

Emerging membrane technology and hybrid treatment systems for the removal of micropollutants from wastewater

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HIGHLIGHTS

- Emerging membrane technology for micropollutant removal was narrated.
- Membrane Bioreactor (MBR) technology for hazardous pollutant removal was illustrated.
- Advanced hybrid treatment for micropollutants removal was discussed.
- Advanced integrated technique for micropollutant from wastewater was elaborated.

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ABSTRACT

Currently, substances have been detected that have always been present but have long been unnoticed; among these substances called emerging pollutants are pharmaceuticals, pesticides, cosmetics, cleaning, and personal care products, among others. Membrane Bioreactor (MBR) technology has gained extensive popularity in enhancing the treatment and reutilization of wastewater. It exhibits efficiencies ranging from 40 to 79 % for treating high organic loads such as dairy wastewater while achieving impressive rates of up to 90 % for effectively removing emerging contaminants. However, traditional MBRs with ultrafiltration or porous microfiltration membranes are not intended to remove hazardous micropollutants from water, which are constantly present in trace concentrations and can adversely affect terrestrial and aquatic ecosystems. Several effective techniques have been used to remove hazardous micropollutants from water, especially for hydrophilic and resistant chemicals. These tactics consist mainly of improving the operating environment, using high-retention membranes instead of porous membranes, adding functional materials to the bioreactor, and including additional procedures for effluent treatment. To ensure almost complete removal of micropollutants, the MBR can be complemented with effluent treatment, recently the use of high retention membranes, hybrid MBR-nanofiltration systems, or other processes such as electrocoagulation or advanced oxidation (AOP) have been reported. Nevertheless, additional investigation is required to assess these strategies' technical and economic viability, particularly regarding real-time effectiveness. This evaluation is crucial for identifying the most appropriate techniques for industrial applications.

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1. Introduction

Emerging contaminants that seriously threaten the ecosystem include poly-fluoroalkyl compounds, endocrine disruptors, biocides, and pharmaceutical and personal care goods [1,2]. Long-term exposure to micropollutants over an extended period can have a detrimental impact on both human health & wildlife [3,4]. As a result, micropollutants are frequently found in waters with concentrations ranging from a few nanograms to grams per liter [5–7]. Traditional wastewater treatment facilities (WWTP) are less successful in removing emergent micropollutants than WWTP designed to remove conventional contaminants [8–10]. Therefore, creating more efficient technology to eliminate micropollutants is critical. Alternative activated sludge treatment methods include membrane bioreactor (MBR), which has evolved during the past few decades. To produce better quality effluent, less toxic to the ecosystem and reduce the generation of sludge, it can be run at a greater biomass content & extended sludge retention time (SRT) [11]. To progress wastewater treatment and reuse, MBR can increase micropollutant removal. Conventional MBR has made strides in wastewater treatment. However, several micropollutants, particularly recalcitrant and hydrophilic substances, including carbamazepine, atrazine, etc., cannot be removed [12–18]. Consequently, numerous approaches have been employed to enhance MBR's efficacy in removing micropollutants. Furthermore, to provide high-retention MBRs for improved micropollutant elimination, a number of tight membranes, including nanofiltration (NF) & forward osmosis (FO), can be used in place of porous MF or UF membranes [19,20]. Wang et al. (2023) recently has reported the use of novel macrocyclic polyamines in nanofiltration membranes (1, 4, 7, 10-tetraazacyclododecane- trimesoyl chloride) for the treatment of methylene blue & tetracycline with a removal of 99.9 % and 92.7 % respectively [21], on the other hand, a recent study reported a nanofiltration-based MBR system which achieved good removal of most MP (pharmaceuticals in synthetic domestic wastewater) operating at ultra-low fluxes, resulting in a very low fouling rate, reduction of carbon emissions by 45.2 % over the microporous membrane system due to the decrease in energy consumption by the pumping systems [22], while another research group used coagulation combined with the electro-Fe⁰/H₂O₂ reaction to treat MBR effluent in landfill leachate, their results shown a removal percentage of COD (74.18 %) and chromaticity (72.22 %), with an operating cost of only 4.18 \$/m³ [23]. However, a systematic comparison of the applicability of various approaches to enhance MBR efficiency for removing micropollutants has not been conducted. The utilization of ultrafiltration (UF) and microfiltration (MF) technologies has gained significant popularity in the treatment of water bodies due to their compact design, ease of automation control, and ability to produce high-quality effluent. [8,24]. However, the pore size limitation results in poor micropollutant rejection for both methods. Furthermore, it should be noted that micropollutants have the potential to accumulate within the pores or on the surface of ultrafiltration and microfiltration membranes, leading to fouling and reduced permeability as time progresses. [25].

To establish which important tactics are preferred in practice, this study reviews essential approaches for improving the performance of MBR for the removal of micropollutants critically. Micropollutant transit and fate in MBRs were clarified. The key factors influencing MBR's performance in removing micropollutants have been identified to facilitate process optimization. Furthermore, the effectiveness of innovative MBR technologies in eliminating micropollutants has been emphasized. Suggestions for future improvements in MBR systems have been provided to enhance micropollutant removal and promote water reuse.

2. Classification of micropollutants (MPs)

Micropollutants, primarily pharmaceuticals, industrial chemicals, personal care items, pesticides, metallic trace elements, and polycyclic

aromatic hydrocarbons, have frequently been found in a variety of waste streams at concentrations ranging from a few ng to mg/L [7,14,26–30]. Municipal wastewater collects a variety of waste streams, such as those from residential, commercial, & industrial areas [31–33]. The increased usage of synthetic products, such as pesticides, cosmetics, and medications, has led to a significant rise in micropollutants in municipal wastewater. It has been estimated that over 100 different micropollutants have been detected and reported in such wastewater [34–39]. For instance, two wastewater treatment facilities in Southern California have found four antibiotics, including trimethoprim, triclosan, erythromycin, and sulfamethoxazole, with concentrations ranging from 20 mg/L to 2.15 g/L [40]. Currently, no specific regulatory standards exist for the allowable concentration of micropollutants. However, it is essential to note that industrial wastewater must undergo appropriate treatment to comply with the required quality standards before being discharged into municipal wastewater pipelines. So industrial wastewater is a significant source of micropollutants for water bodies or municipal wastewater, especially from the chemical and pharmaceutical industry [41]. An example of this is hospital wastewater, recognized as a significant contributor to the presence of antibiotics in the environment. In a study conducted by Mazzitelli et al. 2018, the maximum observed concentration of paracetamol in treated psychiatric hospital wastewater reached up to 37 g/L. This demonstrates the substantial levels of pharmaceutical substances found in specific wastewater sources. [42]. According to Aydin et al. (2019), the total amount of antibiotics in 16 separate hospital wastewater effluents ranged from 497 ng/L to 323 g/L in the winter to 21 ng/L to 5 µg/L in the summer [43]. Some nations, like the European Union and the United States, have designated various micropollutants (such as bisphenol A and diuron) as significant indices to assess water quality due to their ecotoxicological impacts and associated health hazards.

3. Removal of pathogenic bacteria by advanced membrane technology

The eradication of pathogens has released global attention, and a solution is attained by developing new sophisticated membrane technologies. Membrane technology combines filtration technology which is enhanced by the membrane's chemical & physical characteristics [44]. This section briefly discusses the major membrane technology that rejects/removes pathogens: nanocomposite membrane, membrane bioreactor, photo-catalytic membrane reactor, thermal-driven membrane technology, and pressure-driven membrane technology. Advanced membranes containing nanoparticles distributed in their polymer matrices are nanocomposite membranes. They are utilized for liquid-liquid, liquid-solid, gas-gas separations, proton exchange membrane fuel cells (PEMFCs), sensor applications, and methanol fuel cells. Nanocomposite membranes can be categorized into four groups based on membrane structure and the location of the nanomaterial: (1) surface-located nanocomposite; (2) conventional nanocomposite (NC), and (3) thin-film composite (TFC) or thin-film nanocomposite (TFN) with nanocomposite substrate. (See Figs. 1 and 2.)

3.1. Conventional membrane

The four nanofillers used in NC membranes are organic, inorganic, hybrid materials, and biomaterial. Phase inversion fabricates nanocomposite membranes that can be manufactured as hollow fibers or flat sheets. Nanofillers are disseminated in the polymer solution and are primarily utilized in microfiltration (MF) & ultrafiltration (UF) processes. Since the filler particles have a large specific area & high surface-to-volume ratio, the polymer chains and the filler are bound together. The remarkable properties of nanoparticles are incorporated into the composite by creating a solid interfacial connection. Robust membrane engagement makes effective stress transmission across the contact possible [45].

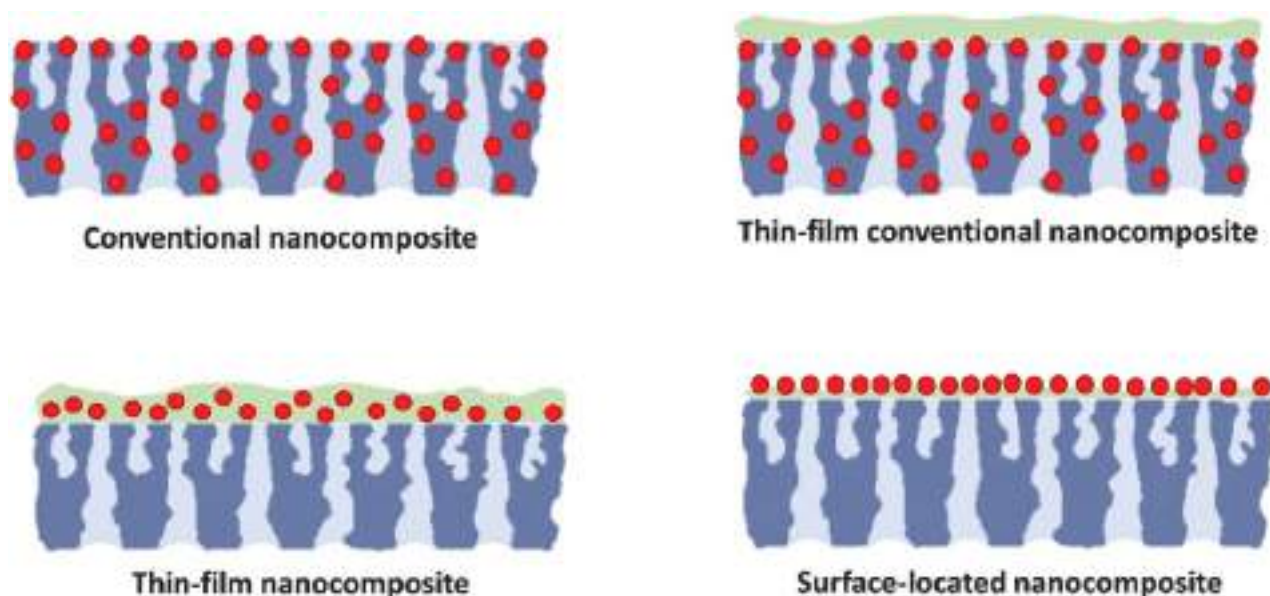


Fig. 1. Different types of nanocomposite membranes based on the structure.

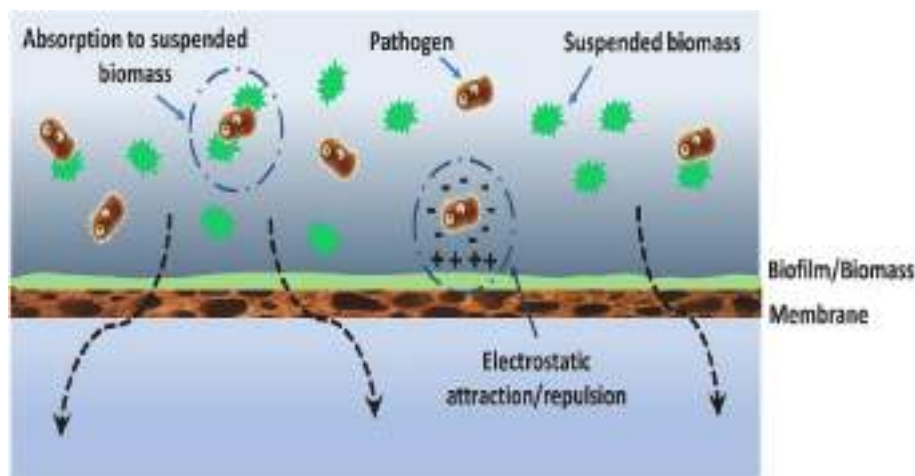


Fig. 2. Mechanism representing the working of membrane bioreactor.

Because the filler's nanoparticles have a large specific area, they probably have a high surface-to-volume ratio, which causes the polymer chain and the filler to be extensively bound [46]. The remarkable properties of nanoparticles must then be transferred into the composite by creating a solid interfacial connection. Consequently, robust engagement makes effective stress transmission across the contact possible. Some of the nanofillers used are TiO_2 [47], SiO_2 , Al_2O_3 , Fe_3O_4 , ZnO [48], CNTs [49], Cyclodextrin [50], Polypyrrole [51], Aquaporin [52], $\text{TiO}_2/\text{MWNTs}$, Au/xGnPs [53], Fe/Mn [54], and SiO_2/GO [55]. These studies reported that adding nanomaterial to polymers can modify physicochemical properties and membrane structure (charge density, porosity, hydrophobicity, thermal, mechanical, and chemical stability). In addition, as well add special functionalities like antibacterial and photo-catalytic properties. Exfoliated graphite nano-platelets and AuNPs (gold nanoparticles) were incorporated into the membrane [53] to create a multifunctional membrane. When NaBH_4 was used to reduce 4-nitrophenol to 4-aminophenol, their membranes demonstrated improved compaction resistance, permeability, and catalytic properties. AuNPs acted as the catalyst in this reaction. Furthermore, adjusting the nanofiller's relative contents regulates the structure and catalytic activity. Such as silver (Ag) [56], zinc oxide (ZnO) [57], nickel (Ni) [58],

copper (Cu) [59], selenium (Se) [60], TiO_2 [61], and silica (Si) [62] possess antibacterial and antimicrobial characteristics. Arjan et al., 2022 fabricated a PLA/CMC/GO-f-COOH@Ag membrane that showed strong π - π interactions that were efficiently used in photo-catalytic behavior for degrading dyes in water bodies [63].

3.2. Thin-film composite (TFC) membrane

In thin film composite (TFC) membranes, a thin barrier layer is applied onto a porous supporting layer. These membranes are commonly employed in the desalination of brackish water and seawater, as well as in the removal of hardness and various organic micropollutants, including pesticides, disinfection byproducts (DBPs), endocrine disrupting compounds (EDCs), and pharmaceutically active compounds. [64]. Forward & pressure-retarded osmosis (PRO) procedures have recently accelerated the development of TFC membranes because they provide the possibility for energy savings during water treatment operations. TFN membranes have also been created using nanomaterials such as CNTs (carbon nanotubes), silica, zeolites, Ag, and TiO_2 , previously used to construct conventional nanocomposite membranes [65]. The in-situ IP procedure involves an organic solvent such as

trimethyl chloride (TMC) and aqueous *m*-phenylenediamine (MPD). Wang et al. (2023) designed a type of TFC on a structural-controlled substrate using metal oxide frameworks (ZIF 8). A polyethersulfone substrate was fabricated by incorporating ZIF-8 with various morphology, namely, hollow tube, spherical and flowerlike structure. The membrane exhibited excellent rejection of sodium chloride, which can be helpful in desalination [66]. Qi et al. (2023) incorporated covalent organic frameworks (COFs) nanomaterial to form a thin-film nanocomposites (TFN) RO membrane with excellent rejection ability of NaCl of about 93 %, anti-fouling resistance, excellent chlorine resistance, and acid/alkali resistance [67]. Kallam et al. 2022, used polydopamine and titanium oxide nanoparticles into polyethersulfone (PES) via the non-solvent induced phase separation (NIPS) process, which showed 0.28 g/L of specific salt flux when compared to the new membrane substrate. These membranes were also used for water reclamation from oil wastewater [68]. Behdarvand et al., 2021 synthesized PVA/PES TFN membranes and studied the effect of carbon nano-materials infiltration. Graphene oxide nanoparticles incorporated membranes showed 35 % enhancement in water permeability and rejection of Na₂SO₄ [69].

3.3. Surface-located nanocomposite

Surface-located nanocomposite membranes are divided based on the assembly of membranes, such as self-assembly, coating, electrostatic attraction, absorption/reduction, chemical grafting, layer-by-layer assembly, and cross-linked process. These membranes showed self-cleaning ability, antimicrobial activity, and excellent rejection ability based on the functional group of the surface of the membrane [70]. Polyimide blended PES, Sulfonated PES, PAA modified polypropylene (PP), and poly(styrene-*alt*-maleic anhydride) (SMA) blended PVDF is commonly used for membranes preparation of self-assembly technique. These membranes are coupled with UV light irradiation for the better rejection ability of the analyte [71]. The coating technique is a standard method where the nanocomposites are coated on the surface of the membranes. These show excellent rejection rates for organic dyes. The major disadvantage of coating is the loss of the composite over time bound with weak van der Waals forces. In the electrostatic attraction technique, the positively charged ions are encapsulated in the negatively charged membrane [72]. A few positively charged ions are AgNPs, modified silica NPs, CuNPs, etc. These membranes show evident antimicrobial activity and enhanced anti-fouling activity. The major disadvantage is the dis-attachment of ions during the cross-flow process [73]. This occurs when the feed has high ionic strength, high pH, and high concentrations of organic ligands. The surface-loaded nanocomposite shows high anti-fouling efficiency with the two effective microbes in water (*E.coli* and *Pseudomonas* sp) [74].

3.4. Membrane bioreactor

Membrane bioreactors (MBRs) are a WWT technology that integrates membrane filtration and activated sludge treatment (biological wastewater treatment processes). In activated sludge, a consortium of microbes is in suspension form, which is utilized to degrade the micropollutants under oxygen conditions, generating biological floc [75]. With membrane filtration, bacteria are entirely contained inside the bioreactor, giving researchers more control over biological processes and the ability to alter the environment of microorganisms in the aerated tank. MBRs can be explained in three mechanisms: The first strategy is that enteric pathogens get absorbed into the biomass. Biomass is a mixture of suspended solids and bacteria larger than the membrane pore size. Hirani et al. (2010) reported that native MS-2 coliphage eliminated added particles compared to seeded MS-2 coliphage in numerous AeMBR systems [76]. The second strategy relies on membrane adsorption and rejection. The pathogen binding to the membrane pores obstructs them; the biofilm that has adhered to the surface membrane helps with rejection. Since the adequate pore size is

smaller and fewer pores can be accessed, virus particles have more difficulty penetrating the membrane. Degradation and inactivation in the mixed liquid phase is the third strategy. The main mechanisms are responsible for the impact of mixed liquor on viral destruction, and inactivation is assumed to be predation by another enzymatic and bacterial breakdown. MBR process studies for pathogen removal with the Log removal viable factor are Norovirus GI (1.82) [77], T4 Phage (5–7) [78], MS-2 Phage (1.7) [79], Enterovirus (0.3–3.2) [80], Somatic coliphage (3.24) [81], Adenovirus (4.10–6.30) [82], and Sapovirus (3) [83]. Based on the studies, membrane characteristics and composition have significantly removed the pathogen. Nevertheless, this system has significant downsides, including membrane fouling issues, high operational and investment costs, and membrane lifespan replacement. Using a longer hydraulic retention time, high inoculation sludge concentration, and an anti-foulant layer was suggested to achieve efficient future pathogen eradication by the MBR system.

3.5. Photo-catalytic membrane reactor

A photocatalytic membrane reactor (PMR) combines photocatalysis by separating the membrane in a single step [84]. Photocatalysis and membrane separation are the fundamental mechanisms for pathogen-removal processes in PMR. In the photocatalysis process, the hydroxyl radicals ($\cdot\text{OH}$) tend to oxidize the organic contaminants in the water [85]. The use of platinum-loaded TiO₂ for disinfection was initially reported in 1985 and involved the catalytic removal of microbial pathogens such as *Saccharomyces cerevisiae*, *E. coli*, and *L. acidophilus*. The common photo-catalysts implemented are TiO₂, WO₃, ZnO, BaTiO₃, Fe₂O₃, BiOBr, and CuS [86]. The use of photo-catalyst has been well developed, as evidenced by the use of photo-catalyst in different processes, including wastewater treatment & the removal of bacteria and viruses such as SARS-CoV-1 and influenza H1N1 [87]. Bacteriophage MS-2 [88], Coliphage, *E. coli* K12 PHL849 [89], *E. coli* K12 PHL1273, Bacteriophage T4 [90], *Lactobacillus casei* Phage PL-1, and Bacteriophage kNM1149 [91] are some pathogen removal using photo-catalytic membrane technique with the removal efficiency of 98–99.9 %. In conclusion, PMR technology can potentially remove pathogens from wastewater. PMRs also provided a superior alternative to suspended photocatalysis since they combined photocatalysis and membrane synthesis into a single process [92]. However, the PMRs have a few disadvantages. It damages the polymeric membrane caused by prolonged exposure to reactive oxygen species and UV light, as well as the possibility of an accumulation of photo-catalysts on the membrane surface that decreases the active catalytic area for the sanitization of pathogens.

3.6. Pressure-driven membrane technology

Microporous membranes (MPM) are used in pressure-driven membrane processes to hold suspended colloids and particles roughly between 0.1 and 20 μm in size [93]. Physical separation is the working principle of the MP process. Due to the lack of potentially toxic chemicals like ozone and chlorine used in the ozonation and chlorination processes for pathogen removal, the low-pressure-driven membrane process offers a better option for disinfection [94]. The MF method operates at low pressures of 0.5 to 5 bar (up to 3.4 logs), producing high solvent flux (65–85 LMH/bar) and equivalent somatic *E. coli* removal. A microfiltration membrane was fabricated by Wang et al. using a microscale polyethylene terephthalate (PET)/two-layer nanoscale polyacrylonitrile (PAN) fibrous scaffold composed of cellulose nanofibers with a membrane diameter of 5 nm. These membranes can eliminate *E. coli* by pore exclusion while maintaining a high penetration rate of 0.19 L/m²h/Pa and a removal log 4 for obliterating the MS2 virus. The construction of these membranes is based on fabricating a web-like structure for the adsorption of polluted molecules with a large surface area and high charge density per unit volume. Wang et al. studied the hybrid MF/UV-C membrane for rejecting *Candida albicans*, *E. faecalis*,

and *E. coli*. The hybrid membrane demonstrated synergy and a more effective intensification process when it operated at low pressure of 0.5 bar and 8 s [95]. In conclusion, the traditional MP technique eliminates more prominent pathogens like bacteria or protozoa. However, it is ineffective in removing viruses because of the increased size of membrane holes.

3.7. Thermal-driven membrane technology

Membrane distillation (MD) is a technique that works on thermal-driven membrane technology. This method uses a temperature difference/vapor pressure over a porous hydrophobic membrane as the vital force to move water vapor from a polluted feed to a distillate stream [96]. The trans-membrane temperature difference causes the development of a vapor pressure differential that permits water vapor to pass through the influent stream to the distillate side while preventing non-volatile species like salt from doing so. [97]. In MD, the pathogen in the feed is primarily disinfected by the high operating temperature, which also serves as a secondary barrier. Hardikar et al. (2021) reported the impact of influent temperature on microbe elimination by bench-scale DCMMD (MS2 and PhiX174 bacteriophage). When the temperature rose, both viruses showed a decline in activity at 45 °C; after the initial reduction in the first hour, both virus concentrations fell by around 2 log in the next 3 h. Viable MS2 concentrations dropped by over 4 logs at 55 °C, although PhiX174 concentrations dropped by >2 logs. Infectious viral levels in the influent were dramatically reduced by <6 logs after 4 h with optimized temperatures (> 65 °C), according to temperature sensitivity studies conducted on the surrogate viruses used in the study [98]. PVDF nanofiber membranes with embedded SiO₂ NPs impregnated with a composite of AgNPs. The carboxylated multiwalled carbon nanotubes (f-MWCNTs) showed exceptional antimicrobial characteristics against bacteria by a 47 % flux reduction in anti-fouling and high salt rejection (99.8 %) rates. The MD process's thermal conditions allow it to eliminate pathogens and viruses effectively. Nanoparticles with specific biocidal activity, such as Ag NPs, CNTs, and GO, can reject thermophilic pathogens. However, using high operating temperatures might raise the cost of operation [99]. And therefore, pathogens in wastewater may be removed using the MD process by using alternative heat sources like geothermal, solar, or even industrial waste.

4. Pretreatment techniques for membrane processes

The main motive of pretreatment is to make the feed water compatible with the membrane. Pretreatment is necessary to reduce membrane fouling, scaling, and degradation and extend the effectiveness and life of membrane components. To restore the membrane flow rate and salt rejection efficiency, regular backwashing/cleaning is frequently required. Membranes without pretreatment techniques can have increased system downtime, more expensive (chemical), irreversible performance loss, and membrane deterioration [100]. The first technique is pressurization, a simple method. Pressurization at 25 bar provides wetting of membranes and does not result in the unintended compaction of composite membranes. Deionized water can be pumped through the membrane under pressure for 15 min with deionized water pressure of 25 bar [101]. The second technique uses a highly alkaline P3-Ultrasil® 10 cleaner or chemicals.

The maximum flux recovery can be achieved using chemical cleaning. Nevertheless, this method usually necessitates an upstream filtering unit, damages the membrane, and produces secondary contaminants [102]. Ultrasound/ozone is a potential physical cleaning technique; however, repeated usage can harm the membrane surface. The use of a saturated CO₂ solution during backwashing is another cleaning method. Electro-coagulation is an alternate pre-treatment technique [103] [104]. Nonetheless, coagulation continues to be the most popular and economic pre-treatment strategy. Combining several pre-treatment techniques maximizes each method's advantages, or integrating techniques

can prevent membrane fouling and preserve membrane performance.

Other techniques can be coupled with MBR to enhance the removal of micropollutants. To fulfill the discharge wastewater regulations and create high-quality water for recycling, effluent purification is explicitly required for MBR and other techniques. High-retention membranes (such as NF, FO, MD, and RO), electrochemical processes AOPs, and activated carbon filtration are a few of the frequently utilized approaches. In an electrochemical MBR, electrokinetic events, including electrophoresis, electrocoagulation, and electroosmosis, can increase sludge floc size and lower the zeta potential to enhance the adsorption and biodegradation of micropollutants. Therefore, electrochemical MBR can be the best-coupled technique for removing microcontaminants [105]. The characteristics of electrocatalytic/electrochemical membrane materials have a significant role in micropollutant removal and electrochemical oxidation that is responsible for the removal efficiency. Currently, the most common types of electrode membranes are micro-filtration membranes made of carbon-based, magnéli-phase, porous-Ti, polymer composite materials, and electrochemical ceramics. Even though several research articles have been published in this area recently, electrocatalytic membranes' ability to remove micropollutants is still in its infancy. The significant parameters considered for the design of an electrochemical membrane to improve the removal efficiency are current density, operating voltage, temperature, pH, and membrane flux.

In general, as the working voltage or current density rises, correspondingly raises the removal effectiveness of micropollutants. Additionally, a higher operating voltage or current density leads to a greater energy need, increasing the cost of operation. The removal efficiency and operation cost must thus be carefully balanced when determining the ideal operating voltage or current density. The solution conductivity, electrolyte content, and composition significantly influence the functionality and stability of membranes. Typically, as the electrolyte concentration rises, micropollutants' removal efficiencies increase [106]. Under acidic circumstances, micropollutant removal effectiveness is higher than in alkaline conditions. At the surface of the membrane, numerous H⁺ ions interact with O₂ to produce H₂O₂ in an acidic environment, which is favorable for the production of OH⁻ ions [107]. Increasing temperature over a range is beneficial for micropollutant removal. For instance, multi-walled carbon nanotube filters had removal efficiencies of sulfamethoxazole (SMZ) of 77 %, 90 %, and 96 % at 15 °C, 25 °C, and 35 °C, respectively. This is primarily because elevating the ambient temperature leads to a decrease in solution viscosity, which speeds up the mass transfer of contaminants to the membrane surface [108].

5. Structure approaches towards membranes for micropollutant (MPs) removal

Membranes classification is reported in several works and is classified according to the state of the material, structure, chemical composition, morphology, superficial charge, etc. [109,110] being the most common, however; we can also find them classified according to gradient structures, surface patterning, permeability, selectivity and stability [111]. The different configurations will be discussed below (Fig. 3).

5.1. Liquid membranes

A liquid membrane (LM) system requires a fluid or quasi-fluid phase, which will separate two other immiscible phases from the LM [112]. This membrane type has been used to recover or eliminate contaminants, like metals, micropollutants (organic or inorganic compounds), and other biomedical and analytical chemistry applications. The transport in a liquid membrane involves liquid-liquid extraction processes following separation utilizing membranes using a serial device. It is usual to use supports for the liquid membrane, and such support can be

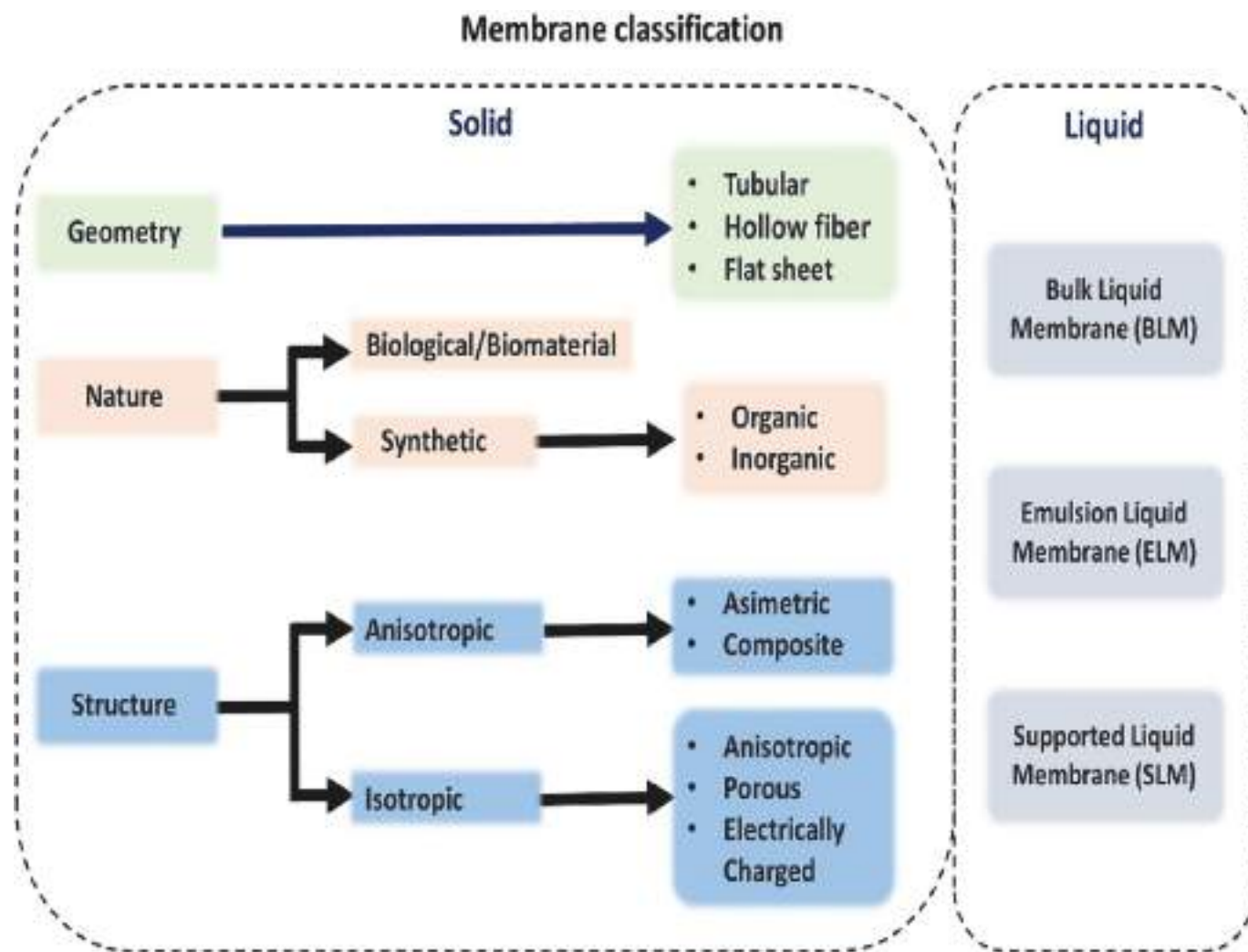


Fig. 3. Membrane classification and their distinct characteristics.

microporous of inorganic type or formed by polymers. This type of membrane is known as a supported liquid membrane (SLM) [113]; on the other hand, there are the bulk liquid membranes (BLM), which are liquid membranes immiscible in water that separate the feeding and receiving phases [114]. In contrast, liquid emulsion membrane (ELM), Regardless of the type of emulsion, the system has the function of allowing the transfer of specific components (one or more), so it can be helpful in the removal of contaminants such as hydrocarbons or pharmaceutical residues or recovery of metals of potential interest [115]. Although the membranes mentioned above belong to the general classification, variants have been developed in line with scientific and technological progress, including hybrid liquid membranes (HLM), hollow-fiber-contained liquid membranes (HFCLM), flowing liquid membranes (FLM), and hollow-fiber liquid membrane (HFLM).

One of the applications of liquid membranes in wastewater treatment is due to some contaminants' organic and hydrophilic character. In this case, bioreactors with BLM have been used [116]. Examples are the work reported by Chang S.H. for the removal of some metals like nickel, copper, silver, cadmium, mercury, lead, and cerium [117]. On the other hand, Khalaf, Z. A. & Hassan, A. A. studied different parameters (stripping and feed rate, stirring velocity, initial feed concentration, carrier concentration, temperature, pH, etc.) for removing cadmium using BLM [118]. Regarding using a supported liquid membrane with strip dispersion (SLM-SD), 99.67 % of removals have been reported for the antihypertensive drug amlodipine [119]. In comparison, green emulsion

liquid membrane has shown promising results with a removal efficiency of 91.27 % for a pharmaceutical micro-pollutant called norfloxacin [116,120].

5.2. Solid membranes

5.2.1. Based on nature

Membranes of synthetic nature can be made of polymers, mainly organic or inorganic materials; for polymeric membranes, the most common materials used are polypropylene (PP), polytetrafluorethylene (PTFE), polycarbonate (PC), polyvinylidene fluoride (PVDF), polyamide (PA), polyethylene (PE), polysulfone (PSU), polyethersulfone (PES), and cellulose acetate among others [121], while inorganic include ceramics (Alumina, titania, zirconia, glass, and silicon carbide), metals, zeolites, carbon, silica and mixture of these materials [122]. At the same time, biomembranes are manufactured from biomaterials like nano cellulose, chitosan, residual agricultural and industrial waste, silk, and other natural fiber [12]. Other examples of these materials are composites, made with the finality of improving or reinforcing some particular properties, such as the case reported by Perendija et al., with the modification of cellulose with diethylenetriamine and (3-Glycidyloxypentyl)trimethoxysilane as well as lignin with epichlorohydrin and tannic acid for the removal of Cr(VI), Ni²⁺, Pb²⁺ and Ni²⁺ [123].

5.2.2. Based on the structure

This classification has two groups, symmetric or isotropic membranes and asymmetric or anisotropic membranes. The isotropic membranes can be subdivided into a) Isotropic porous membranes, a material with high porosity, where the pores are physically random and interconnected. The separation of target compounds is performed by size difference, mainly by ultra or microfiltration. Like the research presented by Luo et al., with the usage of a mesoporous catalytic ceramic membrane of $\text{AlOx/La}_2\text{CoMnO}_{6.8}$ for the removal of carbamazepine,

atrazine, and sulfamethazine [124]; b) Dense isotropic membranes (non-porous), it is not porous as the previous case, it is a dense material which requires a driving force with a gradient either of pressure, electric potential or concentration to carry out the separation of components, it is usual to use them in reverse osmosis. For example, polyvinylidene fluoride (PVDF) membranes have been used for the treatment of oily wastewater from the petrochemical industry [125]; and c) Electrically charged membranes: They can be either porous or dense, they are electrically charged (positive or negative charges), and according to this

Table 1

Illustrates the different types of membranes used for water filtration with their removal efficiencies.

Type of membrane	Material	Pollutant	Operation conditions	Removal efficiency	Reference
Emulsion liquid membrane	Oil phase: span 80 (surfactant), D2EHPA (extractant) magnetic Fe_2O_3 particles (co-surfactant) Stripping phase: $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$	Pb^{2+}	Span 80 at 2 % v/v; carrier at 4 % v/v, and Fe_2O_3 at 0.3 % w/v, with a pH = 3, 1:1 internal: organic phase, agitating speed of 250 rpm, emulsification time of 10 min and contact time of 8 min, with 0.3 % emulsion breakage	97.2 %	[128]
Emulsion liquid membrane (ionic liquid)	Organic membrane phase (Span-20/[TMAm][Cl]/sunflower oil) Water-oil emulsions (W/O): NaOH/membrane phase	Salicylic acid	Stripping agent: NaOH 0.005 M as emulsifier: Span-20 at 0.2 wt% [TMAm][Cl], agitation speed of 4000 rpm, 3:1 treat ratio with 25 min extraction time	90.04 %	[129]
Synthetic membrane (Polymer/photocatalyst)	Poly(vinylidene fluoride) membrane (PVDF- TiO_2)	Steroid hormone 17 β -estradiol	60 $\text{L}/\text{m}^2\text{h}$ flux, 25 mW/cm^2 , and Ti 6.5 %	> 96 %	[130]
Synthetic membrane (catalytic ceramic/N-doped carbon)	NC@CM/PMS CM: TiO_2 and ZrO_2 NC: N-doped carbon PMS: Peroxymonosulfate	Bisphenol A	0.01 g L^{-1} of PMS, 2 mg L^{-1} of BPA, pH = 7 using a phosphate buffer 0.05–0.2 MPa	81–100 %	[131]
Synthetic Cyclen-TMC LNF	Polyamide polymer	Methyl blue (MB) and tetracycline (TC)	pH = 9 MB = 50 ppm TC = 200 ppb 17.7 cm^2 filtration area of 4.5 L/min cross-flow velocity	99.9 % (MB) and 92.7 % (TC)	[132]
Biomembrane	Bacterial cellulose (BC) biopolymers	Polystyrene microplastics (PS-MPs) at 10 mg/L	20 filtration cycles of 25 mL of MPs	93–99 %	[133]
Isotropic porous	Commercial Polyamide (ESPA1–2521) membrane modified with grafting poly (glycidyl methacrylate)	EDC, PhAC— (acetaminophen and Bisphenol-A and carbamazepine)	Pressure 10–20 bar Feed flow rates range: 600–680 L/h and 330–400 L/h; 100–120 mg/L for Bisphenol-A, 10–12 mg/L for carbamazepine, and 100–120 mg/L for acetaminophen	Solute passage % Bisphenol-A 0.09 \pm 0.007 Carbamazepine n.d. Acetaminophen 0.41 \pm 0.03	[134]
Electrically charged	PC-SA and PC-SK membranes (PCA GmbH, Germany)	Salinity	PCCell ED 64-004 cell of PC-SA (7 pieces) and PC-SK (8 pieces), Pt/Ir-coated titanium electrode, 5693 mg/L total solid dissolved, 8 V, 8 L, 40 min	Water recovery 50 % 322.1 mg/L TDS	[135]
Asymmetric	Poly(lactic acid) (PLA)	Bovine serum albumina (BSA)	BSA (1 g/L) of 10 mM phosphate buffer solution pH = 7.2 Membrane area = 12.56 cm^2 20 psi for 60 min	92 %	[136]
Asymmetric composite	Polyamide membranes for reverse osmosis (self-assembled aromatic nanoparticles, SANs)	Organic micropollutants (OMPs)	25.5 cm^2 effective area NaCl at 2.0 g L^{-1} or 0.2 mg L^{-1} 25 °C and 1.55 MPa	NaCl rejection (>98.3 %) OMPs rejection (>99.4 %)	[137]
Tubular membrane	$\text{TiO}_2/\text{Al}_2\text{O}_3$ composite tubular membrane	Emerging contaminants di-(2-ethylhexyl) phthalate (DEHP), di-n-butyl phthalate (DnBP), caffeine (CAF), acetaminophen (ACE), sulfamethoxazole (SMX) and cefalexin (CLX).	Adsorption 0.1 g/L graphene (maximum time 12 h) Al or Fe anode, SS 216 cathode, 294 kPa transmembrane Pressure and 30 V/cm electric field	89 \pm 2 % DnBP; 85 \pm 3 % DEHP; 99 \pm 2 % ACE; 94 \pm 3 % CAF; 100 \pm 0 %; CLX, and 98 \pm 2 % SMX	[138]
Hollow fiber Nanofiltration (NF)	Modified polyethersulfone with a high chlorine tolerance	Micropollutants (MPs) Atenolol Atrazine Sulfamethoxazole Naproxen	Temperature 20 \pm 0.5 °C 35 mg/L of each MPs Feed solution of 20 \pm 2 $\text{L}/\text{m}^2\text{h}$ Time: 72 h	MP removal (>85 %)	[139]
Flat sheet Ultrafiltration (UF)	Polycarbonate/acrylonitrile-butadiene-styrene flat sheet membranes	Humic acid	Surface area of 14.7 cm^2 3 L influent tank 0.2 bar Airflow of 4 L/min	98.6 % for the blend membrane	[140]

charge, they are either an anion or a cation exchange they are used in electrodialysis. While anisotropic membranes include: a) asymmetric membranes, which generally are made up of a very dense upper layer supported by another porous layer (sublayer) thicker than the upper one, but also can be a multilayer. The composition layers could be different. Like the polyelectrolyte multilayer membranes poly (allylamine hydrochloride)/poly(styrene sulfonate) PAH/PSS bottom layer and a thin poly(acrylic acid)/poly(allylamine hydrochloride) PAA/PAH top layer for the removal of a mix of micropollutants [126]. Finally, b) anisotropic composite membranes are made up of layers of different materials, the main one being the top layer, which will significantly define the separation capacity of the overall system; it also falls into this category as a membrane modified with a particular material which can be micro or nanoparticulate, e.g., asymmetric composite PTFE membranes with the incorporation of different wt% of kaolin for application in textile wastewater treatment [127].

5.2.3. Based on the geometry

Tubular membranes are porous with tube-shaped walls (internal diameter ≥ 2 mm). This type of configuration is carried out by tangential cross flow; they are usually used for processes with feed flows with a high concentration of suspended and dissolved solids, oils, and fats. It favors turbulence and thus better avoids fouling and scale formation. On the other hand, hollow fiber membranes comprise many long, high porosity filaments (internal diameter < 1 mm) encapsulating inside a polymeric body, such as PVC. They can be used for microfiltration or reverse osmosis; unlike tubular ones, they only handle laminar flow. While Flat sheet membranes are placed on plates, are supported on the permeate side and have spacers, and are thus intercalated to form a module. The membrane surface per unit volume varies between 300 and 500 m^2/m^3 ; they are usually used for ultrafiltration, but they have the disadvantage of being unable to be cleaned by backwashing [109]. Table 1 illustrates the different types of membranes and their use and efficiencies.

When contaminated water presents a complex combination of pollutants, it is challenging to treat adequately with conventional wastewater treatment alone, as some MPs may persist. Several studies have shown that advanced treatments like oxidation processes (AOP) and membrane filtration can increase MPs' removal rate [141–144]. Membrane filtration processes include nanofiltration (NF), microfiltration (MF), ultrafiltration (UF), membrane distillation (MD), forward Osmosis (FO), and reverse osmosis (RO), which can be used in stand-alone or hybrid form in the configuration in which the best performance has been obtained.

6. Removal of micropollutants using a hybrid technique

When contaminated water presents a complex combination of pollutants, it is challenging to treat adequately with conventional wastewater treatment alone, as some MPs may persist. Several studies have shown that advanced treatments like oxidation processes (AOP) and membrane filtration can increase MPs' removal rate [141–144]. Membrane filtration processes include reverse osmosis (RO), ultrafiltration (UF), microfiltration (MF), forward Osmosis (FO), nanofiltration (NF), and membrane distillation (MD) which can be used in stand-alone or hybrid form in the configuration in which the best performance has been obtained.

6.1. Micropollutants (MPs) removal using hybrid reverse osmosis and nanofiltration (NF)

Reverse osmosis (RO) is a physicochemical treatment that mimics nature by removing contaminants from water using selective semi-permeable membranes. Unlike osmosis, which is a natural process, it requires an external pressure that can overcome the osmotic pressure. This treatment removes suspended solids (SS), mono and divalent ions,

inorganic and organic compounds, microorganisms, and some pathogens. RO can remove SS and monovalent ions that nanofiltration cannot [145]. The most commonly used types of membranes are a) cellulose triacetate and its derivatives and b) thin-film composite membranes. In this case, cellulose triacetate-based membranes have the advantage of being resistant to chlorination and easy to manufacture and recycle; Their disadvantages are low flow that can be handled, poor mechanical stability, and prone to fouling. While thin film composite membranes are less prone to fouling and scaling, can remove $\cong 98\%$ of contaminants, have higher flow resistance, reduced salt rejection, and better mechanical properties, they are also more corrosion-resistant [146].

The process is affected by different parameters, such as flux, pressure, fouling, solute concentration, pH, time, diffusivity, temperature, polarization concentration, and solute partition coefficient, so selecting the appropriate operating parameters is essential to obtain optimum performance. In this sense, it has been proposed to use the diffusion model (in dissolution); applying the Maxwell Stephan mathematical formula makes it possible to calculate the mass transfer steps through a reverse osmosis membrane. The diffusion model is used at the research and application level, and it contemplates the following processes: a) reverse osmosis, b) gas separation, and c) pervaporation [147]. Fig. 4 shows a typical diagram of the hybrid NF and RO system for removal of micropollutant from water.

A practical application of the Model was presented by Maddah & Almugahwi [148]; reverse osmosis permeation can be written as follows:

$$J_i = A(\Delta p - \Delta \pi) \quad (1)$$

$$A = \frac{P_i c_{i0} v_i}{RTl} \quad (2)$$

where J_i - membrane flux of component i (gfd), Δp - applied pressure drop across the membrane (psi), $\Delta \pi$ - osmotic pressure drop across the membrane (psi), A - water permeability constant ($\text{cm}/\text{atm}\cdot\text{s}$), P_i - component permeability i (cm^2/s), c_{i0} - initial mole concentration (ppm), v_i - molar water volume (cm^3/mol), T - temperature (K), R - gas constant ($\text{m}^3\text{atm}/\text{mol}\cdot\text{K}$) and l - membrane thickness which is assumed to be similar to spacer thickness (mil).

Membrane resistance constant for each Brackish Water Reverse Osmosis (BWRO) Toray membrane can be calculated from:

$$J_i = \frac{p}{kR_m} \quad (3)$$

k - dynamic viscosity of water ($\text{lb}\cdot\text{s}/\text{ft}^2$) & R_m - membrane resistance (ft^{-1}).

The osmotic pressure in a dissolution (aqueous) system can be determined using the Van't Hoff equation, which relates it to the molar concentrations (M) of the dissolved species. The required osmotic pressure drop ($\Delta \pi$) for a water treatment plant can be calculated using the following Eq. (4).

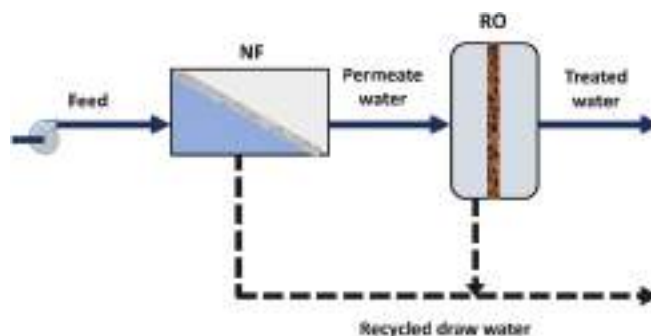


Fig. 4. Typical diagram of the hybrid NF and RO process for removal of micropollutant from water.

$$\pi = M\mathbb{R}T \quad (4)$$

where M - molar concentration of dissolved species (mol/L), \mathbb{R} - ideal gas constant (0.08206 L•atm/mol•K), & T - temperature of water (K).

The ability of a membrane to separate solutes, specifically salt, from the feed solution can be determined by employing Eq. (5), which calculates the percentage of membrane removal. (χ). This percentage increases as a function of applied pressure.

$$\chi = \left(\frac{c_{j0} - c_{jl}}{c_{j0}} \right) \times 100 \quad (5)$$

where χ in the membrane removal percentage (%), c_{j0} is the initial mole concentration of component j (ppm), and c_{jl} is the final mole concentration of component j (ppm).

According to the study results, it is essential to highlight the usefulness of using the dissolution-diffusion model to determine the most favorable membrane parameters in the case of salt removal; this model can be helpful for other cases. On the other hand, nanofiltration (NF) can be used to filtrate effluents contaminated with organic compounds, salts, divalent ions, and some microorganisms. The membrane pore size used by NF is (RO < NF < microfiltration (MF) and ultrafiltration (UF)). Membranes of hydrophilic character in a tubular or spiral configuration are mainly used, the most common material being cellulose acetate. This type of membrane has a very high remotion capacity (>99 %) for multivalent ions (e.g., sulfate, iron, and calcium ions) with low to moderate remotion capacity ($\cong 70$ %) and high rejection (>90 %) for monovalent ions (e.g., K^+ , Na^+). The parameters contributing most to membrane performance are solvent viscosity, pH, temperature, flux, pressure, pore size, membrane loading, concentration, membrane hydration, and membrane fouling. The flux rate, one of the critical parameters, can be calculated from Eq. (6):

$$F_p = \frac{V}{At} \quad (6)$$

where F_p is the flux rate of permeate (L/m²•h), ΔV the cumulative volume of permeate (L), A - membrane surface (m²), and Δt the time of filtration (h).

Fouling, influenced by pressure and concentration polarization, leads to a decrease in permeate flux. This decrease can be attributed to additional resistance to mass transfer within the membrane. As proposed by Silva, the relationship between the permeate flux and the pressure variation applied on both sides of the membrane can be described by Eq. (7). [149]:

$$J = \frac{1}{\mu R_T} P \quad (7)$$

where μ is the permeate viscosity, R_T is the total resistance of flow, and ΔP the pressure difference.

The total resistance of flow (R_T) can be calculated from Eq. (8):

$$R_T = R_m + R_a + R_b + R_g + R_{cp} \quad (8)$$

where R_m is the resistance caused by a membrane, R_a & R_b the resistance occurs due to adsorption & pore blockage, R_g & R_{cp} the resistance caused by the gel layer of the membrane, and the resistance produced by concentration polarization.

Wastewater treatment is a process that can be physicochemical and biological, aiming to eliminate pollutants from both sanitary and industrial effluents. This process may have three or more stages, depending on the physicochemical and microbiological characteristics of the wastewater and the types of contaminants present in it; in particular, micropollutants are those agents of either biological or chemical origin that reach the water in quantities equal to or less than micrograms per liter. Different technologies have been used to treat them, from the more traditional ones, such as adsorption, coagulation,

and biological treatment, to more advanced treatments, such as membrane separation and its variants. Hybrid systems can be couplings of different membrane processes or even the coupling of other physicochemical and biological processes with membrane processes. For example, they incorporate TiO₂ and graphene oxide in reverse osmosis membranes. In this research, they studied dye removal (100 %) and pharmaceutically active compounds (up to 95.7 %) [150]. A hybrid system that consists of combining different technologies, reverse osmosis, nanofiltration, and electrocoagulation. For biologically treated textile effluent, this study reports promising removal efficiencies for the textile industry (chemical oxygen demand COD >93 %, conductivity 99 %, chloride 97 % and TDS 91 %) [151]. Each technology has advantages and disadvantages; sometimes, you may opt for one in particular or a combination. Table 2 shows recent results for RO and NF in removing micropollutants.

6.2. Micropollutants (MPs) removal using hybrid forward osmosis (FO) and membrane distillation (MD)

Forward osmosis (FO) is a separation technique that harnesses the natural osmotic process to transport water molecules across a semi-permeable membrane. The feed solution (e.g., industrial wastewater) is brought into contact with the membrane during this process. Simultaneously, an extraction solution with a higher total dissolved solids (TDS) is present on the other side of the membrane. The disparity in TDS concentration between the two solutions generates an osmotic pressure, which facilitates water movement from the feed solution into the extraction solution. As a result, contaminants present in the feed solution are retained while the water molecules pass through the membrane. [152]. The extraction solution can be a simple mixture of water, salt, or another compound specific to the desired application. The principal difference between RO and FO is how the water passes through the membrane; RO requires external pressure, while FO uses natural osmotic pressure. Working at low pressure allows FO to present fewer problems with membrane fouling. Fig. 5 illustrates the hybrid forward osmosis (FO) and membrane distillation (MD) system for micropollutant removal.

The water flow through the membrane (J_w) considering a FO membrane is expressed as (Eq. (9)):

$$J_w = A(\pi_D - \pi_F) \quad (9)$$

where A - hydraulic permeability constant (m/s/atm), π_D - draw solution osmotic pressure (atm), and π_F - feed solution osmotic pressure (atm).

It is essential to highlight that Eq. (9) does not account for the influence of concentration polarization on the osmotic difference and the driving force in the forward osmosis process. However, Eqs. (10) and (11) consider the effects of internal and external concentration polarization when calculating water flux. These equations account for two different operating modes: the active layer facing the feed solution (AL-FS) and the active layer of the membrane facing the draw solution (AL-DS). By considering these modes, the impact of concentration polarization on water flux can be accurately estimated [153]:

$$(AL - DS) J_w = A \left(\frac{\pi_{D,b} \exp\left(\frac{-J_w}{k}\right) - \pi_{f,b} \exp\left(\frac{J_w}{D}\right)}{1 + \frac{B}{J_w} \left(\exp\left(\frac{J_w}{D}\right) - \exp\left(\frac{-J_w}{k}\right) \right)} \right) \quad (10)$$

$$(AL - FS) J_w = A \left(\frac{\pi_{D,b} \exp\left(\frac{-J_w}{k}\right) - \pi_{f,b} \exp\left(\frac{J_w}{D}\right)}{1 + \frac{B}{J_w} \left(\exp\left(\frac{J_w}{k}\right) - \exp\left(\frac{-J_w}{D}\right) \right)} \right) \quad (11)$$

where $\pi_{D,b}$ is the osmotic pressure of the bulk draw (atm), $\pi_{f,b}$ the bulk feed solutions (atm), k is the mass transfer coefficient (m/s), B is the solute permeability coefficient (m/s/atm), and K as the solute resistivity

Table 2

Illustrates the NF, MF, and RO usage, operating conditions, and removal efficiencies towards particular pollutants.

Technology	Membranes materials	Pollutant	Operation conditions	Removal efficiency	Reference
Activated Carbon with Al ₂ O ₃ NPs, NF, and RO Membranes	NF: Thin film of polyamide RO: Composite Polyamide	Oil field wastewater	Effective area 140 cm ² . 0–69 Bar Operating pressure range. 1.8 L/min Feed flow rate	TOC: 95.2 % TDS: 99.67 %	[167]
Microfiltration-adsorption (MF-GAC) NF and RO	MF: A flat sheet membrane of polyvinylidene fluoride (PVDF) Granular-Activated Carbon (GAC) NF: Two hydrophilic NP030 (polyethersulfone) and NTR729HF (Polyvinylalcohol/polyamides) RO: polyamides	Micropollutants (analgesics, lipid regulators, antimicrobial agents, beta-blockers, pharmaceuticals, flame retardants, and pesticides)	MF-GAC: surface area 0.2 m ² , 10 L reactor tank, 2 g/L GAC dose 20 °C temperature, 4 bar, clean water flux 12 and 62 L/m ² ·h RO: 40 bar. 23.5 L/m ² ·h clean water flux	MF-GAC and NF: 70 to 95 % RO: >90 %	[168]
Nanofiltration and Reverse Osmosis	RO: thin composite film membrane polyamide-urea copolymer barrier layer. NF: thin-film polyamide	Heavy metals	Feed capacity 1/2 L, maximum pressure of RO 50 bar and NF 38 bar	RO: Cr ³⁺ (99.2 %), Pb ²⁺ (98.8 %), Cd ²⁺ (98.6), As ³⁺ (99.2 %), Ni ²⁺ (98.4 %), and Sb ³⁺ (98.8 %) NF: Cr (98.2 %), Ni (97.8), Cd (92.3 %), Pb (76.9 %), As (52.5 %), and Sb (64.1 %) >99 %	[169]
Reverse Osmosis/Nanofiltration (RO/NF) and Membrane bioreactor	Commercial NF90/RO XLE	Erythromycin, azithromycin, diclofenac, clarithromycin, methiocarb, clothianidin, thiamethoxam, oxadiazon, acetamiprid, imidacloprid, tri-al-late	Experiments of NF/RO were in batch mode for 3 h with recirculation. The flow rate of 3 L/min and pressure of 12 bar		[170]
hybrid system coagulation-reverse osmosis (RO)-nanofiltration (NF)/ultrafiltration (UF)	UF ceramic tubular membrane NF polyamide spiral wound membrane RO dense polyamide spiral wound membrane	Landfill leachate	FeCl ₃ and Al ₂ (SO ₄) ₃ as coagulants UF 10 bar, NF 41 bar (area = 7.6 m ²), RO 69 bar (area = 7 m ²)	62 % for TOC, 89 % for TSD, and 62 % for iron	[171]
Hybrid nanofiltration with adsorptive supports	PVC UF hollow fiber (HF) membranes Natural bentonite and LDH nano clays were used as adsorbents in the support layer.	Boron	16.1 L/m ² ·h Water flux Pressure 4 bar 25 cm length of 50 fibers with and ~300 cm ² effective surface area	83 %	[172]
Forward osmosis membrane bioreactor-membrane distillation (FOMBR-MD)	FO: cellulose triacetate embedded support (CTA-ES) MD: polytetrafluoroethylene (PTFE) membrane	Dairy wastewater TOC (920 ± 45.8 mg/L) COD (3000 ± 121.7 mg/L) TN (81 ± 12.1 mg/L) TP (11.7 ± 1.55 mg/L) NH ₄ ⁺ (26.4 ± 4.30 mg/L) NO ₃ ⁻ (5.3 ± 0.85 mg/L) Ca (26 ± 0.36 mg/L) Na (68 ± 2.9 mg/L) pH (7.4–8.5) Conductivity (942 ± 18.4 µS/cm)	Bacillus-FOMBR FO effective surface area = 90 cm ² MD effective membrane area = 400 cm ² Configuration of MD = AGMD, 70 L/h 40 operation days	TN efficiency (40–79 %) almost 100 % of the contaminants	[173]
Forward osmosis (FO)-membrane distillation (MD) hybrid system	FO: cellulose triacetate (CTA) (FTS H2O™ flat sheet membrane). MD: polytetrafluoroethylene (0.2 µm pore size, CF042)	Human urine	FO effective area of 34 cm ² MD flat sheet membrane of 37 cm ² effective area Configuration of MD = DCMD 40 °C for draw solution 24 h operation time	Anions, Cations, and DOC rejection >79.5 % and 93.7 % in the hybrid system and FO side	[174]
Ultrasonic stripping-membrane distillation (US-MD)	Hydrophobic polytetrafluoroethylene (PTFE) membrane	Mariculture wastewater	Power input of ultrasonic probe = 80 W MD configuration = DCMD Effective area = 50 cm ² US time = 2 h Temperature 80 °C of water bath kettle and 15 °C condensate tank	NH ₄ ⁺ -N MD (71.7 %) US-MD (86.8 %)	[175]
Hybrid nanobubble-forward osmosis system (NBs-FO)	Thin film composite-polyamide flat sheet	Aquaculture wastewater	Ozone and air NBs 0.00231 m ² Active membrane surface	OM, TDS, and PCC (~98 %)	[176]
UV-LED-membrane distillation process (UV-LED-MD)	Flat-sheet MD of polytetrafluoroethylene (PTFE) in a non-woven PPY layer	Hormones (17α-ethynylestradiol and estrogenic activity)	89.25 cm ² effective membrane area. 24.4 cm ² Quartz plate area 4 W UVC LED Time for each test of 550 min 60 L/m ² ·h flux, 40 min filtration Time, 100 mL/min Flow rate with 2 mg/L concentration	(>97.7 %) hormone removal efficiencies & >99.0 % estrogenic activity removals	[177]
Catalytic ceramic membranes (CCM)	Commercially ceramic membranes (0.1 µm, Sinotsing Environment, China)-MnCe oxide	Atrazine		99.99 %	[178]

(continued on next page)

Table 2 (continued)

Technology	Membranes materials	Pollutant	Operation conditions	Removal efficiency	Reference
	β -SiC nano-wires/g-C ₃ N ₄ @ β -SiC nano-wire heterogeneous membranes	Organic dyes (methylene orange)	1.0×10^{-4} m ² effective membrane area, 1 bar trans-membrane pressure photo-catalytic degradation of 50-min	100 %	[179]
	Aluminum oxide (Al ₂ O ₃), copper oxide (CuO), and titanium oxide (TiO ₂)	Bisphenol A	~15 cm ² effective membrane area of four ceramic HF was BPA solution 10 mg/L using batch mode and 0.5 mM PMS concentration 25 °C temperature	91.46 %	[180]

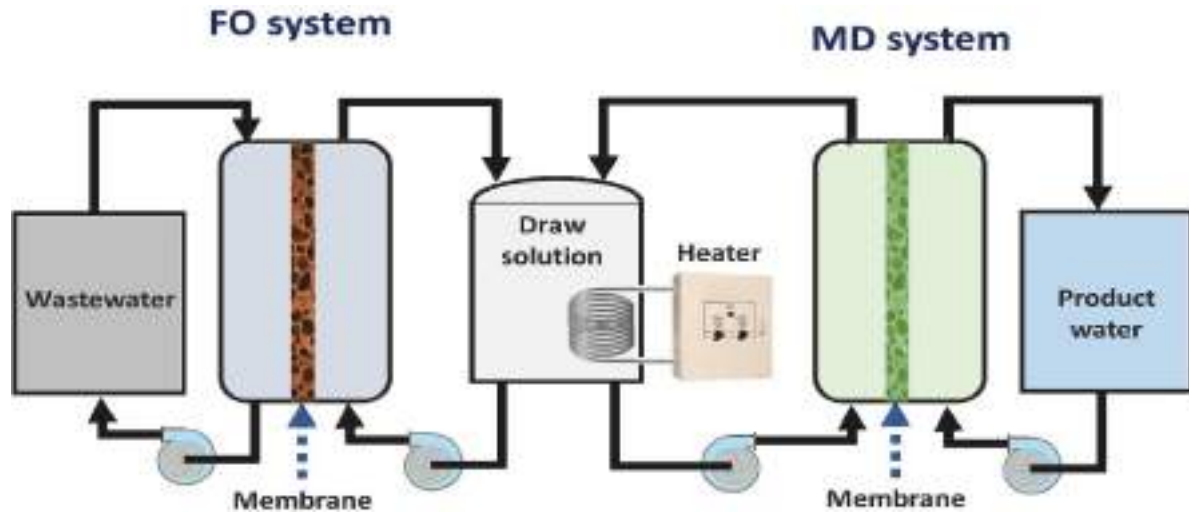


Fig. 5. Schematic diagram of hybrid forward osmosis (FO) and membrane distillation (MD) system for micropollutant removal.

(s/m). Solute resistivity and Mass transfer coefficient are the coefficients representing ICP and ECP.

The bulk osmotic pressure at higher concentrations can be calculated considering the water activity according to Eq. (12):

$$\pi = - \left(\frac{RT}{V} \right) \ln(a_w) \quad (12)$$

where a_w - water activity & V - molar water volume. On the other hand, water activity can be intended using the Pitzer from Eq. (13):

$$a_w = \exp \left(-0.01802 \sum_i M_i \right) \quad (13)$$

M_i - molality (moles) of the solute per kg of solvent, and ϕ - osmotic coefficient. It can be estimated from Eqs. (14)–(18).

$$-1 = z_m z_x F + 2m \left(\frac{v_m v_x}{v} \right) + 2m^2 \left(\frac{(v_m v_x)^{1.5}}{v} \right) C_{mx} \quad (14)$$

$$F = - \frac{0.3921^{0.5}}{1 + 1.2 I^{0.5}} \quad (15)$$

$$I = 0.5 \sum_i m_i z_i^2 \quad (16)$$

$$|z_m z_x| = \frac{\sum_i m_i z_i^2}{\sum_i m_i} \quad (17)$$

$$B_{mx} = B_{mx}(0) + \exp(-2.0 I^{0.5}) \quad (18)$$

where z_m and z_x are the charges of x and m ions, and v_m and v_x are the respective stoichiometric coefficient of the ions, C_{mx} , $B_{mx}(0)$, and $B_{mx}(I)$

are the Pitzer equation constants specific to each solute.

The parameter k , which is related to the effect of ECP, can be calculated by the dimensionless Sherwood number (Eq. (19)):

$$k = \frac{Sh D}{d_h} \quad (19)$$

where Sh - Sherwood number, D - diffusion coefficient (m²/s), and d_h - hydraulic diameter of the membrane channel (m). The parameter d_h can be calculated from Eq. (20):

$$d_h = \frac{4WH}{W + H} \quad (20)$$

H and W are the height and width of the rectangular membrane channel. To estimate the Sherwood number, it is necessary to know the flow conditions according to the Reynolds number (Re): $Re \leq 2100$ laminar, $Re > 4000$ turbulent, and between $2100 \geq Re \geq 4000$ in transition, using Eq. (21).

$$Re = \frac{d_h v \rho_F}{\mu_F} \quad (21)$$

Being v the velocity (m/s), ρ the liquid density (kg/m³), & μ the dynamic viscosity. Then according to the Re number, Sh values can be estimated (Eqs. (22) and (23)):

$$\text{Laminar flow } Sh = 1.85 \left(Re Sc \frac{d_h}{L} \right)^{0.33} \quad (22)$$

$$\text{Turbulent flow } Sh = 1.85 Re^{0.75} Sc^{0.33} \quad (23)$$

where L is the length, and Sc is the Schmidt, and it can be estimated from Eq. (24):

$$Sc = \frac{\nu}{D} \quad (24)$$

Being ν the kinematic viscosity. On the other hand, the parameter K , which is related to the ICP effect, is defined by Eq. (25):

$$K = \frac{\tau t}{D\varepsilon} \quad (25)$$

where τ - tortuosity, t - support layer thickness, and ε - porosity. Then, the term $\tau t/\varepsilon$ is identified as the membrane structure parameter S and replaced in Eqs. (10) and (11) to obtain Eqs. (26) and (27):

$$(AL - DS) J_w = A \left(\frac{\pi_{D,b} \exp \left(\frac{-J_w K}{k} \right) - \pi_{f,b} \exp(J_w K)}{1 + \frac{B}{J_w} (\exp \left(\frac{J_w K}{k} \right) - \exp \left(\frac{J_w K}{k} \right))} \right) \quad (26)$$

$$(AL - FS) J_w = A \left(\frac{\pi_{D,b} \exp \left(-J_w K \right) - \pi_{f,b} \exp \left(\frac{J_w}{k} \right)}{1 + \frac{B}{J_w} (\exp \left(\frac{J_w}{k} \right) - \exp \left(-J_w K \right))} \right) \quad (27)$$

The modeling of FO can help prognosticate the water flux in the systems under various experimental conditions at the laboratory level.

Up to this point, the principle of operation and the equations for modeling the system have been discussed. However, it is complicated to use FO alone because it is necessary to combine it with another separation process in many applications to separate the extraction water from the final product or to use FO as an advanced pretreatment process. This adaptation with another technology is called hybrid, like membrane distillation (MD).

MD is a membrane technology for treating saline water and wastewater treatment; it is a thermal separation process in which exclusively steam can go through the membrane (hydrophobic porous membrane, e. g., PTFE, PVDF, or PP). For this to occur, there is a vapor pressure difference between the membranes. The membrane is fed with a hot flow, and cold water is fed on the other side (permeate flow), so the vapor will penetrate via pores and condense on the colder side [154]. There are four configurations for the MD, the difference between which lies in the way in which the permeate is collected and driving force generation: a) direct contact membrane distillation (DCMD), in this mode, the hot feed is in direct contact with all the exterior of the hot side of the membrane, the evaporation and entrainment of vapor molecules occur on the feed side membrane surface and moves into the permeate, condensing inside the membrane module; b) air gap membrane distillation (AGMD) In this case, the feed solution will only be in contact with the hot side of the membrane, on the other hand, the air is introduced and will be stagnating between the cold surface and cold solution. The vapor will pass through the membrane from the hot side and continue moving towards the air gap to the permeate side. This system has the particularity of increasing the resistance to mass transfer because of the presence of air, but it is more energy efficient. Some examples of applications are in water desalination, removal of certain salts with high concentrations (chlorides, carbonates, sulfates) and some volatile organic compounds; c) sweeping gas membrane distillation (SGMD), unlike the previous case, an inert gas is employed to sweep the vapor from the permeate side, condensation takes place outside the membrane zone. The gas acts as a barrier which helps to reduce heat loss and is not stationary. This configuration is often used for the removal of volatile compounds in solution; and d) vacuum membrane distillation (VMD), Which requires a vacuum pump on the side of the permeate membrane. The vapor transported through the membrane constantly moves from the vacuum zone to the external condenser. It has the advantage that the conduction heat is negligible.

It is also possible to mathematically model for DCMD configuration by considering mass and heat transfer, as reported by Olantunji & Camacho [155]; such a study can help determine the optimum operating

conditions and reduce the effect of temperature & concentration polarization on permeate flux decrease. Similar to the above technologies, other physical, biological, and chemical processes can be combined with forward osmosis, membrane distillation, or other methods (reverse osmosis or nanofiltration). Like the research work published by Fatima et al., where a sequential system was used for treating Poultry Slaughterhouse Wastewater, obtaining a removal of COD (100 %), total solids TS (100 %), total volatile solids TVS (100 %), total suspended solids TSS (100 %), total fixed solids TFS (100), total phosphorus TP (100 %), and total nitrogen TN (62 %) [156]. Or the information published by Liu et al., who used an integrated loose nanofiltration (NF) - membrane distillation (MD) system for the recovery of resources from leachate MBR effluent, they recovered 96 % organics, 87 % salts and 82 % high-purity water [157]. Table 2 shows the NF, MF, and RO usage, operating conditions, and removal efficiencies towards particular pollutants.

6.3. Wastewater reuse and recycling

Presently, the combination of population growth, climate change, and heightened pollution levels has resulted in a significant water scarcity crisis in numerous regions across the globe. This issue will escalate as one of the most critical environmental concerns in the forthcoming decades. [158]. Recycling and reusing water for non-potable applications or replacing it with potable water are increasingly important alternatives for water resource management, offering an effective solution to the problem of water scarcity. This way, wastewater reuse can increase water supply for agriculture, industry, and drinking water use. Therefore, this alternative should be a high priority for enterprises since it offers advantages in reducing water supply costs and protecting the environment. Membrane technologies have a crucial role in wastewater treatment for water reuse, as they can be integrated with various pretreatment processes to meet the necessary quality standards for reutilizing treated water. This integration allows for achieving the required water quality for reuse purposes. As a result, the advancement of membrane technology in recent years has led to a notable increase in the implementation of water reuse projects globally.

Reverse osmosis is a crucial membrane technology for producing water with high purity from wastewater treatment; this technology offers better guarantees for applications where potable water is required due to its ability to simultaneously remove a wide range of contaminants, such as pathogens (bacteria and viruses) total dissolved solids, and low molecular weight chemical contaminants [159]. This membrane technology is also a viable alternative for producing potable water from seawater or brackish water [160]. Utilizing nanofiltration membranes in wastewater treatment is promising for various applications, including recycling, water reuse, and recovering valuable products in textiles, food, oil, mining, tannery, pharmaceuticals, and paper manufacturing [161]. Moreover, hybrid membrane processes offer an exciting alternative for wastewater purification and subsequent reuse, providing additional opportunities for efficient and sustainable water management. [162]. Recently, Racar et al. evaluated the effect of incorporating the NF/RO system in a wastewater treatment process with MBR technology; in this study, they compared the decrease of physicochemical and microbiological parameters and the removal of eleven micropollutants detected. The results indicated that the incorporation of NF90/RO treatment after MBR allows obtaining an effluent with the necessary quality to be reused for irrigation; these results cannot be obtained with the simple use of MBR or the additional treatment with nanofiltration (NF270). In addition, the NF/RO treatment showed excellent removal rates (>99 %) for all the micropollutants detected [163].

6.4. Cost-effectiveness and sustainability of membrane-based technologies

Organic micropollutants (MPs) are more common in water and wastewater due to the increasing use of medications, endocrine

disruptive substances, and personal care products for healthcare and improving living conditions. Providing secure and safe water supplies is complicated because conventional wastewater treatment plants have inherent limitations in dealing with these substances. Membrane technology has been identified as a potentially helpful strategy for dealing with this new issue. To ensure the effectiveness and practicality of membrane-based treatment approaches in large-scale applications, it is crucial to comprehend the behavior of microplastics (MPs) and the underlying mechanisms involved in their removal. Advanced membrane technologies, such as nanofiltration, reverse osmosis, ultrafiltration, membrane distillation, and forward osmosis, have emerged as effective methods. These cutting-edge membrane-based treatment technologies, among others, are highlighted in the paper as viable options for addressing the challenge of removing MPs from various water sources.

Pharmaceutical active compounds (PhACs), endocrine-disrupting chemicals (EDCs), and personal care products (PCPs) are just a few examples of organic micropollutants (MPs) that continue to pose a significant problem for the water ecosystem. The issue of organic MPs directly into water systems is expected to increase due to the growing urbanization and the greater reliance of modern civilizations on pesticides, bactericides, herbicides, and fungicides. The potential complexity of MPs' harmful effects on human health, primarily upon chronic exposure via the water supply, has recently raised growing concerns. The European Union (EU) has implemented stringent controls on the discharge of organic MPs due to rising concerns about their presence in water bodies [8,24].

Similarly, the USEPA has placed numerous organic MPs on a Contaminant Candidate List to track their prevalence, entry points, and possible dangers to human health. Water/wastewater treatment and reuse have grown increasingly interested in MP removal utilizing cutting-edge membrane technology. Several specialized review studies have summarized the extensive work done on this topic due to its widespread interest. The coverage of novels and emerging membrane technologies, such as membrane distillation (MD) and forward osmosis (FO), has been relatively restricted in prior evaluations, which have mainly focused on commercial reverse osmosis (RO) and nanofiltration (NF). Despite the topic's relevance and imminent nature, no severe evaluation has focused on developing membrane technologies to eliminate organic MPs [25].

Both RO and NF use more energy than other pressure-driven membrane-based treatment systems like microfiltration (MF) and UF since they are pressure-driven processes. Although RO and NF require a lot of pressure, they are increasingly used for desalination and water purification. Due to the excellent purity of NF/RO-treated effluents, their usage in tertiary treatments in wastewater/sewage treatment facilities is also being advocated [19,20].

Indeed, to achieve an effective removal of microplastics (MPs) using nanofiltration (NF) and reverse osmosis (RO) processes, it is essential to comprehend the fundamental mechanisms at play. These mechanisms include electrostatic interaction, hydrophobic interaction, and size exclusion. MP particles possess diverse physicochemical characteristics, such as size, charge, solubility, diffusivity, and hydrophobicity. Understanding these characteristics is crucial for optimizing NF/RO processes and tailoring them to effectively remove MPs by leveraging the appropriate mechanisms based on the specific properties of the MP particles. MPs, with size exclusion and adsorption being the primary means of eliminating them. Ibuprofen, dipyrone, and diclofenac are all examples of negatively charged MPs, and they were more easily removed from the body than neutrally charged MPs like acetaminophen and caffeine due to the electrostatic repulsion between the membrane surface and the negatively charged MPs. Few came to a similar conclusion, reporting a robust inverse association between neutral-hydrophilic MP size and passage. The link was not as strong between moderately hydrophobic MPs and depression. Hydrophobic moieties like aromatic rings and hydrocarbon chains have an affinity for the active layer of RO membranes, which leads to poor removal, as explained by the author. The primary

strategy for removing some MPs is size exclusion. Bisphenol A, for example, was released at a lower rate (74.1 %), compared to ibuprofen (98.12 %) and salicylic acid (97 %), due to its nonionic composition at the chosen pH.

In comparison to deprotonated/negatively-charged ibuprofen (pKa 4.9) and salicylic acid (pKa 2.9), bisphenol A exhibits a higher pKa value (9.6–10.2), which results in a less significant contribution of electrostatic interaction between the negatively-charged membrane surface and the compound due to its lower degree of ionization. Consequently, the electrostatic contribution is mainly absent or negligible in the case of bisphenol A. [43]. Therefore, the size exclusion mechanism was the primary removal mechanism for bisphenol A, in contrast to ibuprofen and salicylic acid. Size exclusion was the principal mechanism for hydrophilic neutral chemicals with high water partitioning coefficients, consistent with previous observations. Regarding positively charged microplastics (MPs), their removal effectiveness can be reduced due to electrostatic interactions with the negatively charged membrane surface. These interactions lead to the adsorption of positively charged MPs onto the membrane, hindering their diffusion and potentially diminishing their removal efficiency. Therefore, electrostatic interactions can impede the removal of positively charged MPs effectively during membrane-based treatment processes. [43].

6.5. Mechanisms of removal by different membrane types

Through effluent discharge and reuse of treated sludge and sewage used in agricultural applications, their residues enter the food chain and accumulate, harming human and animal health. Many micro-pollutants have been associated with carcinogenic, mutagenic, and reproductive toxicity hazards and have been found harmful to freshwater invertebrates, mussels, human embryonic cells, and fish [164–166]. Their effects on plant development and growth and agricultural operations are not limited to high environmental concentrations or the use of reclaimed water for irrigation [11]. Recycled water is typically a mixture of the MF/UF and RO permeate. Sydney, Australia's Water Reclamation Scheme (WRAMS) combines MF permeate, and RO permeate to create recycled water. The RO permeate ratio is 50 % and 20 % of the final product water. WRAMS's recycled water contains a higher concentration of micro-pollutants since it is drawn from a more significant volume of Biologically treated sewage effluent (BTSE). The reclaimed water is then resold to homeowners for non-drinking purposes, including watering lawns and gardens, filling fountains and ponds, and irrigating parks [11]. Membrane filtration, adsorption, improved oxidation techniques, and various forms of biological treatment have all been investigated as potential means of purifying micro-pollutant feed water. The majority of the micro-pollutants in BTSE are removed by the intercalation of granular activated carbon (GAC) and MF in our prior experiments (>90 %). Sorbents, including Xylit, lignite, sand, and GAC, were tested, and their Log Kow (octanol-water partition coefficient) values were found to correlate positively with the efficiency with which they removed micro-pollutants. GAC was determined to be the most effective method for treating onsite sewage systems for micro-pollutants. Research into methods of ozonation for removing these micro-pollutants has received some attention. However, it has been noted that ozonation can have harmful side effects. They also suggested using GAC filters as a secondary treatment to eliminate micro-pollutants and other hydrophobic substances. Only a few people have argued that ozonation methods are less effective than activated carbon adsorption for removing micro-pollutants like pesticides, industrial chemicals, antibiotics, antidepressants, and surfactants. Micro-pollutants in feedwater were effectively removed by membrane filtration, making this a feasible treatment option. Although nano-filtration (NF) was shown to be effective at eliminating micro-pollutants, reverse osmosis (RO) was found to be even more so (>90 %) [164–166].

Permeate quality is comparable to MF (with pore sizes of 200 nm) and UF (3 nm) membranes, which reject micro-pollutants

predominantly by size exclusion. Size exclusion, hydrophobic interaction, electrostatic interaction (Donnan effect), orientation and charge of the molecule to the membrane, H-bonding, etc., are all ways NF/RO processes efficiently remove micro-pollutants. The mechanism driving the removal of micro-pollutants by the various types of membranes must be studied to understand this process. This allows for the development of more tailored and suitable water supply systems [164–166]. Therefore, the BTSE requires a change or addition to the treatment method to maximize the elimination of overlooked micro-pollutants. There have been hardly any investigations into the secure distribution of recycled water free of or containing only trace amounts of micro-pollutants. To generate reclaimed water that is safe for the environment, it is necessary to remove the micro-pollutants from the microfiltered BTSE, and the goal of this research is to identify viable membrane-based treatment solutions, such as the microfiltration-adsorption (MF-GAC) hybrid system, which combines the NF and RO process. In addition, the existing dual membrane technology is expensive because it combines MF and RO to produce recycled water. The report highlights using energy-efficient membrane-based hybrid treatment systems such as the NF and MF-GAC to remove micro-pollutants from BTSE.

6.6. Factors affecting the performance of membrane-based technologies for micropollutant removal

For evaluating Membrane-based technologies, we should include membrane properties, physicochemical characteristics of chemicals to be removed, transport mechanism, and matrix influence. Size exclusion, adsorption, electrostatic repulsion, solute-solute interaction, diffusion, fouling, and so on all play a role in the membrane's selectivity [38,39]. Adsorption of ibuprofen by RO and NF membranes was evaluated by a few researchers, who concluded that the electrostatic repulsion between the membrane and the pollutant is directly related to the pH of the solution. By lowering the pH below ibuprofen's pKa (acid dissociation constant), the membrane becomes positively charged, allowing for the easier adsorption of the negatively charged ibuprofen inside the membrane. NF and RO membranes were more effective at removing ionic chemicals (97 %) than nonionic molecules (82 %). This mechanism, known as diffusion, reduces the membrane's adsorption capacity and, thus, its ability to remove chemicals. Due to electrostatic repulsion between this micropollutant and the membrane, RO membranes (negative surface charge) eliminated diclofenac at >95 % removal efficiencies. Diclofenac and other medications and personal care items with negative charges in solution, like ibuprofen, glimepiride, naxoprene, and sulfamethoxazole, were found to have the same impact in a separate investigation. Micropollutants with neutral or positive charges were much more difficult to remove. Adsorption removed nearly all of the for-naxoprene, whereas it only removed around 20 % of the acetaminophen (which is neutral) and 60 % of the atenolol (which is positive). Substances with high pKa, and poor hydrophobicity, such as diatrizoate and carbamazepine, were shown to have negligible adsorption by NF and RO membranes. Hence size exclusion was found to be the efficient mechanism. The notion that hydrophilic chemicals with neutral charges are only faintly adsorbed in the absence of fouling was also validated. Similar findings were also reported by a small sample size, substantiating that improved removal rates are observed for larger values of the octanol-water partition coefficient ($\log K_{ow}$).

6.7. Recent developments in membrane materials and designs for improved performance and scale-up challenges and opportunities

The commercialization process has been painfully slow because of all the obstacles. Despite its many benefits, commercial development of membrane distillation process (MD) and MD membranes has been painfully sluggish. Minimal advancement was made in the first three decades, and most of the studies conducted during this time were fundamental, seeking a better understanding of the MD process and its

various configurations. In addition, MOFs are the most often employed fillers in mixed matrix membranes due to their exceptional physical and chemical capabilities. MOFs have excellent filler characteristics because their organic ligands strongly bind polymeric chains [11]. In addition, the pore size and structure of membranes based on MOFs can be adjusted to meet the needs of a given application, allowing for more control over membrane selectivity and permeability. Because of their well-organized structure, large pore volume, and specific surface area, they can alter the membrane's structure and increase transmembrane flux. When fabricating multifunctional composite membranes, carbon-based nanomaterials stand out as a superior class of nanomaterials thanks to their unique properties, such as a high surface area to volume ratio, high mechanical strength, and a reduced fouling propensity. Due to their high-quality physicochemical features and low production cost, quantum dots have recently gained a lot of interest as a membrane modification for MD applications. In this study, we closely examine the latest generation of NPs employed in MD research, with an eye on how they might improve membranes' inherent anti-fouling and superhydrophobic qualities. Pure water flux, salt rejection, pore size, porosity, water contact angle, liquid entry pressure (LEPw) value, and membranes' longevity in operation are just some metrics used to assess modified membranes [11]. In addition, we have elaborated on membrane modification techniques, which are often glossed over when talking about MD membrane production. The primary objective of this review was to provide a broad overview of the many different types of cutting-edge membranes created for MD applications out of innovative materials such as metallic NPs, carbon nanotubes, graphene and its derivatives, and metal-organic frameworks. While other works have addressed improving membrane characteristics by incorporating NPs, our research seeks to provide in-depth scientific analyses of the mechanisms responsible for the enhanced properties of nanoparticle-modified membranes. Effectiveness and tactics for designing membrane nanostructures for enhanced performance in MD are the topics of this review paper. This article also discusses the pros and cons of several nanoparticles (NP) and nanomaterial-based membranes currently developing.

7. Chemical aspect of membrane treatment

One of the best ways to enhance membranes' characteristics and boost their performance in several applications is through modification. To minimize the rejection of micropollutants, it is based on the adaptation of organic contaminant removal mechanisms by membranes, including hydrophobic interaction, size exclusion, and electrostatic interaction [181]. In this context, different membrane modification processes and their effect on contaminant removal are listed as follows.

7.1. Based on surface charges

The zeta potential or membrane surface charge determines the membrane driving force. The electrostatic attraction between a membrane's surface and a microbe's surface charge affects how well a membrane function [182]. The majority of membrane surfaces have a negative charge at neutral pH levels. However, the charge may be changed by modifying the solution conditions since the chemical groups on a membrane surface are amphoteric [183]. Hu et al. examined the removal of the MS2 bacteriophage virus using RO membranes made of various materials and operating under various pressures. According to Hu et, polyamide RO membrane was used to accomplish a better log removal for MS2 bacteriophage at a 100 psi operating pressure [184]. Moreover, Antony et al. demonstrated that polyamide RO membranes with a slight negative charge outperformed cellulose acetate and polysulfone membranes regarding viral rejection. The isoelectric point of MS2 bacteriophages is pH 3.9, indicating that the charge is negative at higher pH [185]. Consequently, these repelling forces aid in rejecting viruses when both the membrane and the viral particles have a negative

charge. Under this approach, the preparation of a nanofiltration membrane with a high negative surface charge and its evaluation for removing the anionic dye was recently reported. The membrane was fabricated using a monomer of sulfonamide, trimethyl chloride (TMC), and 2,4,6-trimethyl-3,5-diamino benzenesulfonamide (TMDBSA) by interfacial polymerization. The results indicate that under optimized conditions, the membrane shows high water permeability ($19.6 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$) and effective rejection of dyes (anionic) with different molecular weights. The membrane's exceptional antifouling properties and long-term stability are also a result of its flat surface and steady surface charge. [186]. On the other hand, Huang et al. evaluated the effect of nanofiltration membrane modification on the removal of 34 organic micropollutants commonly detected in wastewater. This work used commercial membranes NF90 from DuPont Filmtec™ was used, which were modified by forming a polydopamine coating using different deposition times. In general, the removal of micropollutants improved with membrane modification, obtaining average removal values of 70 %, with only a 26.4 % decrease in water permeability. The membrane modified with the highest negative charge showed the best removal for the loaded organic compounds [181].

7.2. Based on surface modification

To change the membrane surface, several modifying agents can be utilized. Some membrane modification methods are sulfonation, amination, carboxylation, and epoxidation [187]. One of the most commonly used methods for adding a hydrophilic group to the surface of polyether membranes is sulfonation, which uses chlorosulfonic acid, sulfuric acid, and chloromethylation, which decreases the anti-fouling effect and oxygen permeability. Using glutaraldehyde, Zhang et al. cross-linked sulfonated polyvinyl alcohol (PVA) to thin-film conventional membranes. The PVA was evenly distributed throughout the, increasing the binding energy between surface and chemical moiety. The surface-modified membranes with glutaraldehyde showed 99.8 % rejection of NaCl [188]. Intending to cause the repulsion of macromolecules by steric repulsion, polyethylene glycol is one of the widely used surfactants to modify the membrane. These membranes can draw water molecules and tend to surround themselves with larger complexes. It has been demonstrated that zwitterionic species, including phosphocreatine, phosphatidylcholine, carboxy betaine, and sulfobetaine, are ideal for the surface modifier to avoid anti-fouling [189]. The formation of a surface coating is another standard method to modify the surface of membranes; in this way, Yadav et al. modified a commercial nanofiltration membrane (TRISEP® UA60) with a coating with graphene oxide and cross-linked kappa-carrageenan (κ -CGN) to evaluate to improve salt rejection and its anti-fouling property. The selectivity of rejection of divalent ions and the water recovery ratio were both dramatically enhanced by the -CGN-GO coating. Under the optimum conditions, the modified membrane showed a 95.73 % flux recovery ratio and an 18.9 % total fouling percentage with a slightly lower water flux than the commercial UA-60 membrane [190]. These substances retain charge neutrality depending on their surroundings despite having negatively and positively charged functional groups in the same side chain segment. To generate long-lasting anti-fouling capabilities, the fundamental concept behind the modification and the structural relationships between the modifier and membrane surface remain challenging to comprehend.

7.3. Based on electrically conducting ceramics

The control of membrane fouling by applying electric current is a novel technique. In this technique, the increase in charge density can be achieved by conducting membranes where the externally applied electrical currents or potentials are immobilized onto the surface of the membrane. The three main forces are electrostatic, electrophoretic, and electro-osmosis, allowing particle mobility during membrane filtration

[191]. However, the membrane's non-conductive nature complicates this method's application. Thus, a field of opportunity is the development of conductive membranes. Stainless steel (SS), titanium (Ti), copper (Cu), and nickel (Ni) membranes are the most common conductive porous membranes [192] [193]. SS and Ti membranes are widely used due to their exceptional mechanical stability and corrosion resistance. The conductivity and thickness determine a membrane's electrical resistance. The resistivity of the thick membrane is lower. Although thin-film conducting materials like carbon nanotubes (CNT) have excellent conductivity, their thickness ($1 \mu\text{m}$) creates a difficulty in resistance. Nevertheless, high membrane resistance might decrease the surface area due to the loss in conductivity in conducting ceramics, which are much thicker (0.25 cm) and have lower conductivity than CNT materials [194]. Recently, the formation of carbon nanotubes and dopamine deposition were reported elaborating on a conductive ceramic membrane. This study first coated the ceramic surface with polydopamine to create an adhesive interlayer before the conductive carbon nanotube coating. Once the membrane was prepared, the effectiveness of this method of controlling membrane fouling during the treatment of synthetic wastewater was evaluated. The developed ceramic membrane functioned well in filtering synthetic wastewater, including inorganic materials or organic contaminants (oil emulsion), and displayed outstanding electrical conductivity and stability. The results obtained show that the fouling rate of the membrane was reduced by approximately 50 % by applying a negative charge on the membrane and maintaining a continuous voltage of 2.0 V. This may be because the applied electric field exerts an electrostatic force on negatively charged contaminants, such as oil droplets and suspended particles, which prohibited the pollutants from adhering to the membrane surface [195].

Finally, electrical conductive membranes are mainly employed as supporting materials in electrocatalytic membrane reactors to remove persistent organic contaminants such as Rhodamine, methyl orange, paracetamol, norfloxacin, bisphenol A, atrazine, tetracycline, perfluorooctanoic acid, N Nitrosodimethylamine and amoxicillin [191].

7.4. Based on catalytic ceramic membranes (CCM)

Ceramic membranes are considered ideal catalyst substrates for their stability and long lifetime. These advantages make ceramic membranes a good choice for combining with oxidants to form an advanced oxidation process (AOP). In the area of wastewater treatment with micropollutants, CCM demonstrates the synergistic roles of ceramic membrane filtration and catalytic oxidation [196]; by example, coupling CMs with catalytic ozonation (O_3) has proven to be an efficient technology to improve hydroxyl radical generation and avoid membrane fouling [197]. The catalytic ceramic membrane further offers a platform for accommodating the catalyst to speed up the physical separation and chemical reaction by the membrane [198]. This technology has gained the attention of researchers as an alternative to the degradation of organic micropollutants. He et al. prepared a catalytic ceramic membrane incorporating MnMe oxide ($\text{Me} = \text{Co}, \text{Fe}, \text{Ce}$) and evaluated its efficiency in removing atrazine. The membranes prepared with Ce oxide presented the best performance, achieving atrazine removal close to 100 % in 40 min; such results are attributed to the high performance in the catalytic ozonation of the membrane (MnCe-CM), being the $\cdot\text{OH}$ radical, the main ROS that contributed to the degradation of the micropollutant. Another critical factor is that the reactions on the surface and inside the membrane micropores were very effective due to the high specific surface area of the membrane. The results show that a new MnCe-CM membrane with catalytic filtration and ozonation functions can be used in wastewater treatment [178]. Lee et al. (2019) proposed a mechanism of catalytic ozonation using CeOx alumina membranes for removing micropollutants, in which roles of ceramic membrane and catalyst (the sources of radical generation and the pathway of contaminant degradation) were established [199]. Another recently reported alternative is preparing a ceramic membrane with SiC nanoparticles

designed to improve its antifouling performance. The prepared membranes proved highly efficient for eliminating organic dyes like methylene orange under artificial sunlight, in strong alkaline or acidic conditions, reaching 100 % removal in 35 min at pH = 13 and 20 min at pH = 1. The superoxide radical ($\text{O}_2^{\cdot-}$) was confirmed as the main reactive species, and the hydroxyl radical (OH^\cdot) was in a smaller proportion. This work provides new knowledge for the design of CCM with auto-cleaning function and for more organic wastewater treatment applications in chemically aggressive environments, serving as a scientific basis for other catalytic filtration systems [179].

8. Development of a low-cost membrane from an alternative material

New membranes are being developed utilizing natural raw materials as alternatives. In addition to clays like kaolin, researchers are developing membranes from waste materials. Creating membranes from waste has attracted much interest by utilizing the “turn trash into gold.” There are three categories into industrial, animal, and agricultural ash. Their techniques to develop a membrane are discussed below.

8.1. Slip casting and tape casting approach

Advanced membrane preparation can be accomplished using the straightforward and affordable slip-casting process. This technique has long been employed in conventional ceramics [200]. Preparing a membrane in the slip-casting method includes a thoroughly mixed suspension before being added into a mold, allowing the solvents to diffuse over the pores due to the capillary action, which acts as the driving force. The cake layer is formed by a quick membrane condensation over the mold [201]. Water and clay/kaolin are typically used in the slip-casting mixture. Mohammadi and Pak reported ceramic membranes made of kaolin clay by combining an electrophoretic technique and slip casting to create a kaolin foundation for more durable zeolite NaA membranes. However, regulating the membrane thickness is problematic with slip casting [202]. Tape casting is a popular membrane manufacturing process for obtaining thin, smooth ceramic sheets. This method entails adding the suspension into a mold/reservoir, which is then passed over a movable knife that regulates the thickness of the membrane [203]. The resulting membrane moves through a drying zone where the solvent is removed from the surface of the membrane. One advantage of the tape casting method is that the membrane can be sliced into various shapes, round and rectangular [202]. The first inexpensive ceramic membrane created via tape casting comprises calcium carbonate, kaolin, quartz, boric acid, sodium metasilicate, and sodium carbonate. The membrane developed had a pore size and porosity from $\sim 0.7 \mu\text{m}$ and 25–55 %, respectively. It has been determined that the developed membrane is suitable for completely removing pathogens from water.

8.2. Pressing method approach

A common technique for manufacturing membranes is pressing. This technique often relies on pressing dry powder using a press machine/tool [204]. In fact, after thoroughly mixing the powder, the resulting mixture is pressed uniaxial, i.e., it is stressed in a mold with immovable dividers to get the appropriate membrane shape and configuration. The produced flat membrane requires a heat treatment for consolidation, typically at a temperature equal to that of the sintering of the reactant components [205]. In a membrane of concentrations of silver, lead, & copper (I) sulfide with kaolin with 0.1 MPa pressure, Hirata and Higashiyama 1970 reported a membrane with 15 mm dia and 2–3 mm thickness [206]. The significant disadvantages of the pressing procedure are that it is expensive and requires high-temperature sintering. Moreover, this method only permits the development of a symmetrical membrane structure.

8.3. Extrusion method approach

Another common technique for manufacturing membranes is extrusion, frequently used to develop porous tubular configurations. A plasticizing agent and a binding agent are required in this method to create a ceramic pulp with rheological properties. The mixture shaping technique determines the shape of the finished membrane support. This method is a deformation process where plastic is forced to move over the die hole using a simple piston process [207]. This process determines the tubular membrane porosity rate, pore size, and shape distribution. The moisture level of the mixture is controlled by composition with appropriate surfactant, binder, coagulant, de-flocculant, plasticizer, lubricant, and preservatives in the blending process. Using the extrusion technique, Issaoui et al. 2019 [208] produced tubular ceramic membranes using 80 % kaolin and 20 % starch; the water permeability efficiency was around 612 L/(h m² bar) after sintering at 1250 °C.

8.4. Phase inversion and sintering technique – an innovative step

Phase inversion technique is a technique that occurs when a polymer suspension transitions from a liquid state to a solid state and can be brought on by both dry and wet processes [209]. In the wet process, a non-aqueous solvent is used to submerge the polymer suspension, whereas, in the dry process, it is exposed to a solvent environment [210]. In the early 1960s, Loeb and Sourirajan developed composite membranes by polymer solution utilizing a wet phase inversion approach. Hubadillah et al. (2018) produced the first kaolin clay ceramic membrane using phase inversion and sintering. The ceramic membranes developed were flat sheets with different concentrations of kaolin/PES ratios and sintering temperatures of the slurry mixture and others. This method produces as-symmetrical configurations with a sponge or finger-like pores, which can be accomplished using sintering and phase inversion techniques [211]. However, these techniques had limitations in terms of mechanical efficiency and strength. The focus of the majority of current research has been on producing an affordable ceramic membrane with hollow fibers.

9. Future research trends

To render membrane processes an economically acceptable and practical option, further research and development areas of membrane-based technologies are needed in future work, especially on economic analysis, large-scale processes, performance optimization, and sustainability. NF and RO approaches have emerged as the most effective way to remove numerous micropollutants from the wide range of traditional procedures. Hybrid technologies could be more appropriate than conventional approaches. The recent hybrid of MBR and forward osmosis approaches have gained much traction in the current environment when used with techniques like AOPs. Compared to other traditional water treatment technologies, it is understood that the MBR is relatively successful at achieving removal efficiencies despite several intrinsic constraints. The primary trends in this area are discussed, with suggestions for future advancements and improvements to the existing situation, knowledge gaps, and future orientations.

Therefore, further work is required to develop treatment methods for their detection, toxicity, degradation, and separation. An in-depth investigation of the polymer structure, conductivity, and membrane resistance after prolonged operation, cleaning, and membrane reuse requires comprehensive research. Understanding the intricate relationships between the composition of the membrane material, ions, and foulants need computational pieces of evidence to prove the mechanism. Another intriguing technique that hasn't been thoroughly investigated is forward osmosis. Compared to RO and NF, forward osmosis uses much less energy to accomplish the same efficiency. Therefore, future research should increase efficiency by creating newer membranes and incorporating unique hybrid technologies, such as AOP and other approaches.

10. Conclusion

In conclusion, this study has shown the use and effectiveness of many membrane-based technologies in the treatment of wastewater (contaminated with hazardous micropollutants) from various sources. Considering the problematic characteristics of wastewater, several membrane-based technologies can be incorporated into a wastewater treatment process or coupled with other systems to increase the efficiency of water treatment. Various combinations of membrane techniques for the integrated approach have been investigated with multiple goals, including zero liquid discharge, resource recovery, process improvement, and fouling mitigation.

Abbreviations

MBR	membrane bioreactor
AOPs	advanced oxidation processes
WWTP	wastewater treatment facilities
SRT	sludge retention time
NF	nanofiltration
FO	forward osmosis
MF	microfiltration
UF	ultrafiltration
MPs	micropollutants
PEMFCs	proton exchange membrane fuel cells
NC	nanocomposite
TFC	thin-film composite
TFN	thin-film nanocomposite
AuNPs	gold nanoparticles
DBPs	disinfection byproducts
EDCs	endocrine disrupting compounds
PRO	forward & pressure-retarded osmosis
CNTs	carbon nanotubes
TMC	trimesoyl chloride
MPD	<i>m</i> -phenylenediamine
ZIF 8	metal oxide frameworks
COFs	organic frameworks
PES	polyethersulfone
NIPS	non-solvent-induced phase separation
PVDF PAA	polyvinylidene fluoride-polyacrylic acid
PP	polypropylene
SMA	poly(styrene- <i>alt</i> -maleic anhydride)
AeMBR	aerobic membrane bioreactor
PMR	photocatalytic membrane reactor
SARS-CoV-1	severe acute respiratory syndrome coronavirus 1
H1N1	a subtype of influenza A virus
MPM	microporous membranes
DCMD	direct contact membrane distillation
GO	graphene oxide
PVC	vinyl polychloride
SS	suspended solids
TDS	total dissolved solids

CRediT authorship contribution statement

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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.

Data availability

No data was used for the research described in the article.

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The authors certify that the submission is not under consideration for publication elsewhere.

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