Bibliometric approach to the perspectives and challenges of membrane separation processes to remove emerging contaminants from water

Jéssica Stefanello Cadore, Lucas Fernando Fabro, Thuany Garcia Maraschin, Nara Regina de Souza Basso, Marçal José Rodrigues Pires and Vandré Barbosa Brião

ABSTRACT

The presence of contaminants in water is concerning due to the potential impacts on human health and the environment, and ingested contaminants cause harm in various ways. The conventional water treatment systems are not efficient to remove these contaminants. Therefore, novel techniques and materials for the removal of contaminants are increasingly being developed. The separation process using modified membranes can remove these micropollutants; therefore, they have attracted significant research attention. Among the materials used for manufacturing of these membranes, composites based on graphene oxide and reduced graphene oxide are preferred owing to their promising properties, such as mechanical resistance, thermal and chemical stability, antifouling capacity, water permeability, high thermal and electrical conductivity, high optical transmittance and high surface area. Membrane separation processes (MSP) can be used as secondary or tertiary treatment during the supply of wastewater. However, the efficient and accessible applications of these technologies are challenging. This study aims to demonstrate the main concepts of membrane separation processes and their application in the removal of emerging contaminants. This study reports bibliometric mapping, relevant data on studies using membranes as water treatment processes, and their viability in industrial applications. The main challenges and perspectives of these technologies are discussed in detail as well.

Key words | composite membranes, graphene, micropollutants, MSP, water treatment

Jéssica Stefanello Cadore (corresponding author)
Vandré Barbosa Brião
University of Passo Fundo (UPF), Faculty of Engineering and Architecture (FEAR), Postgraduate Program in Civil and Environmental Engineering (PPGEng),
Passo Fundo, RS,
Brazil
E-mail: *jescadore@gmail.com*

Lucas Fernando Fabro
Thuany Garcia Maraschin
Nara Regina de Souza Basso
Marçal José Rodrigues Pires
Postgraduate Program in Technology and Materials Engineering,
Pontifical Catholic University of Rio Grande do Sul (PUCRS),
Porto Alegre, RS,
Brazil

HIGHLIGHTS

- Electrostatic interactions and steric effects can occur between the membrane surface and the solutes, which can significantly influence the separation efficiency.
- MSP present significant advantages compared to other technologies used for CECs removal.
- The use of graphene-based materials, such as GO and rGO, for removing CECs in water treatment processes has been a rapidly growing area of interest in the recent literature.

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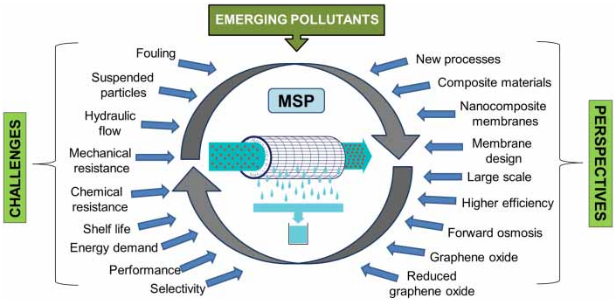
GRAPHICAL ABSTRACT



INTRODUCTION

Since the end of the 1970s, the monitoring of micropollutants in the environment has attracted attention (Hignite & Azarnoff 1977). This is due to the effects that it can cause, such as aquatic toxicity, genotoxicity, and endocrine disturbance in animals (Aquino *et al.* 2013).

Water bodies affected by effluent discharge face a serious impact on their quality and, consequently, on public safety and health (Stylianou *et al.* 2015). These contaminants arise from various anthropogenic sources, such as industries, agriculture, mining, and domestic residences (Ali *et al.* 2019a). Fatalities caused by water contamination with untreated industrial waste have caused public health crises in several countries such as Brazil, Jordan, China, USA, Bangladesh, South Africa, and Libya (Ward 2014; Power *et al.* 2018).

Due to the potential disturbances caused by their chemical composition in animal reproductive systems and humans, endocrine disruptors (EDs) have attracted significant attention and efficient treatments are being designed to remove them from potable and wastewater (Bila & Dezotti 2007). EDs can be classified as synthetic substances called xenoestrogens (pesticides, alkylphenols, phthalates, polychlorinated biphenyls, pharmaceutical agents, and bisphenol) and androgens and phytoestrogens (Bila & Dezotti 2007; Aquino *et al.* 2013). In general, these environmental micropollutants are known as contaminants of emerging concern (CECs) and have been a cause of increasing concern in recent years.

Methods of biological treatments such as filters or activated sludge are mostly insufficient for the removal of a wide range of contaminants with high toxicity content, such as drugs, pesticides, and heavy metals (Power *et al.* 2018). Other treatments such as coagulation and flocculation are also insufficient for the removal of CECs. Chlorination, used for the disinfection of water, in turn can result in undesirable effects such as unpleasant taste and odor. In addition, the use of chlorine to disinfect water generates concerns for its possible contribution to the production of carcinogenic by-products (Varma *et al.* 2013; Power *et al.* 2018). Other methods such as ozonization, ion exchange, and photocatalysis have the limitations or risks of generating undesirable by-products in treated water.

An alternative for the removal of various contaminants is membrane separation processes (MSP). A membrane is a two-phase separating barrier, which fully or partially restricts the transport of chemical species present in these phases (Habert 2006). Membranes that use the pressure gradient as a driving force are classified as ultrafiltration (UF), microfiltration (MF), nanofiltration (NF), and reverse osmosis (RO) (Power *et al.* 2018). Abundant research has been conducted to accomplish improvements in the functionality, control, characterization, and efficiency of membranes. Thus, the present study aims to conduct a bibliometric review of the literature on the general perspectives of membrane manufacturing, new methods and materials developed and the main challenges of their application in the removal of CECs.

METHODOLOGY

The review was organized through search for studies published to date in the Web of Science database, due with important and current information, and can serve as a facilitated access for the development of future studies.

In the first phase of the bibliometric research, the articles were selected through a combination of the terms 'membrane' AND 'emerging contaminants OR emerging pollutants OR emerging micropollutants' to contextualize the application of membranes for the removal of these environmental micropollutants (Figure 3). The use of the 'AND' operator in database searches indicates that both terms should appear in the publications (Visentin *et al.* 2019). When using the operator 'OR' at least one of the defined terms must be contained in the search. Data were selected based on titles, abstracts, and keywords of published studies, without delimiting the time period. Therefore, 426 publications were found, exported in BibTex format and imported into Rstudio software through the Bibliometrix package (RStudio Team 2020). In Table 1, recent studies are cited that have used membranes to remove different contaminants to contextualize their applicability.

In the second phase of the bibliometric research, Figure 4 was generated by the Bibliometrix package through the search for the terms 'membrane' AND 'industrial application OR large-scale' AND 'challenge' resulting in 262 publications, enabling a bibliometric approach of the main challenges of MSP in large-scale applications (RStudio Team 2020).

In the third phase, a search was conducted combining the terms 'MSP OR membrane separation process OR microfiltration OR ultrafiltration OR nanofiltration OR reverse osmosis' AND 'graphene oxide OR GO OR reduced graphene oxide OR GOr'. Thus, the combination of previously determined terms found searches in titles, abstracts, and keywords of the relevant publications available in the Web of Science database. The research comprised a total of 1,508 publications. These files were analyzed according to the year of publication (Figure 8).

Moreover, a literature review was carried out seeking the contextualization of the use of MSP in water treatments aimed at the removal of CECs, including recent studies, new materials and processes, and the main challenges and perspectives for future studies.

MEMBRANE SEPARATION PROCESSES FOR WATER TREATMENT

MSP that use pressure gradients as the driving force are one of the most promising technologies for water purification. These technologies comprise MF, UF, NF, and RO. The membranes impel barriers to the passage of different pollutants and remove microorganisms, suspended particles, and colloidal materials or dissolved solids. Thus, the systems act by inhibiting the passage of micropollutants, forming a barrier to block the contaminants, acting through various separation mechanisms. These consist of a physical system, operating without heating and with high selective control, and these characteristics are some of their advantages when compared to other methods (Habert 2006; Alonso *et al.* 2018; Aquarden Technologies 2019). The MSP have a simple operation, thereby allowing compact projects that do not require a large area for implementation. Therefore, it is possible to combine them with other treatment methods without the use of additive products (Rodríguez

Table 1 | Recent studies using membranes for water treatment

Retention mechanisms	Materials	Applications	Reference
Nanofiltration	Polyacrylonitrile and Fe_2O_3	Diazinon removal	Pordel *et al.* (2019)
Nanofiltration	Chitosan and graphene oxide	Diazinon removal	Chen *et al.* (2019a)
Ultrafiltration	Polyethersulfone	Caffeine removal	Acero *et al.* (2017)
Ultrafiltration	Guanidyl-functionalized graphene/polysulfone mixed matrix	Antibacterial and antimicrobial	Zhang *et al.* (2019)
Filtration adsorption	β-cyclodextrin polymers	Bisphenol A removal	Wang *et al.* (2019a)
Solid-phase extraction	Porous carbon derived from amine-functionalized MIL-125 metal-organic framework	Bisphenol A and 4-tert-butylphenol	Sánchez *et al.* (2019)
Nanofiltration	Graphene oxide/attapulgite composite membranes	Dyes removal	Wang *et al.* (2019b)
Reverse osmosis	Industrial membrane – RE2521-SHF	Ciprofloxacin removal	Alonso *et al.* (2018)
Microfiltration	Industrial membrane – hydrophobic and negatively-charged PVDF	Ciprofloxacin removal	Guo *et al.* (2018a)
Nanofiltration/reverse osmosis	Polyamide thinfilm composite (TFC) membranes	Pesticides	Fini *et al.* (2019)

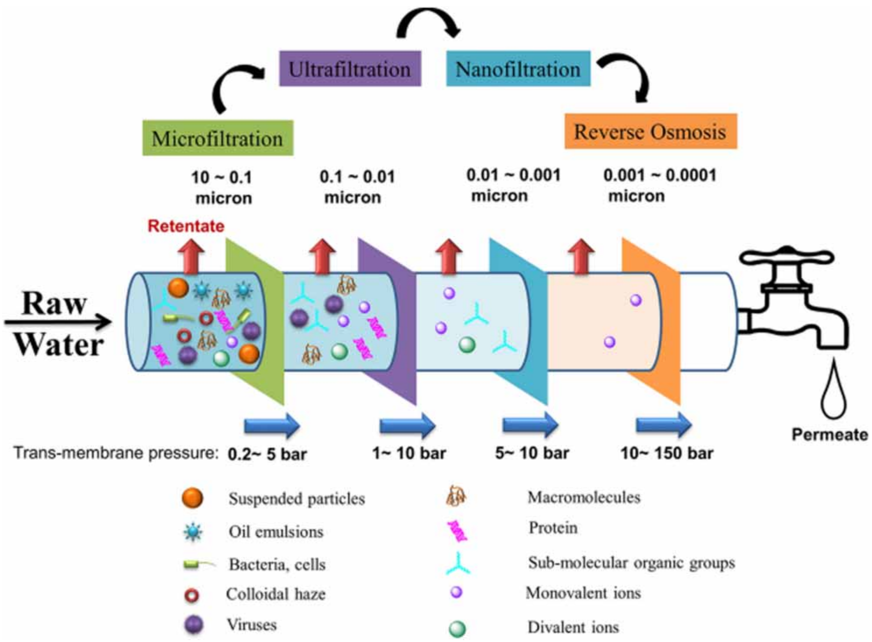


Figure 1 | Schematic of a water treatment system by MSP. *Source:* Liao *et al.* (2018).

et al. 2006; Mulder 2012). Figure 1 presents a summary of a water treatment system through an MSP.

Pore size is not the only factor responsible for the MSP. Electrostatic interactions and steric effects can occur between the membrane surface and the solutes, which can significantly influence the separation efficiency (Heberer *et al.* 2002; Garcia-Ivars *et al.* 2017a, 2017b). The pH value and isoelectric point (IEP) can also be a major contributor, as it can affect the hydrophilicity and solubility of substances (Licona *et al.* 2018). Rigueto *et al.* (2020) demonstrated this behavior in MSP to remove caffeine from water, as shown in Figure 2.

Several studies have been conducted on the mechanisms of the transport of solutes in membranes. However, further studies are required to understand the influence of the properties of the solute, membrane parameters, feed water parameters, and operational parameters on the mechanism (Nghiem *et al.* 2010; Simon *et al.* 2012; Yüksel *et al.* 2013) to improve the technique and increase the efficiency and durability of membranes.

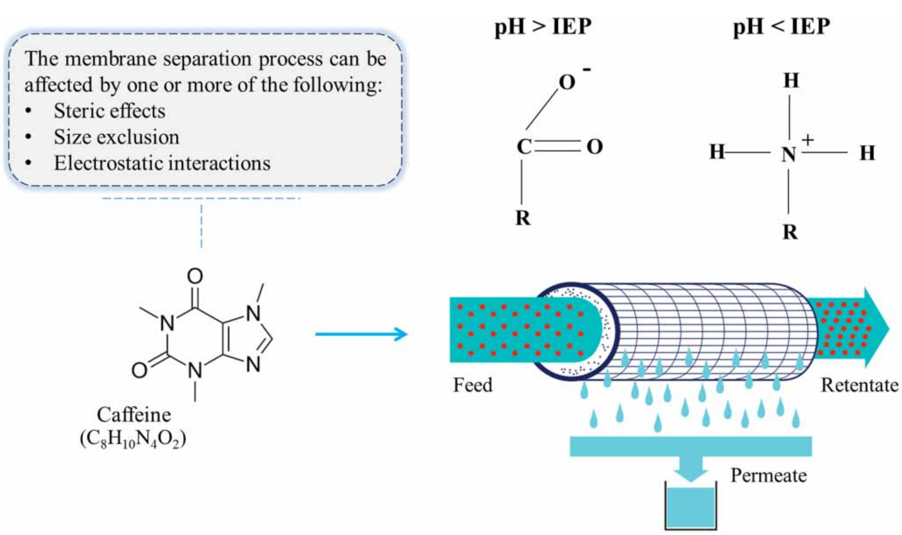


Figure 2 | Effect of feed solution pH on the functional groups on the surface of the membrane. *Source:* Rigueto *et al.* (2020).

Depending on the chemical composition and process type, membrane materials can vary widely. However, membrane manufacturing aims to achieve an ideal material that has mechanical strength, maintains high flow, and is selective for the desired permeate component. The latter two are mutually conflicting. Achieving high selectivity is usually possible using membranes with small pores, thereby preventing a high hydraulic flow. Also, the resistance of the membrane is directly related to its thickness. Thus, the ideal characteristics of the physical structure of membranes are thin layer of material, small pore size range, and high porosity (Judd & Jefferson 2003).

MEMBRANE SEPARATION PROCESSES TO REMOVE EMERGING CONTAMINANTS

CECs encompass a wide range of pollutants such as drugs, pesticides, hormones, and EDCs, among others. They are generally present in concentrations of micrograms or nanograms per liter, but even at these concentrations they can affect aquatic biota (La Farre *et al.* 2008; Richardson & Kimura 2016). As they have different chemical characteristics, their removal is limited in conventional water or sewage treatment plants, as they are not designed for this purpose.

Effective treatment methods for these pollutants have been studied recently, including MSP. This is mainly because the processes remove soluble molecules having small dimensions. Thus, studies that have used membranes for the removal of pharmaceutically active compounds (PhACs), caffeine (Taheran *et al.* 2016; Garcia-Ivars *et al.* 2017c; Mendez *et al.* 2017; Alonso *et al.* 2018; Couto *et al.* 2018; Egea-Corbacho *et al.* 2019; Lopera *et al.* 2019), pesticides (Vásquez *et al.* 2018; Fini *et al.* 2019; Nguyen *et al.* 2019; Pordel *et al.* 2019), and PhACs (Morone *et al.* 2019; Ouyang *et al.* 2019; Wang & Huang 2019), among others, have been increasingly developed.

MSP present significant advantages compared to other technologies used for CECs removal. The MSP are widely used in a large number of applications (Singh 2014; Zheng *et al.* 2015) producing high-quality permeates without formation of toxic by-products or metabolites (Kim *et al.* 2018). Despite the great diversity of methods, full-scale advanced oxidation processes (AOPs) have mainly been used for drinking water treatment and water reuse applications (Rodriguez-Narvaez *et al.* 2017; Miklos *et al.* 2018). In addition, the generation of transformation products (TPs) with toxic potential was observed in the water treatment by different AOPs (Luo *et al.* 2014; Sharma *et al.* 2018). The efficiency of the adsorption processes is significantly affected by the presence of organic matter or suspended particles and the used adsorbent must be regenerate or properly disposed of (Dhangar & Kumar 2020; Vieira *et al.* 2020a). MSP are also phase-changing technologies; however, the treatment/disposal of rejected effluent can be handled more easily; for instance, by coupling with AOPs in hybrid systems (Ganiyu *et al.* 2015; Dhangar & Kumar 2020). The biological treatments could present low degradation capacity for some compounds (e.g. EDCs) and significant concentrations of hydrophobic and recalcitrant contaminants remain in the biosolids, which must be disposed of properly (Ahmed *et al.* 2017; Rodriguez-Narvaez *et al.* 2017). On the other hand, MSP present high removal efficiency for EDCs (Vieira *et al.* 2020a), low solid waste generation (Crini & Lichtfouse 2019), and can be associated with biological processes increasing their removal capacity for a wide range of CECs (Luo *et al.* 2014; Dhangar & Kumar 2020).

Figure 3 shows the cluster of words found in the bibliometry. CECs include endocrine disruptors, bisphenol A, pharmaceutically active compounds, antibiotics and pharmaceuticals; that is, most studies on CECs have a focus on PhACs and personal care products (PPCPs). One can visualize the main treatment methods used in scientific literature, including UF, ND and RO, combined with other processes like adsoption, AOPs and solid-phase extraction. The membrane bioreactor (MBR) also appears in this mapping. It is applied in municipal sewage treatment plants, and may be linked to other processes such as pre- or post-treatment including biodegradation and activated sludge.

Several studies have been conducted to investigate the removal of CECs through membranes. These have made necessary modifications through new materials and have strived to achieve greater efficiency and enable the removal of specific contaminants. Table 1 shows recent studies that have used MSP for water treatment.

Mamo *et al.* (2018) investigated the removal of 13 PhACs and 20 of their metabolites and different TPs in the different treatment steps of urban raw wastewater (sewer, primary treatment, MBR and RO/NF). The analgesic acetaminophen, which was found at the highest concentrations in the sewer and influent samples (18–74 μg L^{-1}), was fully eliminated during MBR treatment. Those PhACs that were only partially removed after the MBR were almost completely removed (>99%) by the RO membrane. For similar average permeate fluxes (18 L m^{-2} h^{-1}), the NF membrane showed high removal efficiencies (>90%) for all of the

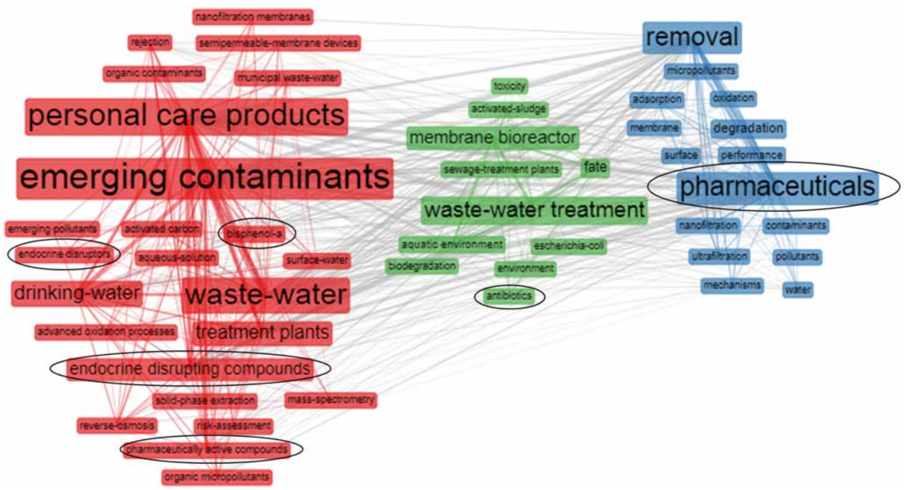


Figure 3 | Combined mapping of the most cited keywords in the domain of 'membrane' AND 'emerging contaminants OR emerging pollutants OR emerging micropollutants'.

PhACs and their metabolites, though lower than those achieved by the RO membrane.

Rodriguez-Mozaz *et al.* (2015) used a pilot plant based on MF and RO applied to the tertiary treatments of conventional waste-water treatment plants. The authors noted that only filtration through the combined MF–RO system in the pilot plant provided quantitative removal for most of the pharmaceuticals (>97%).

Meanwhile, in their studies, Fini *et al.* (2019) investigated the removal of specific pesticides – two phenoxy acid herbicides (2-methyl-4-chlorophenoxy-acetic acid and 2-methyl-4-chlorophenoxy-propionic acid) and a pesticide TP (2,6-dichlorobenzamide) – through NF and RO membranes. The authors have verified that the increase in rejection values influences the increase in the recovery values of the membranes. This is attributed to the effects of pore blocks. This effect is known as fouling, and occurs when there is a buildup of micro particles on the membrane surface. On reaching the extremities, it results in a pressure drop and reduction of the permeate.

Pinto *et al.* (2018) conducted studies with an MBR to evaluate its efficiency in the filtration process of industrial wastewater containing pesticides and hazardous residues. The results of the treatment have indicated that it is necessary to couple MF or UF with the existing conventional pretreatment of RO to improve the aspects of final water quality and minimize the problems of fouling, thus reducing operating costs. It is known that in MBRs, fouling problems are mostly due to the presence of biomass (Zheng *et al.* 2011). Coupling different treatment processes has been tested as a promising alternative in MSP.

An experimental study combining biological treatment with flocculation and UF membrane technology was conducted, separately and in combination, seeking to increase insight into the capability of such treatment processes to remove CECs. UF processes yielded better results than their separate use, leading to the highest removal rates of 15 of the 17 compounds (Melo-Guimarães *et al.* 2013).

Challenges and future perspectives

According to Figure 4, among the most cited words in an article titles about the challenge of industrial application

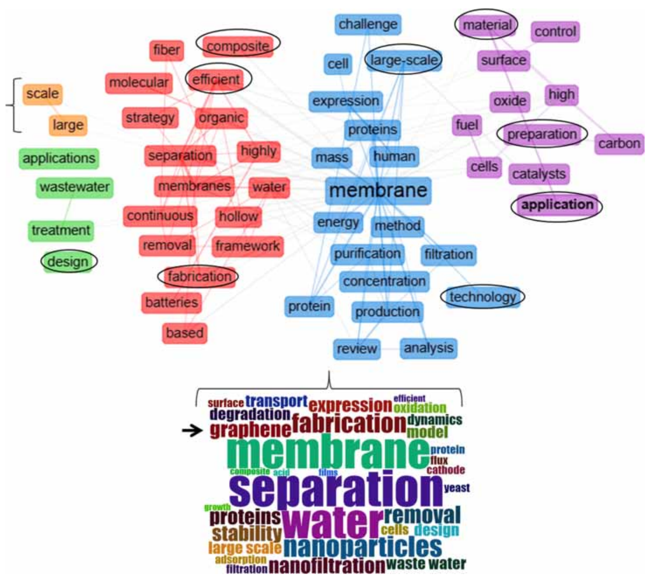


Figure 4 | Combined mapping of the most cited words in the article titles and word cloud with the most cited keywords in the domain of 'membrane' AND 'industrial application OR large-scale' AND 'challenge'.

of membranes, are: technology, composite, efficient, preparation, fabrication, application and design. Furthermore, in the word cloud it is possible to observe the word graphene, which can be a challenge in the manufacture of composite membranes at large scale. According to Ali *et al.* (2019b), the use of carbon nanotubes (CNT) can have some drawbacks including less dispersion in aqueous solution and low sorption affinity to use for large-scale production of organic and inorganic CNT composites.

Achieving satisfactory performance of membranes is also one of the challenges. It is influenced by the design of the membranes (pore size and distribution of materials), which is directly related to the efficiency of the filtration process (Nasir *et al.* 2019).

For large-scale membrane processes, like industrial or other commercial uses of membranes, large membrane areas are required (Obotey Ezugbe & Rathilal 2020). Most studies are being developed on a laboratory scale, and require large-scale applications to obtain an actual estimate of the economic viability of membranes (Nasir *et al.* 2019).

The main challenges for the application of membranes on an industrial scale, according to Xiao *et al.* (2019), are: more efficient fouling control, higher cost-effectiveness, enhanced pollutant removal and more reasonable positioning in the application fields.

Colmatation is still a challenging factor in membrane processes. This is due to the accumulation of pollutants on the surface, causing fouling and a drop in treatment efficiency. This problem is constantly encountered and it is one of the main challenges in MSP.

The reuse of membranes is also a challenge that has been studied. Membranes have a short life cycle in comparison with other treatment methods, thereby increasing the cost associated with the process. This is due to the wear of membranes by chemical cleaning processes, which are necessary to reduce fouling. The wear is caused by the accumulation or adsorption of micropollutants on the surface and within the pores of the membranes.

NANOCOMPOSITE MEMBRANES

Nanocomposite membranes are a new class of membranes manufactured by combining polymeric materials with nanomaterials. These are emerging as a promising solution for the treatment of water and effluents due to their multiple functionalities (Zhang *et al.* 2019). Given the variety and availability of organic and inorganic nanomaterials, such as titanium dioxide (TiO_2), silicon dioxide (SiO_2), CNT, and graphene, several studies have investigated their challenges, opportunities, and future potential (Lee *et al.* 2011; Pendergast & Hoek 2011; Alzahrani & Mohammad 2014; Daer *et al.* 2015; Goh & Ismail 2018; Anand *et al.* 2018; Johnson *et al.* 2018; Song *et al.* 2018; Xu *et al.* 2019).

Advanced nanocomposite membranes can be designed to meet specific water treatment applications, including the removal of CECs (Karkooti *et al.* 2018) by adjusting their structure and physicochemical properties such as hydrophilicity, porosity, load density and thermal and mechanical stability (Koenig *et al.* 2012; Ganesh *et al.* 2013; Zhang *et al.* 2013; Ionita *et al.* 2014; Wang *et al.* 2016, 2018; Guo *et al.* 2017; Zhu *et al.* 2017; Köhler *et al.* 2018). Additionally, unique features have been introduced such as antibacterial, photocatalytic or adsorbent capabilities (Yu *et al.* 2013; Zambianchi *et al.* 2017; Indherjith *et al.* 2019; Zhang *et al.* 2019) and attempts have been made to reduce the mass and cost of the generally used materials (Sun *et al.* 2016; Sun & Bai 2017; Sun & Li 2018).

Halakoo & Feng (2020) demonstrated the possible linkages between GO and polyethyleneimine polymer (PEI), obtained through fabricate of layer-by-layer. GO nanosheets can interconnect with PEI through electrostatic interactions, which are the main driving force for building up the PEI/GO bilayer. By pairing PEI and GO, strong intermolecular hydrogen and chemical bonding can also occur, as illustrated in Figure 5.

For water treatment processes, it is ideal that materials with good mechanical resistance and controlled hydrophobicity be used (Power *et al.* 2018). The surface hydrophilicity can be indicative of the permeate flux and anti-fouling performance of membranes (Bi *et al.* 2020). Mukherjee *et al.* (2018) developed UF membranes using chitosan as an intermediate matrix between the substrate and graphene oxide (GO) coating. Aiming at the removal of the pesticide atrazine, they achieved $>95\%$ efficiency in the process. Chitosan is an appropriate matrix for the manufacture of membranes applied to water treatment, although its limited mechanical resistance restricts its application.

NF membrane was developed using flexible GO and nylon 6 multilayers, through a layer-by-layer assembly process, combining electrospinning and electrospray techniques. The membrane demonstrated a high flow of organic solvents, which were chemically stable for solvent separation processes. Due to its advantages, GO is increasingly being used in MSP (Chen *et al.* 2018).

Pordel *et al.* (2019) used the polyacrylonitrile and Fe_2O_3 electrospun nanofibrous membrane to remove the pesticide diazinon. The results showed that the highest removal

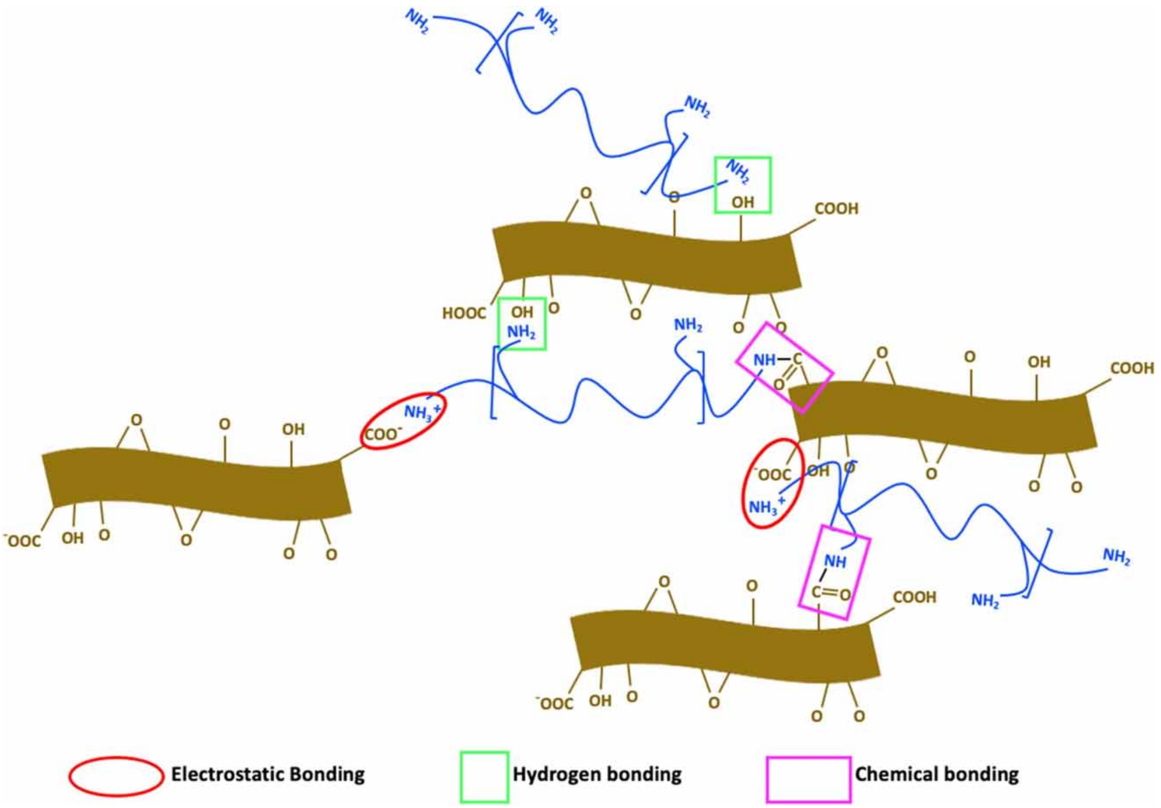


Figure 5 | The possible linkages between GO and PEI. *Source:* Halakoo & Feng (2020).

efficiency under optimal conditions (pH 7, diazinon concentration of 10 mg/l, and 0.1% nanoparticles) was 83.7%, can be applied as an effective and environment-friendly material. Thus, studies using new materials have constantly been carried out, in search of improvements in the efficiency of removal of micropollutants.

Diclofenac degradation system using reduced GO composites combined with titanium dioxide (rGO/TiO_2) and peroxodisulfate (PDS) was evaluated (this was used as an electron acceptor to accelerate the photocatalytic activity of this material). For synthesis, composite materials were triggered by visible radiation with high photocatalytic performance (vis). The vis-rGO/TiO_2/PDS system showed excellent properties in diclofenac degradation. Hence, there is a vast possibility of modifications in the configurations of membranes, as well as their various applications in water treatment processes (Chen *et al.* 2019b).

The effect of incorporating four GO derivatives on the physico-chemical characteristics and permeation properties of polyethersulfone (PES) membranes were reported. The OG derivatives used in the study included graphene nanoplatelet (GNP), GO nano-sheet, longitudinally uncompressed GO nano-tape (GONR-L) and helical uncompressed OG nano-tape (GONR-H). The addition of graphene nanoparticles up to 0.1 wt.% improved the water flow (due to an increase in porosity) and membrane hydrophilicity. Among the effects observed, the results showed that GONR-L at its ideal load (0.1 wt.%) provided the maximum water flow (70 $L/h/m^2$ at 60 psi), rejection of matter by organic membranes (59%) and antifouling properties (30% improvement compared to pure PES membrane) (Karkooti *et al.* 2018).

Recently, research has been conducted in the domain of rationally assembled two-dimensional (2D) nanomaterials (objective-oriented preparation strategies) in lamellar architectures and these have been used as building blocks for advanced membrane devices in the domains of energy and environment (Figure 6). Distinct advantages such as reduced size, energy efficiency and ultra-fine structure have demonstrated gains in technical characteristics (Sun & Li 2018). Additionally, multilayer systems have extended structural properties.

Other analogous types of 2D graphene structures that are derived from their crystals dispersed in layers, include hexagonal boron nitride (h-BN), graphitic carbon nitride (gC_3N_4), transition metal dichalcogenides (TMDCs),

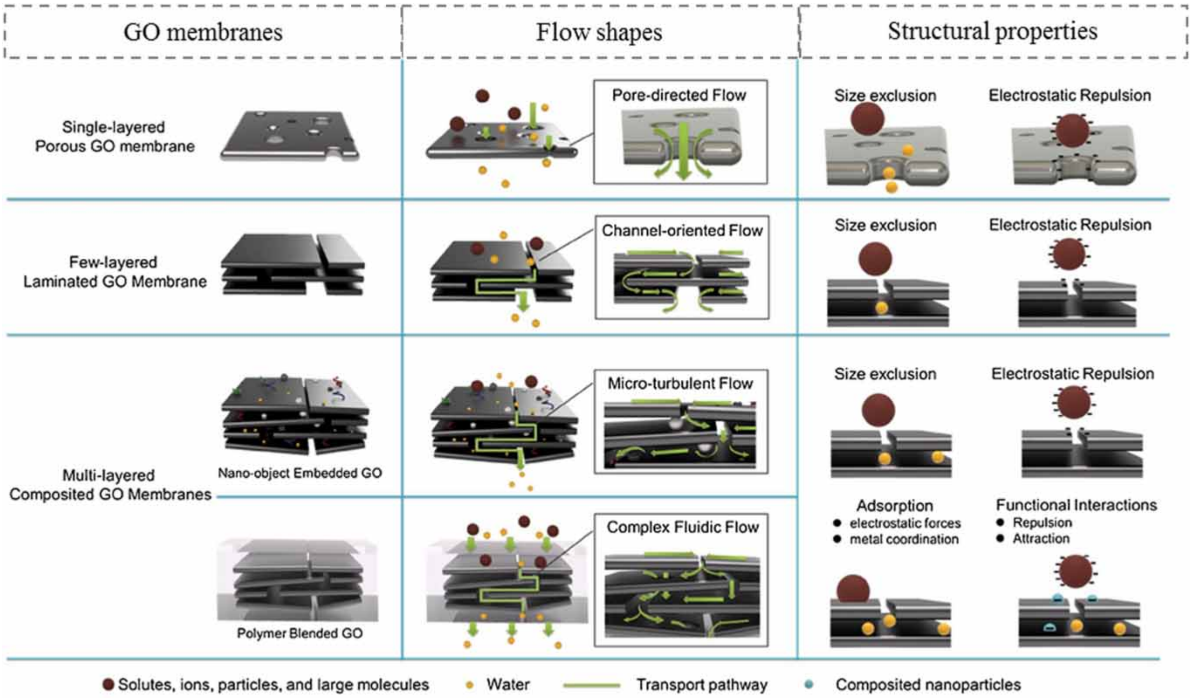


Figure 6 | Schematic diagrams of the functional structures of GO membranes. *Source:* Sun & Li (2018).

among others. These have also been extensively investigated in various domains of science (Wu *et al.* 2012; Xu *et al.* 2013, 2019; Biscarat *et al.* 2015; Shen *et al.* 2016; Ma *et al.* 2017).

Based on the molecular printing technique (Lu *et al.* 2019; Sajini *et al.* 2019), ultrathin films/nanocomposite membranes have become attractive in the domains of purification and separation. Membranes impregnated with antifouling ions and term-sensitive to GO and silicon dioxide printed with europium ($Eu^{3+)}$) were developed by Lu *et al.* (2018), to selectively separate lanthanum (La^{3+}), gadolinium (Gd^{3+}), and samarium (Sm^{3+}) from europium. GO nanosheets and modified silicon dioxide nanospheres ($kSiO_2$) were synergistically stacked on substrates modified with polydopamine to form nanocomposite membranes.

Typically existing in nanocrystalline form, the materials employed include porous ceramics, such as Al_2O_3, TiO_2, ZrO_2, ZnO, and SiO_2 (Bouazizi *et al.* 2017; Szczepanik *et al.* 2017). Composite membranes containing two or more inorganic materials have also been developed, including TiO_2–SiO_2, TiO_2–ZrO_2, and Al_2O_3–SiC (Anisah *et al.* 2019; Farahani & Vatanpour 2018; Guo *et al.* 2018b) as well as several other nanoparticle composites, such as Ag–TiO_2, Zn–CeO_2, and zeolites (Mohmood *et al.* 2013; Kumar *et al.* 2014; Lee *et al.* 2016).

Several authors have researched the removal of emerging contaminants in natural and wastewater by the use of isolated nanomaterials (Sarkar *et al.* 2018; Sophia & Lima 2018; Indherjith *et al.* 2019; Rasheed *et al.* 2019; Sánchez *et al.* 2019; Khalil *et al.* 2020) or incorporated different matrices in membranes to break the paradox between membrane permeability and selectivity (Goh *et al.* 2014, 2016; Wang *et al.* 2015; Ali *et al.* 2019b), improving separation technology (Ji *et al.* 2017; Sheikh *et al.* 2020).

Carbon-based nanocomposite membranes were employed in a continuous mode of operation for the first time. For this purpose, reduced graphene oxide (rGO) doped with nitrogen using melamine as the source (rGO-M), was included as a catalytic active phase in a poly (vinylidene fluoride) matrix (PVDF). The performance of the reduced composite membrane (rGO-M-PVDF) was demonstrated by the degradation of three fluoroquinolone antibiotics to the level of ppb in ultrapure water ($100\,\mu g\,L^{-1}$ each), with removing pollutant rates in the range of 2.05 at 2.73 $mg\,m^{-2}\,h^{-1}$. Conversions of pharmaceutical products in the range of 54–91% were effected after 24 hours of operation in full continuous mode and high resistance to fouling was observed by this new catalytic membrane (Vieira *et al.* 2020b).

Advanced, new and consolidated methods can remove CECs with or without limitations. However a huge local diversity of CECs and the water matrix becomes essential in the optimization for each application (Rizzo *et al.* 2019).

There are seasonal variations that can influence the removal of CECs (Kumar *et al.* 2019). All of these factors reinforce the importance of research on MSP and the use of membrane nanocomposites (Esfahani *et al.* 2019; Salazar *et al.* 2020; Shakak *et al.* 2020) for this purpose.

FUTURE TRENDS OF GRAPHENE

Graphene derivatives, such GO and rGO, have appropriate solubility, excellent processing ability, moderate conductivity, high specific surface area, good biocompatibility, and are an abundant and economic resource. As a novel nanomaterial family, GO and rGO are promising candidates to inhibit bacterial growth due to the ease of production, functionalization, and promising biocompatibility, showing excellent performance in disinfection and a broad-spectrum bactericidal ability (Anand *et al.* 2019; Han *et al.* 2019).

GO sheets are negatively charged when in aqueous environments. Compared to graphene membranes, where nanopores provide transport channels, stacked GO allows the passage of molecules, which provides another means of separation (Nair *et al.* 2012; Yang *et al.* 2017). Figure 7 shows the molecular structure of (a) graphene and (b) GO, with the latter representing in detail the presence of oxygen-containing functional groups located in the basal planes and edges (Yang *et al.* 2017).

The GO and reduced rGO can be used in a broad range of applications and show promise as an integral material to the progression of new and improved products (Smith *et al.* 2019). As far as effective adsorbents are concerned, graphene-based materials, for example, GO and rGO, are efficient adsorbents for composite membranes in removing CECs. GO and rGO show availability of different functional groups, such as epoxide, hydroxyl, carboxyl, and carbonyl, on its surfaces. The oxygen-containing groups allow GO to act as an adsorbent for a broad range of pollutants. In addition, the high specific surface area and the possibility of formation of π-π interaction between the aromatic ring of GO and rGO make it a relevant adsorbent for fabricating nanocomposite membranes for effective removal of CECs pollutants (Borthakur *et al.* 2018; Khalil *et al.* 2020; Madima *et al.* 2020). Carbon-based nanomaterials thus offer new possibilities for membrane design and fabrication, where the properties of the carbon nanomaterial can be utilized to influence membrane properties or include additional functionalities (Esfahani *et al.* 2019).

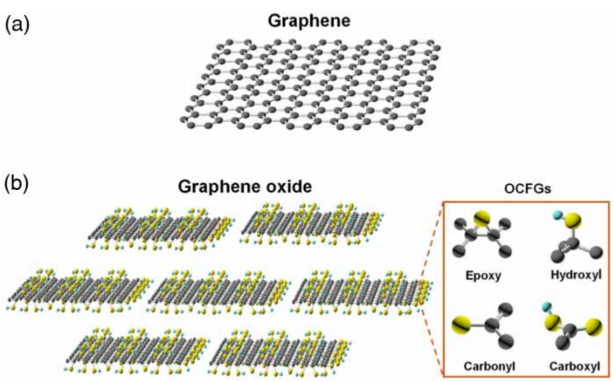


Figure 7 | Scheme of the molecular structure of (a) graphene and (b) GO. *Source:* Yang *et al.* (2017).

Recent work reports on the functionality of graphene and its derivatives when added to composite membranes in order to remove these harmful pollutants (Pedrosa *et al.* 2019; Khalil *et al.* 2020; Madima *et al.* 2020; Vieira *et al.* 2020a).

Khalil *et al.* (2020) evaluated the efficacy of porous graphene as a potential candidate for the removal of six widely utilized pharmaceuticals from their aqueous solutions, such as atenolol, carbamazepine, ciprofloxacin, diclofenac, gemfibrozil and ibuprofen. The results revealed high removal efficiencies for trace concentrations of all selected pharmaceutical contaminants (>99%) at a 100 mg/L dose of graphene (Khalil *et al.* 2020). Another study published by Pedrosa shows a membrane fabricated with N-doped GO powdered material with high catalytic activity for persulfate activation and phenol degradation (Pedrosa *et al.* 2019).

Ma *et al.* (2020) researched the performance of the polyvinylidene fluoride (PVDF) UF membrane with GO-polyethylene glycol as an additive. It can be concluded that the composite membrane has satisfactory performance in improving the membrane hydrophilicity, permeability, and antifouling performance in practical applications compared with a pure PVDF membrane (Ma *et al.* 2020).

NF membranes with high performance in water purification are studied by Cheng. The membranes' retention of rhodamine B is 85.03% with GO, 95.43% with GO/polyacrylamide and 97.06% with rGO respectively (Cheng *et al.* 2019).

GO-based membranes and their use as adsorbent of a mixture of selected CECs, such PhACs, pigments, PPCPs and surfactants are explored by Zambianchi and this work showed high affinity for most of the targeted organic compounds with efficiency higher than 90% after 4 h of treatments (Zambianchi *et al.* 2017).

Graphene-based materials were applied for the removal of PhACs in water and were demonstrated to be stable after their use as catalysts in peroxymonosulfate activation, and had a high adsorption capacity for a mixture of CECs (antibiotics) (Solís *et al.* 2020). A novel modified GO membrane with stable structures, large adsorption capacity and high flux has been successfully developed for efficient removal of bisphenol A by Chen *et al.* 2020a. The work accomplished by Sheng *et al.* shows membrane composite filtration with rGO and CNT for sulfamethoxazole removal in water (Sheng *et al.* 2020).

The study performed by Chu shows a modified ceramic membrane functionalized with GO (ceramicGO) that exhibited higher retention of pollutant compounds: pharmaceuticals (ibuprofen and sulfamethoxazole) and inorganic salts (NaCl, Na_2SO_4, $CaCl_2$, and $CaSO_4$). While the retention efficiencies of pharmaceuticals and inorganic salts in the pristine membrane were 15.3% and 2.9%, respectively, these increased to 51.0% and 31.4% for the ceramicGO membrane (Chu *et al.* 2017).

The Chós study reveals that rGO and GO nanoribbons (GONRs) are used to fabricate a composite membrane that exhibits ultrafast water permeance and precise molecular separation, >95% for dyes with a hydration radius greater than 5 Å (Cho *et al.* 2019).

Oh *et al.* (2017) investigated the potential of GO membranes for removing CECs and multivalent ions from water, demonstrating the promise of using the GO membrane as a pH-responsive membrane. Fryczkowska *et al.* (2020) obtained composite membranes with good transport properties (~390 L/m^2 h) and a high degree of protein retention (85%). Thus, they described that reduced GO has biocidal properties, which depends on the size of nanoparticles and the type of microorganisms. Yoon *et al.* (2020) investigated the effect of the graphene nanomaterial content on the hydrophilic and antifouling properties of the polysulfone membrane. The membrane with GO at 0.10 wt.% was found to provide the best membrane fouling reduction, pollutant removal rate, and best permeability.

Akther *et al.* (2020) explored the effect of GO flake lateral size on forward osmosis (FO) membrane performance and demonstrated that better antifouling property membrane performance can be considerably influenced by smaller GO flakes, improving the membrane flux and selectivity. This demonstrates the applicability of the material in other water treatment processes.

Figure 8 shows the evolution of scientific production regarding the number of annual publications related to researches about MSP with graphene, GO and rGO. Older

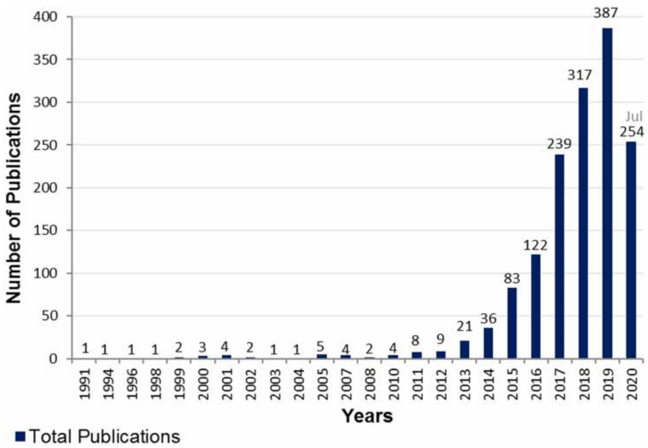


Figure 8 | Temporal evolution of the scientific literature in the domain of 'MSP OR membrane separation process OR microfiltration OR ultrafiltration OR nanofiltration OR reverse osmosis' AND 'graphene oxide OR GO OR reduced graphene oxide OR GOr'.

publications were also accessed. It is possible to observe that there are no studies prior to 1991; however, only from 2011 onwards, studies began to intensify, which indicates that studies using graphene-based materials are recent in MSP. This may be due to graphene derivatives having been discovered in recent decades, and their application in membrane processes being recent. In the last decade, there has been a significant rise in research containing graphene and its derivatives, which is due to its excellent properties, enabling application in several processes, especially those involving environmental remediation. According to Sophia *et al.* (2016), they can be used to reduce the burden of pollution by the adsorption and decomposition of persistent organic and inorganic pollutants. However, few studies have reported on the removal of CECs from membranes containing these nanomaterials, thus enabling several opportunities for further research.

ALTERNATIVE MEMBRANE TECHNOLOGIES

Forward osmosis

With the progress of studies involving membranes, especially those of RO, interest in osmosis engineering applications has increased. FO has applications in water and effluent treatment, food processing, and water desalination (Cath *et al.* 2006).

The growing interest in FO systems for applications, mainly water desalination, is driven by their low energy consumption and low probability of fouling. Under the assumption that the intensity of membrane fouling is directly proportional to the applied pressure, FO exhibits less reversibility to the compaction of the suspended matter layer on the membrane surface (Tufa *et al.* 2019). However, a negative impact observed in FO membranes is related to the decreased flow, which results from the pore blockage of the support layer (Boo *et al.* 2013). Figure 9 shows the schematic diagram of an FO system and its settings for water/wastewater treatment.

Chen *et al.* (2020b) reported that new membranes in the FO system made of electrospun nanofibers impregnated with titanium dioxide nanoparticles (TiO_2) and composite titanium/silver dioxide nanoparticles (TiO_2/Ag) were used to compare the antimicrobial performance and rejection of tetracycline-resistant genes. The characterizations revealed that TiO_2/Ag nanoparticles were dispersed evenly in polysulphone nanofibers and resulted in a membrane that exhibited excellent physicochemical properties, filtration, and antifouling performance in real wastewater.

Seeking to improve the performance of the FO process, Bagherzadeh *et al.* (2020) developed membranes composed of polyethersulfone/polyamide through the incorporation of particles of GQDs@UiO-66-NH_2 (obtained by synthesizing UiO-66-NH_2 metal-organic frameworks modified with graphene quantum dots (GQDs). The authors reported the advantages of this process including the increase of water flow, improvements in selectivity and increased compatibility with the matrix of the polyamide layer.

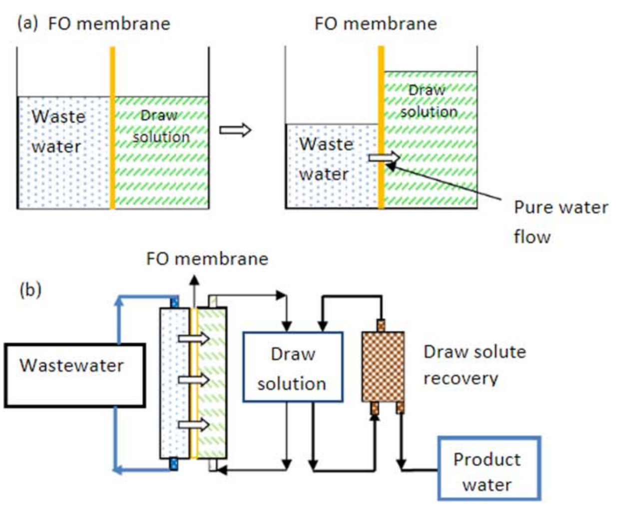


Figure 9 | Schematic diagram (a) of the FO system and (b) of the FO configuration for water/wastewater treatment. *Source:* Hai *et al.* (2014).

Electrodialysis and electrodialysis reversal

Electrodialysis (ED) is a process for transporting salt ions from one solution through an ion exchange membrane to another solution, under the influence of an applied electric field (Mulder 1996). The electrodialysis reversal (EDR) consists of the improved ED process, where the direction of the ionized transport is changed, changing the polarity of the electrodes periodically. This system is carried out to improve the traditional ED system, avoid fouling, and increasing the efficiency of the process (Tanaka *et al.* 2003; Kalogirou 2005). The ion exchange membranes used in the ED and EDR processes are categorized into cation exchange membranes and ion exchange membranes. The ion exchange membranes used in the ED and EDR processes are considered ion exchange resins in the polymeric matrix. In an electrodialysis cell, an applied voltage moves between the electrodes, and the membranes cause the ions to become trapped in the concentration channels, where there is a blockage of negative or positive ions. The behavior of the process can be seen in Figure 10.

ED was introduced and used in industrial applications even before RO (Bernardes *et al.* 2014). Although other membrane-based water treatment processes, such as UF and RO, have been used to reuse drinking water, ED/EDR processes are not very common in recovering municipal wastewater, since EDR can remove only ionized species and therefore, it is not possible to remove organic matter, flavor or odor (Warsinger *et al.* 2018). ED systems are normally used to desalinate mainly brackish waters at lower salinities (Metcalf and Eddy *et al.* 2007).

Gabarrón *et al.* (2016) studied to evaluate the occurrence and removal of CECs such as PhACs and EDCs in a drinking water treatment plant (DWTP) treating raw water from the Mediterranean Llobregat River. The DWTP combined conventional treatment steps with the world's largest EDR facility. A hybrid system combining other processes is used in DWTP. EDR technology demonstrated less efficiency in removing contaminants compared to treatments carried out with adsorption and chlorine dioxide oxidation. On the other hand, EDR technology has been determined as an additional barrier to research the main removal of negatively charged compounds but not neutral compounds.

Roman *et al.* (2019) studied the physicochemical characteristics in the transport and adsorption of organic micropollutants in ED and EDR membranes. Micropollutants with positive charge were characterized by the highest adsorption (50–100%), while micropollutants with negative charge were adsorbed in a significantly smaller amount (10–20%).

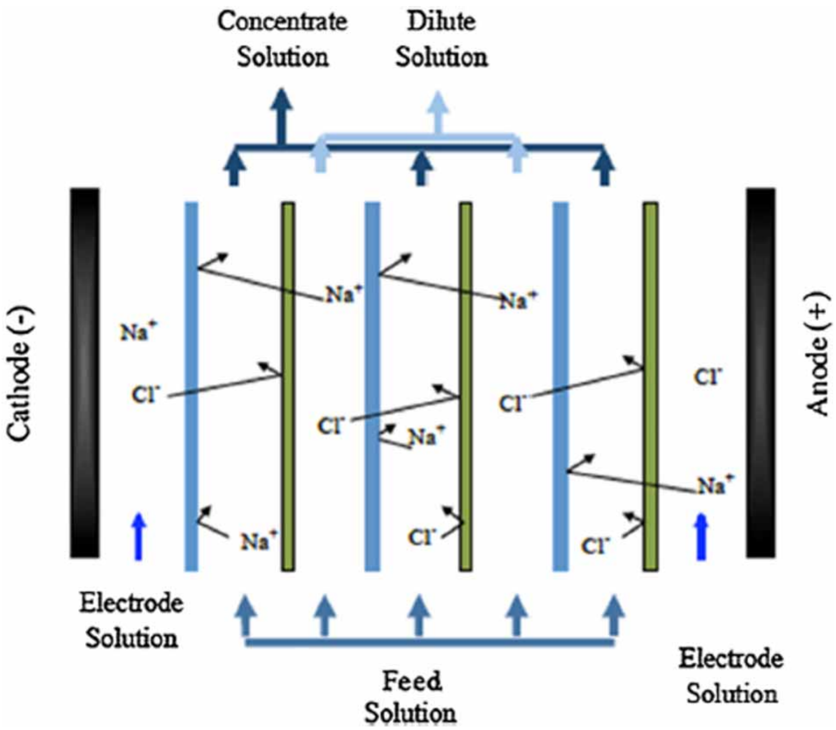


Figure 10 | Schematic diagram of the structure. *Source:* Warsinger *et al.* (2018).

Membrane distillation

The process of using a distillation membrane (MD) consists of the thermal water treatment process. This component is a promising process mainly designed for desalination processes (More & Tyagi 2020). The difference in vapor pressure between the hot feed and the cold permeate is the driving force, which is a major advantage compared to conventional pressure membrane processes (Wang & Chung 2015). The membranes used in the MD process have some requirements: they must use one layer or several layers; at least one of the layers that is in direct contact with the hot current must be hydrophobic; and they must be thin (since the permeation flow is inversely proportional to the thickness of the membrane) (Shirazi & Kargari 2015). The basic principles of the MD process are seen in Figure 11.

Some studies have reported possible applications of the MD processes, ensuring the elimination of different micropollutants from water. Wijekoon *et al.* (2014) examined the feasibility of MD for removing 29 CECs during water and wastewater treatment. All CECs with pK_H (classified as non-volatile) were well removed by MD, while three compounds (i.e. 4-tertoctylphenol, 4-tert-butylphenol and benzophenone) with moderate volatility ($pK_H > 9$) therefore had the lowest rejection efficiencies of 54, 73 and 66%, respectively. When MD treatment was integrated with a thermophilic MBR, near complete removal of all 29 CECs investigated in this study was achieved, suggesting that MD could be a promising post-treatment to be used in conjunction with a thermophilic MBR for CECs removal.

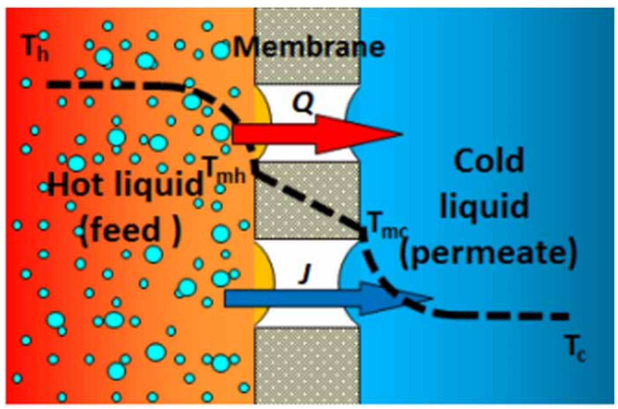


Figure 11 | Basic principles of MD process. *Source:* Ghaffour *et al.* (2013).

CONCLUSION

Developing efficient processes for removing CECs from the environment has become a growing concern. This is due to micro and nano-pollutants, which are emerging through anthropic activities. Conventional water treatment is inefficient for its effective removal. Significant harm to human and animal health can be directly related to the intake of these pollutants.

Membranes stand out among the processes used for the treatment of CECs, mainly due to their high potential for the retention of extremely small particles. Their designs delimit the size of particles to be captured (due to the porosity of the membranes); however, pore size is not the only factor responsible for the MSP.

According to the materials and techniques used for their manufacture, membranes can be classified as MF, UF, NF, and RO. MSP cover a wide range of applications and can be useful as a single treatment or coupled with other treatment methods. They require low energy consumption compared to other existing technologies, resulting in a lower cost per cubic meter treated. Another advantage is that they do not introduce chemical additives into the process.

Several processes and materials have been studied for application in MSP. Composite materials of graphene, GO, and rGO are the target of increasing studies for application in MSP, due to their numerous excellent properties and promising characteristics.

The properties and functionalities of graphene-based nanocomposite membrane separation for CECs removal from water were studied. The use of these graphene-based materials, such GO and rGO, for removing CECs has been a rapidly growing area of interest in the recent literature.

The application of MSP is still a challenge on a large scale, due to factors such as cost, fouling control, difficult dispersion in aqueous solution and low sorption affinity. Moreover, each membrane technology has specific characteristics that must be taken into consideration to choose the best treatment.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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