compared to 98% in NF90). Kimura et al. (2003) found that MPs could be removed via electrostatic charging effect other than size exclusion mechanism. The researchers investigated the removal efficiency of MPs via ESNA NF and XLE RO membranes using negatively charged MPs (salicylic acid, diclofenac, 9-anthracene carboxylic acid (ACA), dichloroacetic acid (DCAA) and trichloroacetic acid (TCAA)) and

non-charged MPs (primidone, phenacetine, BPA and 2-naphthol) with feed concentration of approximately 100 $\mu\text{g/L}$ via single solute experiments at 5.5 bar. After 24-h filtration test, higher rejections were achieved by XLE RO membrane (ranged from 92% to 96%) and ESNA NF membrane (ranged from 91% to 94%) for all of negatively charged MPs. The high MPs rejection is likely due to the electrostatic exclusion between negatively charged membrane surface and the anionic compounds which reduce the adsorption of MPs onto the membrane surface. Likewise, the XLE RO membrane with lower MWCO demonstrated better rejection compared to ESNA NF membrane. In their study, the size exclusion was weakly correlated to the MPs removal since ESNA NF membrane (MWCO: 200 Da) was able to remove 91% of DCAA (MW: 129 Da), even though the size of DCAA is significantly smaller than the pore size of ESNA NF membrane. All of the non-charged MPs demonstrated lower MPs rejection (ranged from 12% to 87%) after tested using ESNA NF membrane for 8 h.

In addition, Fujioka et al. (2020) utilized commercial ESPA2 RO membrane to investigate the mechanisms underlying the poor rejection of small pesticides by PA RO membranes. The authors noticed that the positively charged diuron, diflufenbuzon and chlorotoluron pesticide compounds were only rejected by 34–65%. When the ESPA2 RO membrane was used to treat negatively charged quinmerac, it demonstrated considerably high rejection of 98% against the negatively charged quinmerac after subjected to 5 $\mu\text{g/L}$ pesticides solution for approximately 70 h at 10 bar. It is worth noting that the hydrogen bond interaction between pesticide compounds and membrane PA layer is likely to be the main reason resulting in low pesticides rejection. The hydrogen bonding can commonly occur between the hydrogen and oxygen atom of pesticide amide groups with the oxygen and hydrogen atom of membrane PA layer, respectively. Consequently, secondary amide pesticides with neutral charge attracted to the PA layer of RO membrane via hydrogen bonding, which ultimately leads to low rejection. These results further suggested that not only size exclusion effect and electrostatic interaction, the hydrogen bond interactions may play an important role in the rejection of pesticides compounds.

Since the MPs contaminants could adsorb on the PA active layer and subsequently diffuse through the membrane, surface modification on the PA active layer becomes a key interest for researchers to improve the membrane performance. For instance, Guo et al. (2016) highlighted the positive effect of membrane surface coating on the MPs removal rate after NF90 membrane was coated with hydrophilic PDA. Four types of hydrophobic EDCs (i.e., ethylparaben, propylparaben, benzylparaben and BPA) were used to determine the removal efficiency of surface-coated NF90 membrane. As shown in Fig. 4(a), the EDCs rejection of ethylparaben, propylparaben, benzylparaben and BPA were improved from ~50%, ~50%, ~35% and ~96%~70%, ~70%, ~60% and 99%, respectively after the NF90 membrane was coated with PDA

Table 3

Summary of TFC membranes with and without surface modification for MPs removal.

^a Membrane type	Optimum modification condition	^b EDC micropollutants	Type	Pressure, bar	Permeate flux, L/m ² .h	Rejection, %	Mechanism	Ref
MA-grafted membrane	60 min grafting	1 mg/L BPA	NF	2.5	~32 to ~24 ↓	74.1 to 96.9 ↑	Size exclusion	Kim et al. (2008)
		1 mg/L ibuprofen			n/a	98.1 to 99.7 ↑	Electrostatic charge	
		1 mg/L salicylic acid			n/a	97.0 to 99.1 ↑	Electrostatic charge	
ED-grafted membrane	–	1 mg/L BPA	NF	2.5	Maintained at ~32	74.1 to 97.6 ↑	Size exclusion	Kim et al. (2008)
		1 mg/L ibuprofen			n/a	98.1 to 98.2 ↑	Electrostatic charge	
		1 mg/L salicylic acid			n/a	97.0 to 97.1 ↑	Electrostatic charge	
SA-grafted membrane	–	1 mg/L BPA	NF	2.5	Maintained at ~32	74.1 to ~98.0 ↑	Size exclusion Electrostatic charge Electrostatic charge	Kim et al. (2008)
		1 mg/L ibuprofen			n/a	98.1 to ~99.0 ↑		
		1 mg/L salicylic acid			n/a	97.0 to ~98.0 ↑		
PDA-coated membrane (4 h coating)	4 h coating	200 µg/L BPA	NF	8	n/a	96 to 99 ↑	All caused by weakened hydrophobicity interaction	Guo et al. (2016)
		200 µg/L ethylparaben			n/a	50 to 70 ↑		
		200 µg/L propylparaben			n/a	50 to 70 ↑		
		200 µg/L benzylparaben			n/a	35 to 60 ↑		
SPM-grafted NF270	0.1 M	500 ppm Na ₂ SO ₄ and 20 ppm BPA	NF	15	~120 to ~100 ↓	~40 to ~70 ↑	All caused by weakened hydrophobicity interaction and salting out	Ben-David et al. (2010)
		500 ppm Na ₂ SO ₄ and 20 ppm Metolachlor			~160 to ~125 ↓	~80 to ~95 ↑		
HEMA-grafted NF270	0.01 M	500 ppm Na ₂ SO ₄ and 20 ppm BPA	NF	15	~125 to ~90 ↓	~53 to ~70 ↑	All caused by weakened hydrophobicity interaction and salting out	Ben-David et al. (2010)
		500 ppm Na ₂ SO ₄ and 20 ppm Metolachlor			~150 to ~120 ↓	~82 to ~98 ↑		
TA-Fe coated NF270	2.4 mM TA: 7.2 mM Fe	200 µg/L methylparaben	NF	10	~550 to ~130 ↓	21.6 to 42.6 ↑	All caused by size exclusion	Guo et al. (2017b)
		200 µg/L ethylparaben			~380 to ~70 ↓	19.9 to 54.4 ↑		
		200 µg/L propylparaben			~350 to ~60 ↓	21.3 to 57.3 ↑		
		200 µg/L benzylparaben			~400 to ~90 ↓	19.6 to 48.3 ↑		
TA-Fe fabricated membrane	2.4 mM TA: 7.2 mM Fe	200 µg/L methylparaben	NF	3	n/a	69.2	All caused by size exclusion	Guo et al. (2019)
		200 µg/L ethylparaben			n/a	93.1		
					n/a	97.6		

(continued on next page)

Table 3 (continued)

^a Membrane type	Optimum modification condition	^b EDC micropollutants	Type	Pressure, bar	Permeate flux, L/m ² .h	Rejection, %	Mechanism	Ref
GMA-modified ESPA1 membrane	2 mM of GMA and 0.045 mM Triton X-100	200 µg/L propylparaben 200 µg/L benzylparaben 100–120 mg/L BPA 100–120 mg/L ACN	NF	20	n/a n/a	99.7 99.66 to 99.91 ↑ 99.30 to 99.59 ↑	All caused by steric effect	Baransi-Karkaby et al. (2019)
ESPA2 commercial membrane	–	5 µg/L Diuron 5 µg/L Protham 5 µg/L Quinmerac	RO	10	n/a	34 79 98	Size exclusion Electrostatic charge Electrostatic charge	Fujioka et al. (2020)
NF90 commercial membrane	–	200 µg/L methylparaben 200 µg/L ethylparaben 200 µg/L propylparaben 200 µg/L benzylparaben	NF	3	n/a n/a n/a n/a	30 60 58 57	All caused by size exclusion	Guo et al. (2019)
XLE commercial membrane	–	200 µg/L methylparaben 200 µg/L ethylparaben 200 µg/L propylparaben 200 µg/L benzylparaben	RO	3	n/a n/a n/a n/a	52.5 76.3 77.4 81.8	All caused by size exclusion	Guo et al. (2019)
ESNA commercial membrane	–	100 µg/L 9-ACA 100 µg/L SA 100 µg/L DCAA 100 µg/L TCAA 100 µg/L diclofenac 100 µg/L 2-naphthol 100 µg/L BPA 100 µg/L phenacetine	NF	5.5	n/a n/a n/a n/a n/a n/a n/a n/a	93 92 91 94 93 12 45 19	Electrostatic charge Electrostatic charge Electrostatic charge Electrostatic charge Electrostatic charge Size exclusion Size exclusion Size exclusion	Kimura et al. (2003)
XLE commercial membrane	–	100 µg/L primidone 100 µg/L 9-ACA 100 µg/L SA 100 µg/L DCAA 100 µg/L TCAA 100 µg/L diclofenac 100 µg/L 2-naphthol 100 µg/L BPA 100 µg/L phenacetine	RO	5.5	n/a n/a n/a n/a n/a n/a n/a n/a n/a	87 96 92 95 96 95 43 99 71	Size exclusion Electrostatic charge Electrostatic charge Electrostatic charge Electrostatic charge Electrostatic charge Size exclusion Size exclusion Size exclusion	Kimura et al. (2003)
TFC-SR2 commercial membrane	–	100 µg/L primidone 100 ng/L esterone	NF	5	n/a n/a	84 >95	Size exclusion Adsorption	Schäfer et al. (2003)
NF270 commercial membrane	–	50 mg/L BPA	NF	10	121	80	Steric hindrance	Yüksel et al. (2013)
NF90 commercial membrane	–	50 mg/L BPA	NF	10	61	98	Steric hindrance	Yüksel et al. (2013)
Self-fabricated TFC membrane	–	250 µg/L CMZ and 10 mM NaCl 250 µg/L DCF and 10 mM NaCl 250 µg/L IBU and 10 mM NaCl 250 µg/L NPX and 10 mM NaCl	FO	Osmotic pressure (Draw: 2 M NaCl)	n/a n/a n/a n/a	~96 ~96 ~95 ~93	All can be caused by electrostatic charge and size exclusion	Jin et al. (2012)

^a MA (methacrylic acid), ED (ethylene diamine), SA (succinic acid), PDA (polydopamine), SPM (3-sulfopropyl methacrylate), HEMA (3-sulfopropyl methacrylate), TA-Fe (tannic acid-ferric ion), GMA (glycidyl methacrylate).

^b BPA (bisphenol-A), Na₂SO₄ (sodium sulphate), 9-ACA (9-anthracenecarboxylic acid), SA (salicylic acid), DCAA (dichloroacetic acid), TCAA (trichloroacetic acid), CMZ (carbamazepine), DCF(diclofenac), IBU(ibuprofen), NPX (naproxen), ACN (acetaminophen).

Table 4

Summary of nanomaterials-modified TFN membranes for MPs removal.

^a Membrane type	Optimum modification condition	^b EDC micropollutants	Type	Pressure, bar	Permeate flux, L/m ² .h	Rejection, %	Mechanism	Ref
AgNPs-incorporated membrane	20 mM AgNPs	200 µg/L propylparaben	RO	20	n/a	~95 to ~98 ↑	Donnan and size exclusion	Yang et al. (2019)
		200 µg/L norfloxacin				~96 to ~97 ↑	Size exclusion	
		200 µg/L ofloxacin				~96.5 to ~97 ↑	Size exclusion	
MOS ₂ -incorporated membrane	40 ml of 0.004 mg/mL of MOS ₂ solution	200 µg/L Methylparaben	NF	8	n/a	25.2 to 53.7 ↑	All caused by weakened hydrophobicity interaction and size exclusion	Dai et al. (2021)
		200 µg/L Ethylparaben				28.1 to 69.1 ↑		
		200 µg/L Propylparaben				38.3 to 79.1 ↑		
		200 µg/L Benzylparaben				59.8 to 91.3 ↑		
MIL-101(Cr) incorporated membrane	0.2 wt/v% MIL-101 (Cr)	200 µg/L Methylparaben	NF	8	n/a	27.7 to 47.4 ↑	All caused by weakened hydrophobicity and enhanced hydrophilicity	Dai et al. (2019)
		200 µg/L Propylparaben				25.2 to 45.9 ↑		
		200 µg/L Benzylparaben				31.3 to 51.1 ↑		
		200 µg/L BPA				64.9 to 79.8 ↑		
PDA-AgNPs modified membrane	4 h AgNPs coating time	200 µg/L Ethylparaben	NF	8	n/a	~50–67% ↑	Size exclusion	Guo et al. (2017a)
		200 µg/L Propylparaben				~50–69% ↑	Size exclusion	
		200 µg/L Benzylparaben				~35–66% ↑	Enhanced hydrophilicity	
		200 µg/L BPA				~97–99% ↑	Size exclusion	
TiO ₂ -incorporated membrane PA/HKUST-1 BTFC-modified membrane	0.5 g TiO ₂ in 2 wt% MPD solution	100 ppm BPA	–	5	n/a	99% ↑	Size exclusion	Anan et al. (2019)
	–	1 mg/L diclofenac	NF	20	0.34 to 1.66 ↑	99.3 to 99.5 ↑	Size exclusion	Paseta et al. (2019)
		1 mg/L naproxen			0.22 to 1.25 ↑	99.3 to 98.3 ↓		
PA/ZIF-93 BTFC-modified membrane	–	1 mg/L diclofenac	NF	20	0.34 to 1.21 ↑	99.3 to 99.0 ↓	Size exclusion	Paseta et al. (2019)
		1 mg/L naproxen			0.22 to 0.53 ↑	99.3 to 99.3 ↓		
Zeolite-incorporated membrane	500 ppm zeolite	PhACs mixture solution (each PhAC with 20 µg/L)	NF	11	~24 to ~63 ↑	Maintained at >90% ↑	Size exclusion, weakened hydrophobicity and electrostatic charge	Dong et al. (2016)
SiO ₂ -incorporated membrane	(0.17 w/v% SiO ₂)	100 µg/L atrazine	NF	10	~14 to ~18 ↑	~94 to ~98 ↑	All caused by weakened hydrophobicity and size exclusion	Rakhshan and Pakizeh (2015b)
		100 µg/L propazine			~11 to ~17 ↑	~84 to ~91 ↑		
		100 µg/L prometryn			~4.5 to ~7.5 ↑	Maintained at ~99 ↑		
GO-modified SW30HR membrane	10 parts of 250 µg/mL	890 µg/L NDMA	RO	27.6	– ↑	76.5 to 82.7 ↑	Size exclusion	Croll et al. (2019)

^a AgNPs (silver nanoparticles), PDA (polydopamine), TiO₂ (titanium dioxide), PA/HKUST-1 BTFC (Cu₃(1,3,5-benzenetricarboxylate)₂(H₂O)₃)-bilayer thin film composite, PA/ZIF-93 BTFC (Zn (4-methyl-5-imidazolecarboxaldehyde)₂), SiO₂ (silica dioxide).

^b BPA (bisphenol-A), PhAC (pharmaceutically active compounds), NDMA (N-nitrosodimethylamine).

for 4 h. The improved EDCs rejection can be attributed to the longer coating duration, leading to a thicker PDA layer formed on the membrane surface and thus creating a barrier resistance for EDCs to reduce their diffusion through the membrane. In addition, the improved MPs rejection can be primarily ascribed to the enhanced membrane surface

hydrophilicity (with water angle reduced from 69.8° to 62° upon 4-h PDA coating), which weaken the hydrophobic interaction between the membrane surface and hydrophobic EDCs. It can be seen from Fig. 4(b) that all of the EDCs tested showed a reduced sorption trend with increasing PDA coating time. As the sorption rate decreased, the EDCs

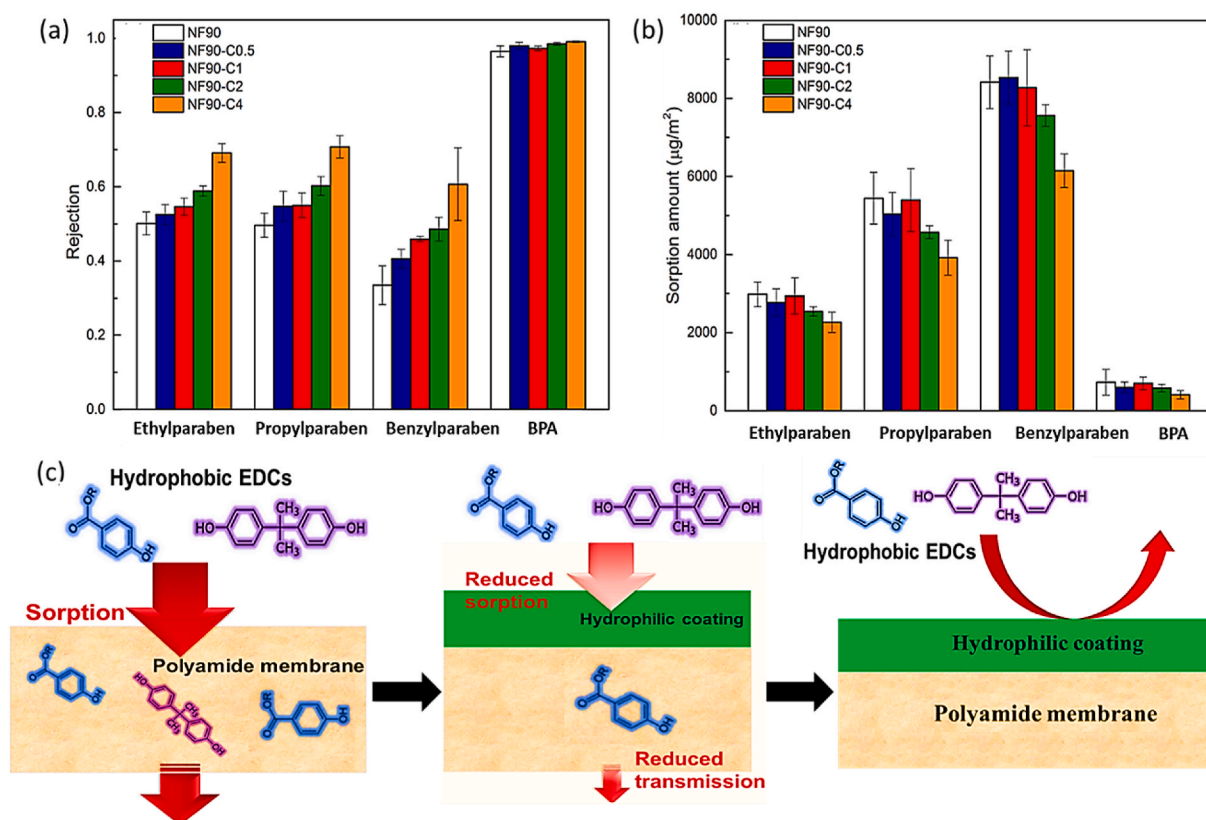


Fig. 4. (a) EDCs rejection performance and (b) EDCs sorption of unmodified NF90 and PDA-modified NF90 (note: C0.5, C1, C2 and C4 represented the PDA coating duration at 0.5 h, 1 h, 2 h and 4 h, respectively), (c) illustration diagram of the sorption and transmission of hydrophobic EDCs through unmodified NF90 and PDA-coated membranes (Guo et al., 2016).

passage was greatly reduced and this resulted in better EDCs rejection (Fig. 4(c)).

Other than PDA coating, Guo et al. (2017b) also utilized green accessible materials, such as tannic acid (TA) and ferric ion (Fe^{3+}) to functionalize the top surface of NF270 membrane via a simple and rapid surface coating method (see Fig. 5(a)). From Fig. 5(b) and 5(c), it can be obviously seen that the Ta/Fe-modified membrane with Ta-Fe molar ratio of 1:3 (labelled as NF270-Fe3) demonstrated outstanding solute rejection against 4 hydrophobic neutral EDCs. This membrane exhibited 21%, 34.5%, 36% and 28.7% higher rejection than the control NF70 for methylparaben, ethylparaben, propylparaben and benzylparaben, respectively after tested using 200 $\mu\text{g/L}$ EDCs for 12 h at 10 bar. Furthermore, the NF270-Fe3 membrane also achieved promising results (>95% rejection) against antibiotics including sulfadiazine, sulfamethoxazole, sulfamethazine, trimethoprim, norfloxacin and ofloxacin. The authors reported that size exclusion effect was the dominant factor contributing to higher EDCs and antibiotics removal as the average pore size of NF270-Fe3 (0.40 nm) was smaller than the control NF270 (0.44 nm). The authors further elucidated that the rejection of NF270 was due to the unfavorable interaction between PA and MPs (e.g., π - π stacking (Schäfer et al., 2011) and hydrophobic interaction (Verliefde et al., 2009)) which promoted the sorption of MPs within the PA layer. Although Ta-Fe coating could enhance MPs rejection, the EDCs permeability for NF270 membrane was reduced by 4–5 times upon the surface coating, as shown in Fig. 5(c).

Separately, Kim et al. (2008) functionalized the surface of a NE4040-70 commercial NF membrane and used it for the removal of EDCs and PhACs. As shown in Fig. 6(a), the surface of NF4040-70 was altered using three different materials via the following modification sequences: graft polymerization of methacrylic acid (MA) atop membrane PA layer to form MA-modified membrane followed by

cross-linking of grafted polymer chains with primary amine groups to form ethylene diamine (ED)-modified membrane and lastly, substitution of carboxylic acid groups to produce succinic acid (SA)-modified membrane. The results showed that all modified membranes could achieve >95% BPA rejection which was significantly higher than that of the control membrane (74.1%). Compared to the control membrane, high amount of ibuprofen was able to be removed by MA-modified membrane (99.7%) and SA-modified membrane (99.0%) compared to the ED-modified membrane. Since BPA is a neutral solute at pH 7, the enhanced BPA rejection can be attributed to the steric hindrance effect resulting from the polymer chains. Contradictorily, the rejection of ibuprofen and salicylic acid was mainly caused by the improved membrane surface negative charge as depicted in Fig. 6(b) upon COOH functionalization atop the PA layer. The presence of COOH groups was evidenced in ATR-FTIR spectra shown in Fig. 6(c) where the peak intensity at 1701 cm^{-1} was found to be greater for the MA-modified and SA-modified membranes.

In an attempt to ameliorate membrane resistant against organic foulants, Ben-David et al. (2010) utilized negatively charged 3-sulfo-propyl methacrylate (SPM) and hydrophilic 2-hydroxyethyl methacrylate (HEMA) to modify the surface of commercial NF270 membrane via surface graft polymerization for the removal of BPA and metolachlor compounds. It must be noted that MPs with higher hydrophobicity could be removed easily by the hydrophilic membrane due to the larger partitioning coefficient. However, 200 $\mu\text{g/L}$ metolachlor compounds with lower hydrophobicity showed higher removal rate (>95%) than 200 $\mu\text{g/L}$ BPA (~70%) compounds after treated using 0.1 M SPM-modified and 0.01 M HEMA-modified membranes at 15 bar. The authors asserted that BPA is a strong hydrogen donor and might show some specific interaction with the HEMA and SPM-grafted membrane, hence leading to weak BPA removal rate compared to metolachlor compounds.

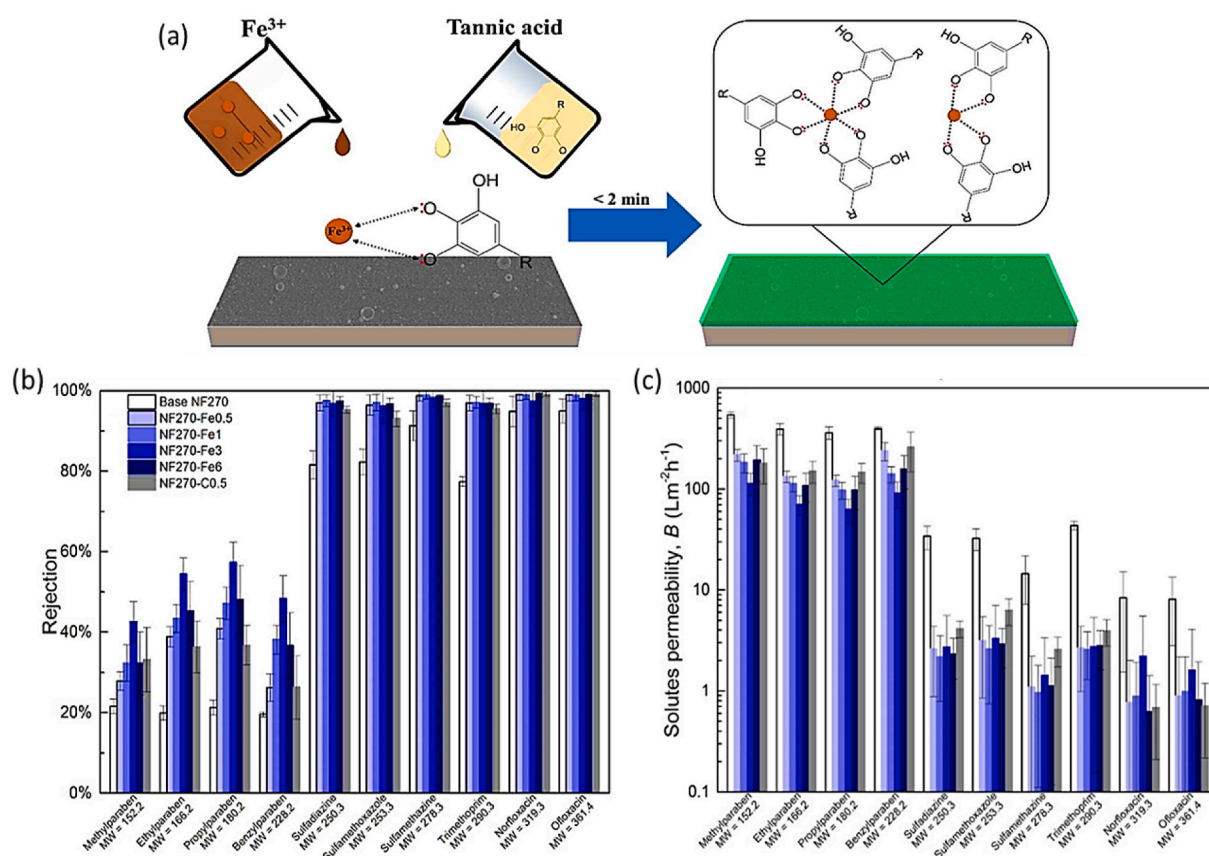


Fig. 5. (a) Schematic diagram of the Ta-Fe coating formed on the membrane surface, (b) MPs solute rejection (c) MPs solute permeability of membranes (note: Fe0.5, Fe1, Fe3 and Fe6 represented the molar ratio of Ta-Fe at 1:0.5, 1:1, 1:3 and 1:6 while C0.5 represent PDA coating time at 0.5 h) (Guo et al., 2017b).

A similar strategy was adopted by Baransi-Karkaby et al. (2019), who modified the surface of ESPA1 membrane using poly (glycidyl methacrylate) (GMA) via surface graft polymerization. Despite a noticeable decrease in permeate permeability from 1.8 to 1.4 $\text{L m}^{-2} \text{h}^{-1} \text{bar}$ after tested using mixed MPs solution, the GMA-modified membrane exhibited greater rejection of BPA and acetaminophen (ACN) at 99.91% and 99.59%, respectively. It was speculated that the additional GMA coating layer helps to tighten and seal the surface defects area of PA layer, which indiscriminately block non-selective passage and improve rejection of all MPs compounds (Ben-David et al., 2010; Bernstein et al., 2010, 2012).

1.8.2. EDCs removal using thin film nanocomposite (TFN) membrane

Although longer surface coating duration could enhance the membrane surface hydrophilicity, it at the same time adversely affects membrane water permeability as evidenced elsewhere (Khoo et al., 2021b; Said et al., 2020; Yu et al., 2013). This is due to the formation of thicker and denser coating layer atop the membrane which acts as a water transport barrier for water to pass through membrane. In order to address this issue, some researchers modified the membrane surface using hydrophilic nanomaterials.

Guo et al. (2017a) immobilized silver nanoparticles (AgNPs) atop the PDA-coated NF90 membrane and used it for MPs removal. Fig. 7 (a)-(b) show the surface morphology of control NF90 and AgNPs-PDA-modified NF90 membrane (designated as NF90-C0.5Ag4). Notably, a lot of nano-sized particles could be found on the membrane surface, indicating the existence of AgNPs on the membrane surface. The presence of AgNPs on the membrane surface was further confirmed using XPS analysis as shown in Fig. 7(c) and about 0.65% of silver contents was detected for the NF90-C0.5Ag4 membrane. It must be pointed out that the silver functionalized membrane led to improved size exclusion effect which

improved membrane selectivity and enhanced surface which weakened the adsorption interaction with hydrophobic MPs. Evidently, the NF90-C0.5Ag4 membrane was able to achieve higher MPs removal efficiency, recording 34%, 38%, 86% and 2% improvement for ethylparaben, propylparaben, benzylparaben, and BPA, respectively compared to control. Fig. 7(d) shows the MP rejection efficiency of different NF membranes.

Generally, nanoparticles such as carbon nanotube, mesoporous silica (SiO_2) and zeolite are reported to be able to provide preferential flow path for water. Hoek and co-workers in 2007 first reported the use of porous zeolite nanoparticles (NPs) to create an additional water pathway within the PA selective layer without jeopardizing its salt rejection. Meanwhile, the improved performance of TFC membranes can also be potentially achieved by incorporating zeolite nanoparticles within the polysulfone (PSf) support, followed by forming a PA layer atop the PSf substrate via IP method (Dong et al., 2016). Dong et al. (2016) reported that permeate flux of control TFC membrane has been enhanced by 2.45 times after incorporating 500 mg/L of zeolite within the PSf layer followed by PA layer formation atop the PSf substrate (designated as TFN-I500). The improved permeate permeability can be ascribed to the larger internal pore size of zeolite nanoparticles (~ 0.70 nm) which allows more water molecules (~ 0.27 nm) to pass through. Another reason could be possibly due to the formation of microporous defects as shown in Fig. 8(a)(ii), whereby the nanoparticles cavities were existed between PA and PSf layer. Despite TFN-I500 able to reject most of the PhACs compounds effectively ($>90\%$) after tested with 21 type of PhACs (with each PhAC compound feed at $20 \mu\text{g/L}$), still only 85%, 82%, 84% and 84% of carbamazepine, metoprolol, ranitidine and chloramphenicol, respectively were removed by TFN-I500 (see Fig. 8(b)). The author claimed that the incorporation of zeolite did not alter the surface hydrophobicity and surface charge of membrane, while the lower PhACs

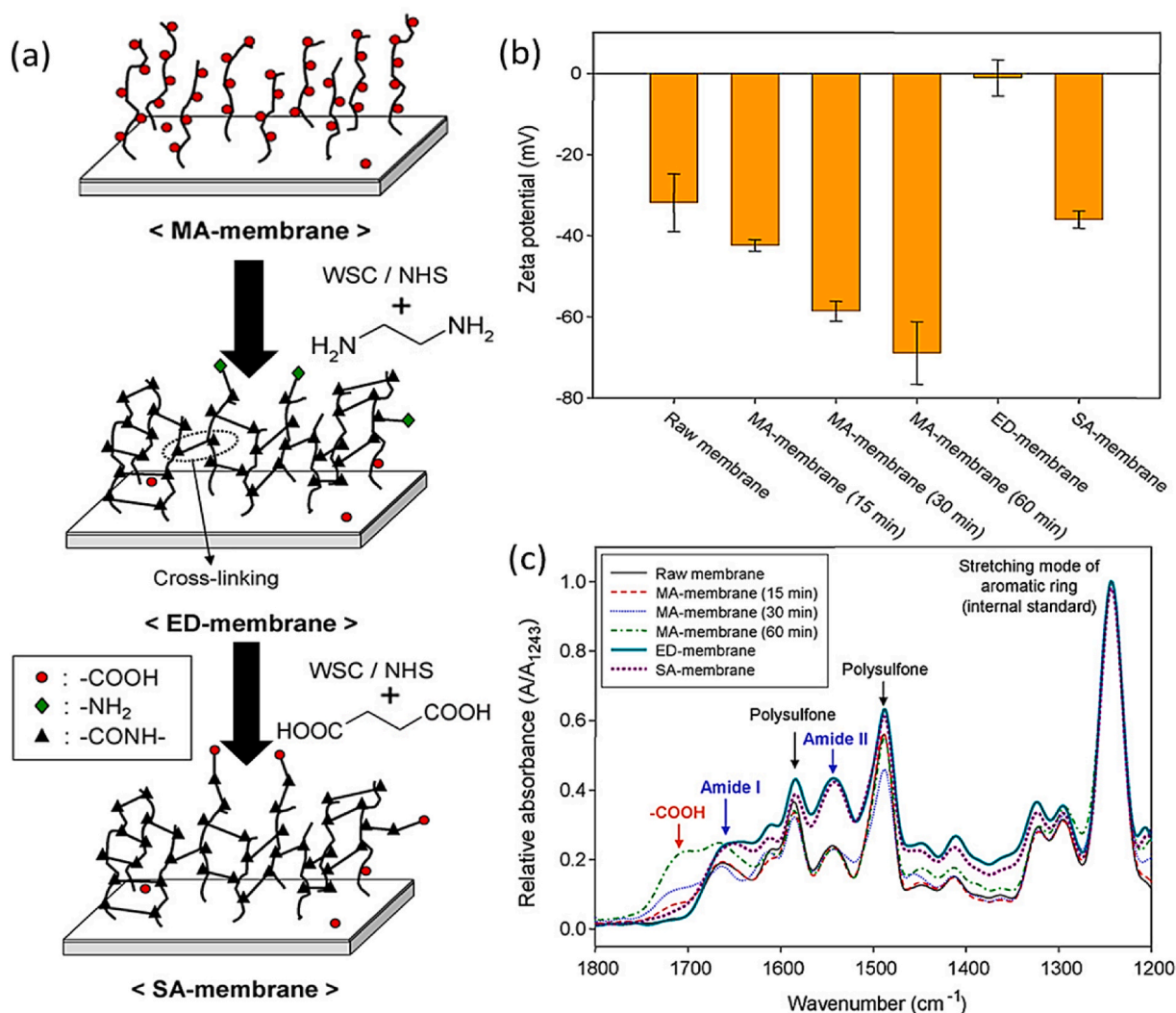


Fig. 6. (a) cross-linking of grafted polymer chains and substituting functional groups, (b) zeta potential analysis and (c) ATR-FTIR spectra of unmodified and modified membranes (Kim et al., 2008).

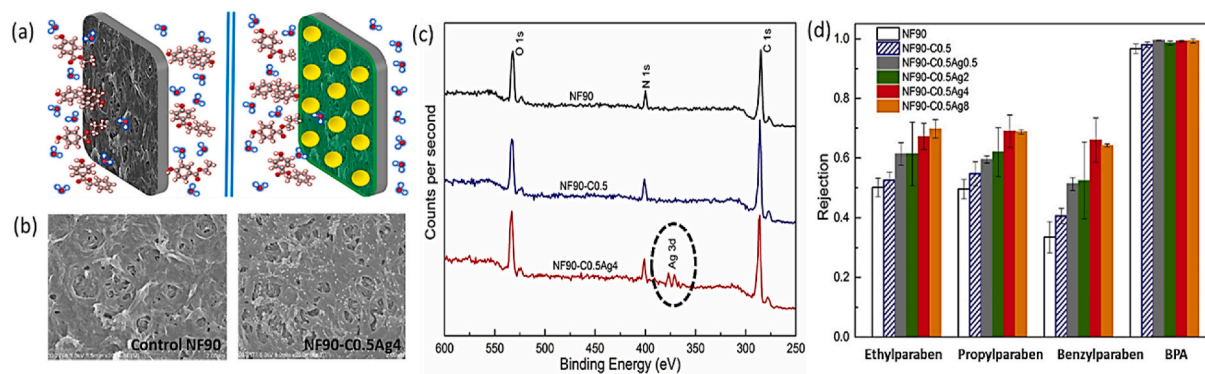


Fig. 7. (a) illustration diagram of unmodified membrane (left) and modified NF90 membrane (right) (b) SEM surface morphology of control NF90 (left) and modified NF90 (right), (c) XPS analysis and (d) MPs rejection performance of NF90 and silver functionalized PDA-coated NF90 (note: C0.5 represented PDA coating time at 0.5 h while Ag0.5, Ag2, Ag4 and Ag8 corresponded to AgNPs loading time at 0.5 h, 2 h, 4 h and 8 h) (Guo et al., 2017a).

rejection can be mainly caused by the enlarged membrane MWCO upon the incorporation of zeolite within PSf layer.

Other than zeolite, Yang et al. (2019) concluded the incorporation of hydrophilic AgNPs was able to generate nanochannels that act as a water pathway within the PA layer. As illustrated in Fig. 8(c), AgNPs were

produced in situ on the PSf layer using 20 mM of AgNO₃ solution and 20 mM of NaBH₄, followed by the formation of PA layer atop the PSf substrate via IP process to form TFN membrane (known as TFC-Ag20). Obviously, the presence of hydrophilic AgNPs could attract the water molecules from surroundings, which in turn hydrolyze the TMC

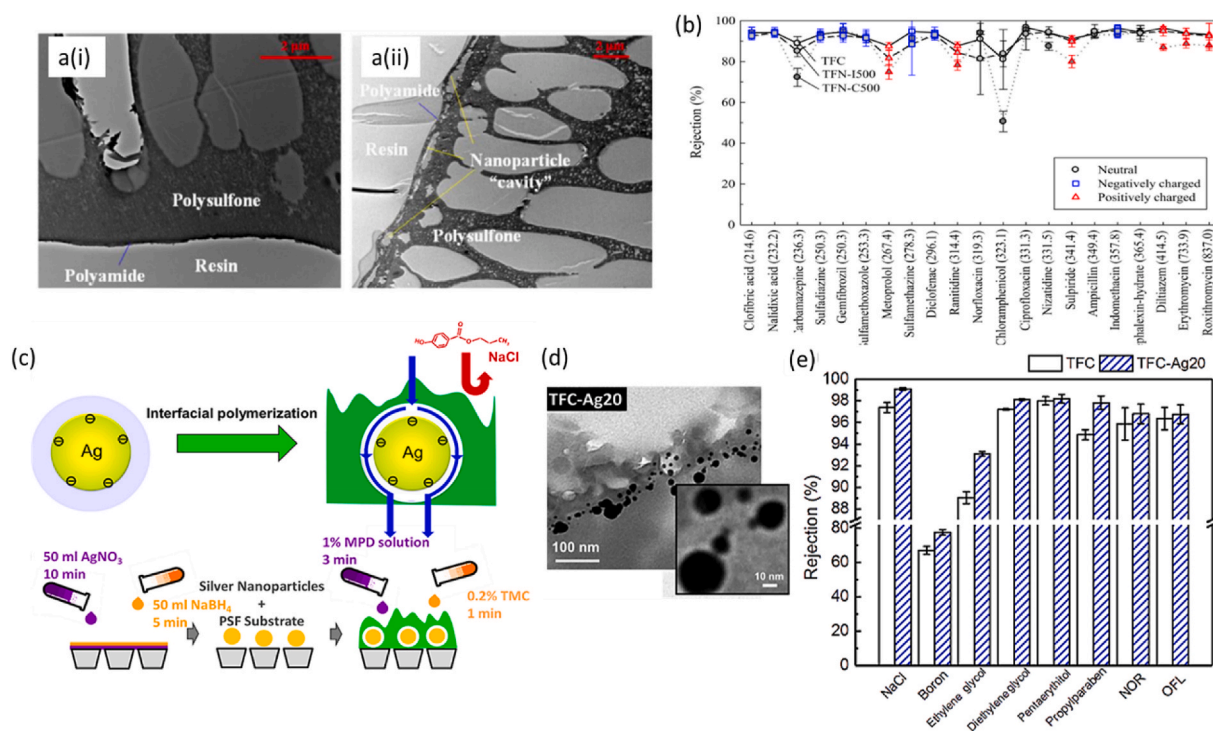


Fig. 8. (a) TEM images of: a(i) TFC and a(ii) TFN-I500 membranes (note: TFC represented control membrane while TFNI500 is the TFN membrane with UF support embedded with 500 mg/L of zeolite nanoparticles) (Dong et al., 2016), (b) Rejection of the 21 individual PhACs using TFC, TFN-I500 and TFN-C500 membranes, (c) schematic diagram of membrane fabrication, (d) TEM views of TFC-Ag20 and (e) membrane rejection performance (Yang et al., 2019).

monomer and terminate the IP process in their vicinity. This, resulting in lower mass density and the formation of nanochannels of approximately 2.5 nm in size around the AgNPs (light colored region as depicted in Fig. 8(d)). Consequently, TFC-Ag20 membrane showed the best water flux performance where 170% of pure water flux was enhanced from 18.6 to 50.0 L/m². h. In addition, TFC-Ag20 was also evaluated for its potential in treating different type of EDCs, such as neutral hydrophilic compounds (pentaerythritol, norfloxacin, and ofloxacin) and

hydrophobic compound (propylparaben). The embedding of AgNPs improved the rejection of pentaerythritol, norfloxacin, ofloxacin and propylparaben rejection by ~0.3%, ~1%, ~0.3% and ~3%, compared to that of TFC membranes when tested with 200 µg/L of individual EDCs for 12 h at 20 bar (Fig. 8(e)). The enhanced rejection against the neutral hydrophilic compounds was mainly due to the increased membrane cross-linking (increased from 54.2% to 62%) and thus greater size exclusion effect. The improved surface hydrophilicity (water contact

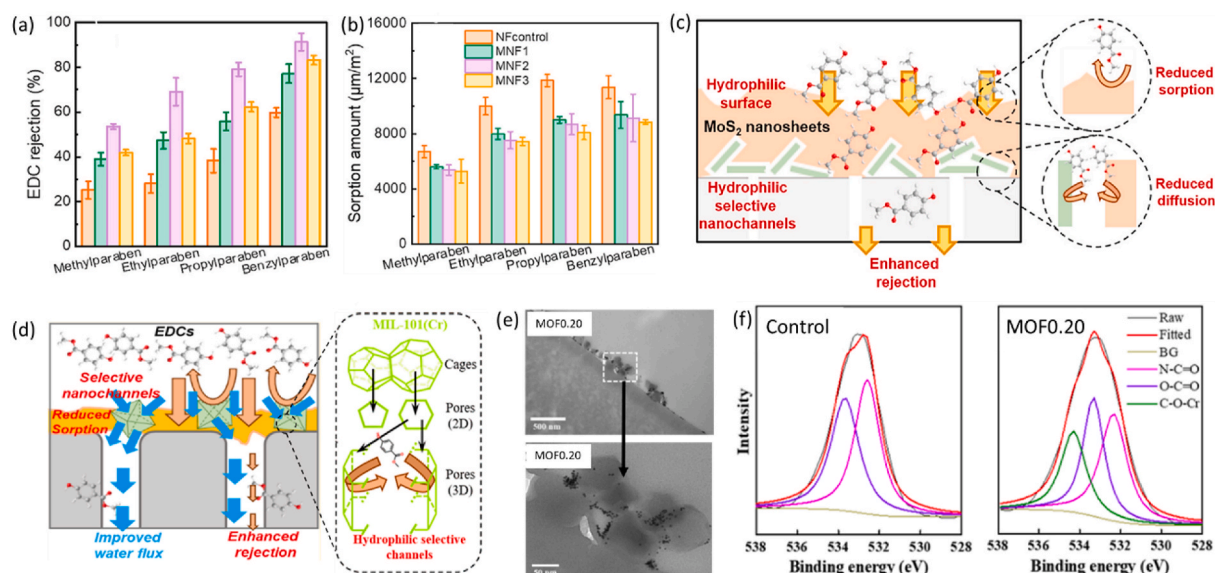


Fig. 9. (a) Rejection of EDCs, (b) effect of MoS₂-incorporated membrane on the EDCs sorption, (c) schematic diagram of the EDCs rejection mechanism by MoS₂-incorporated membrane (note: MNF1, MNF2 and MNF3 represented MoS₂ volume at 10 ml, 20 ml and 40 ml) (Dai et al., 2021) (d) schematic diagram of the MPs rejection mechanism by MIL101(Cr)-incorporated membrane, (e) TEM view of MOF0.20 membrane tested using gold nanoparticle, (f) XPS analysis of control and MOF0.20 membranes (note: MOF0.20 represented MIL101(Cr)-embedded loading at 0.20 wt/v%) (Dai et al., 2019). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

angle reduced from $\sim 84^\circ$ to $\sim 72^\circ$) and increased size exclusion effect could suppress the hydrophobic interaction of propylparaben towards the membrane surface.

Besides incorporating zeolite and Ag nanoparticles in TFC membrane, the sheet-shaped hydrophilic MOS_2 nanosheets with high aspect ratio was also found to be effective in alleviating the hydrophobic interaction between EDC compounds and membrane surface as documented in the work of Dai et al. (2021). As shown in Fig. 9(a), it can be found that the rejection rate of methylparaben, ethylparaben, propylparaben, and benzylparaben using MOS_2 -incorporated TFN membrane with volume of 20 mL (also designated as MNF2) increased from 25.2%, 28.1%, 38.3%, and 59.8%–53.7%, 69.1%, 79.1%, and 91.3%, respectively. However, the embedment of 30 mL MOS_2 nanosheet within TFC membrane was not recommended as over-stacking of MOS_2 nanosheets increased the resistance for water transport and create surface defects in the PA layer. The EDCs rejection was mainly caused by the reduced sorption (see Fig. 9(b)) and the decreased diffusion of EDCs compounds through MOS_2 -incorporated membrane. Particularly, the hydrophilic membrane surface has reduced the initial MPs sorption while EDCs diffusion has been selectively impeded by the hydrophilic nanochannels (see Fig. 9(c)).

Compared to other nanoparticles or nanosheets, the adjustable pore size, highly porous structure and good compatibility with PA layer make metal organic frameworks (MOFs) an emerging alternative for advancing membrane performance (Li et al., 2012, 2017; Ma et al., 2017; Sorribas et al., 2013). In a separate work, Dai et al. (2019) utilized MIL-101(Cr) with 1.2 nm pentagonal/1.6 nm hexagonal openings in order to modify the physicochemical properties of PA layer for enhanced MPs rejection and water flux. The authors claimed that mesoporous MIL-101(Cr) was selected because of its large pore volume and surface area that offers abundant water nanochannels (see Fig. 9(d)). Upon the embedment of 0.2 wt/v% MIL-101(Cr) within the PA layer (designated as MOF0.20 membrane), it possessed nearly 2 times larger water/EDC selectivity (0.52) compared to the NF control membrane (0.25). The significant enhanced water transport can be mainly due to the selective nanochannels produced from MIL-101(Cr). The water pathway within MIL-101(Cr) nanochannels were further confirmed using gold nanoparticle filtration test and TEM as depicted in Fig. 9(e), where certain amounts of fine size gold nanoparticles were able to diffuse through the MIL-101(Cr) nanochannels. In term of MPs rejection, the removal rates of MOF0.20 against methylparaben, propylparaben, benzylparaben, and BPA were recorded at 47.4%, 45.9%, 51.1%, and 79.8%, respectively. These values were significantly higher than that of NF control (27.7%, 25.2%, 31.3%, and 64.9%, respectively). Since MPs rejection were governed by both hydrophobic interaction in addition to size exclusion, the enhanced membrane surface hydrophilicity and selective nanochannels of MIL-101(Cr) plays critical role in suppressing the sorption and passage of MPs through PA layer. As shown in Fig. 9(f), the XPS analysis further suggests an increase of hydrophilic functional group (carboxylic group) for MOF0.20 membrane as implied by the decreased $\text{N}-\text{C}=\text{O}^*$ to $\text{O}^*-\text{C}=\text{O}$ ratio.

Paseta et al. (2019) modified TFC membrane by incorporating two types of hydrophilic MOF (ZIF-93 and HKUST-1, respectively) onto the polyimide support, followed by the formation of PA layer via IP process. The membrane incorporated with HKUST-1 showed higher pore size (6–9 Å) (Chui et al., 1999) than that of the ZIF-93 (3.6 Å) (Cacho-Bailo et al., 2015). Owing to the larger MOF pore size, all of the TFN membranes demonstrated excellent permeate flux (more than 2-fold) than the control membrane upon MOF incorporation. Furthermore, HKUST-1-modified membrane was able to show permeate flux of 37% and 136% higher compared to ZIF-93-modified membrane after subjected to 1 mg/L diclofenac and 1 mg/L naproxen solution, respectively at 20 bar. Although the permeability of MOF-incorporated membranes was found to improve, the naproxen rejection however decreased from 99.3% (control) to 98.3% for HKUST-1-modified membrane while it was maintained for ZIF-93-modified membrane. This can be mainly ascribed

to the larger pore size of HKUST-1 in relative to that of ZIF-93. Thus, naproxen (230.26 Da) can diffuse more easily through the MOF structure.

In an attempt to alleviate the environmental pollution due to the use pesticides and herbicides in agriculture sector, Rakhshan and Pakizesh (2015b) introduced 0.03–0.3 w/v% oleic acid surface modified- SiO_2 nanoparticles within the PA layer via IP process in order to enhance both permeate flux and triazines (atrazine, propazine, and prometryn) removal rate. At the optimum SiO_2 loading of 0.17 w/v%, the resultant membrane showed the highest permeate flux and rejection rate for atrazine ($\sim 18 \text{ L/m}^2 \cdot \text{h}$, 98%) propazine ($\sim 17 \text{ L/m}^2 \cdot \text{h}$, 91%) and prometryn ($7.5 \text{ L/m}^2 \cdot \text{h}$, 99%) compounds when filtration process was operated at 10 bar (each triazines at $100 \mu\text{g/L}$). Compared to the SiO_2 -modified membrane, NF control membrane demonstrated much lower water flux and triazines rejection, recorded at $\sim 14 \text{ L/m}^2 \cdot \text{h}$ and 94% for atrazine, $\sim 11 \text{ L/m}^2 \cdot \text{h}$ and 91% for propazine, $\sim 4.5 \text{ L/m}^2 \cdot \text{h}$ and 99% for prometryn, respectively. These results indicated that the addition of SiO_2 nanoparticle to the PA layer can improve both permeate flux and triazine rejection on account of the enhanced membrane surface hydrophilicity. Besides of the molecular size of triazine, the author highlighted that electrostatic interaction was the main factor in which the low rejection of propazine was due to its larger dipole moment (3.5 Debye). Molecules with high dipole moment become aligned so that their dipole charge is oriented to be opposite to the membrane surface charge. As a result, propazine can be strongly adsorbed onto the membrane surface.

Croll et al. (2019) revealed that the introduction of graphene oxide (GO) atop PA layer of SW30 commercial membrane via amine coupling reaction with ethylenediamine (ED) as a linker, was found to achieve 6% higher N-nitrosodimethylamine (NDMA) rejection (increased from 76.5% to 82.7%) after subjected to $890 \mu\text{g/L}$ NDMA solution for 2 h. The enhanced NDMA rejection can be ascribed to the sealing of surface defects by the covalently bonded GO layer, thus promoting size exclusion effect. However, the GO-modified membrane suffered from reduced permeate permeability (decreased from 1.2 to $1.05 \text{ L/m}^2 \cdot \text{h} \cdot \text{bar}$) despite the improvement observed in NDMA rejection. Such phenomenon can be caused by the blockage of covalently bonded GO nanosheets layer that inhibit water diffusion.

1.9. Challenges and future research trend

MPs are emerging as a new challenge to the worldwide. In order to solve this problem, there are many researchers tend to develop newer and appropriate water treatment technologies since conventional treatment process merely contribute in the degradation or removal of MPs compounds. Among so many water treatment methods, membrane technology such as NF and RO process have proven to be high effective in removing wide range of MP compounds. However, most of the reported studies have been carried out in the laboratories scale and usually limited to single MP component pollution approach in water bodies. Moreover, majority of the studies have performed the experimental work using synthetic MPs components. Hence, future research should focus on using the real water wastewater to address the problem for their better commercial exploitation. In addition, thousands of pharmaceutical products are produced in every year while only <5% of pharmaceuticals compounds are detected for treatment (Dharupaneedi et al., 2019). The pharmaceutical products such as preservatives, flavorants and antioxidants used in the cosmetics have rarely been investigated in wastewater treatment plants. Thus, more efforts are needed on treatment approaches particularly in their detection, toxicities, degradation and separation.

Based on the correlational analysis made between MPs removal rate and properties of MPs, it was revealed that size exclusion and hydrophobic interaction of MPs play important roles in the MPs removal efficiencies using RO/NF membranes. In order to determine the size of the MPs solute, van der Waals volume (vdV) of MPs is one of the best size

parameters to elucidate the size exclusion mechanism towards the given membrane structure. Therefore, the size exclusion is predictable based on molecular size of solute (i.e., edV) and the membrane pore structure (i.e., MWCO). Nevertheless, the degree of adsorption on membrane rejection performance is hard to predict due to the lack of quantitative parameters. Hensen solubility parameters (HSPs) may be the useful tool for determine various intermolecular interactions, including hydrogen bonding, dispersion and polar interaction between membrane and MPs solute (Belmares et al., 2004; Shin et al., 2019). Although HSPs of MPs solute can be calculated using the group contribution theory according to their chemical structure (Stefanis and Panayiotou, 2008), the information related to these parameters is still very limited.

With respect to MPs rejection performance, surface modification of membrane using hydrophilic materials may be an attractive strategy to alter the physicochemical properties of PA layer for enhanced MPs removal rate. However, the additional coated/grafted layer could act as a water transport barrier and led to low water permeability (Khoo et al., 2022). One of the approaches that has been widely used in recent years is to modify the PA layer via incorporation of inorganic nanomaterials as it could create selective nanochannels for water flux to pass through PA matrix while enhanced MPs removal rate efficiently. Besides, the construction of novel membranes is believed to be a potential and promising strategy to improve the MPs removal by tailoring size exclusion, adsorption, and electrostatic charge without jeopardize the water permeability. For instance, branched macromolecules such as star polymers and dendrimers that contain large intramolecular free volume and numerous functional groups can be attractive building blocks for membrane since it is able to produce highly crosslinked membrane with permeable selective layer structure.

2. Conclusion

This present review details the sources, classification and treatment techniques of MP compounds found in the last few decades as well as the potential hazard of MPs towards living organism. Extensive use of PhACs, pesticide, PCPs and EDCs have led to the increase of MPs concentration from ng/L to µg/L in the water bodies, and this causing huge impact to the environment, such as hormonal problems, cancer, infertility and so on. Based on our correlational analysis of the MPs rejection rate and properties of MPs examined, we found that size exclusion effect plays a major role in the MPs rejection rate as MPs with larger molecular weight can be easily removed by surface modified-RO and NF membranes. Other than size exclusion effect, additional mechanisms such as hydrophobic interaction (adsorption), electrostatic charge interaction (attraction), hydrogen bonding and dipole interaction with membrane PA layer can also influence the removal of MPs. In this respect, low dipole moments, larger molecules with similar charge to the membrane surface and low log Kow values tend to exhibit higher MPs rejection. Overall, the functionalization using hydrophilic monomers over the membrane PA layer and the incorporation of nanomaterials led to better MPs rejection performance as it can enhance membrane physiochemical properties in order to repel the MPs compounds.

Credit author statement

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Resources. **Nor salmi Abdullah:** Data curation.

Compliance with ethics requirements

This article does not contain any studies with human or animal subjects.

Declaration of competing interest

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

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