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Recent developments in ultrafiltration membrane technology for the removal of potentially toxic elements, and enhanced antifouling performance: A review

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ABSTRACT

The presence of high concentrations of heavy metals (potentially toxic elements) in water bodies generates serious environmental issues and health problems that led to a dramatic increase in wastewater treatment costs. Therefore, it is imperative to develop low-cost and efficient technologies for removing these contaminants from wastewater. Membrane processes are advanced techniques for water treatment and ultrafiltration membrane (UF) has some advantages over other membranes as it requires low pressure to perform However, the UF membrane separates contaminants mainly by size exclusion mechanism, resulting in poor decontamination performance for potentially toxic elements and high susceptibility to membrane fouling. The performance of the UF membrane is often affected by organic and biofouling. Hence, researchers are still looking forward to developing new types of UF membranes that will make them more effective for potentially toxic elements removal and overcome the obstacle of fouling. This review aims to provide an overview of the application of UF membranes in potentially toxic element removal through bibliometric analysis and literature review. The incorporation of various organic, inorganic, carbon-based, and composite-based nano-materials into polymers such as zeolites, metal–organic frameworks (MOFs), and graphene oxide (GO) gave encouraging results for the removal of potentially toxic elements from water. Moreover, this review discusses the mechanisms of fouling in UF membranes and how different techniques can be used to control it. In the end, all the materials are evaluated and compared based on their efficiency, toxicity, simplicity in preparation, popularity, and cost efficiency to provide an overall critical view of the work done in this area. Also, the major challenges and limitations of the use of UF membranes are provided which will help to set the direction of future research.

1. Introduction

Clean water production is becoming a critical issue due to the high rates of water pollution, mismanagement of water resources, and their threatening negative effects on human health (Youcai, 2018). There are various methods used in different water treatment industries to treat water depending on the water quality entering the plant and the required output standards (Wasim et al., 2017; Hubadillah et al., 2017). The typical water purification unit consists of

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multiple stages, each of which must be performed in order. These include pretreatment, pH adjustment, coagulation and flocculation, sedimentation, filtration, and disinfection (Zheng et al., 2018).

There are several types of water purification techniques used such as electrochemical precipitation, complexation, membrane filtration, ion exchange, and reduction (Simon et al., 2013). The most popular method of treatment is membrane technology because it is effective in removing most of the contaminants from water and is also flexible in design and easy to operate (Lakherwa, 2014). In addition, membrane filtration processes have exceptional separation potentials for achieving many of the existing water standards (Zheng et al., 2014). The membrane technology is very beneficial due to its modular nature as it can be applied at both larger and smaller scales, and due to the comparatively little footprint, the better-treated water quality, and lower energy usage (Judd and Jefferson, 2003). With the increase in water demands and constant improvements in membrane performance, energy demand, and cost, the near future of the water industry will witness a continuous expansion in membrane application (Judd and Jefferson, 2003).

1.1. Membrane technology, types, and uses

Typically, the pore sizes of the membranes range from finely porous to non-porous which can eliminate pollutants like protozoa, bacteria, and even multivalent, divalent ions. Based on the pore sizes, the membranes are divided into four major types i.e., microfiltration (MF), ultrafiltration (UF), nanofiltration, and reverse osmosis. These four membrane processes exhibit varying separation ranges. MF and UF are categorized as low-pressure processes that effectively eliminate suspended solids, colloids, and microorganisms. Whereas nanofiltration and reverse osmosis (RO) are high-pressure processes. Nanofiltration is considered a young membrane process that effectively removes magnesium and calcium to achieve water softening and eliminate some simple organic compounds. The application of RO is well known for seawater desalination and desalting of brackish water and removing either natural or synthetic organic compounds of low molecular weight from water (Nunes and Pienemann, 2001).

UF membrane, a low-pressure-driven membrane process, has been extensively used in different industrial applications and water treatment processes Drioli et al. (2017) due to (1) its ability to produce high-quality effluent, (2) its cost-effectiveness in terms of capital cost and operation cost which makes it suitable for large scale application and, (3) its high potential for macromolecule removal, lower footprint, and low energy consumption (Wenten et al., 2020). UF membranes have a pore size of 0.005–0.1 microns and can remove impurities at a low operating pressure of 1–2 bar (Li et al., 2018; AiCHE, 2013). UF membranes can eliminate pathogenic microorganisms and some viruses, allow the passage of most ionic inorganic matter, and retain ionic and particulate matter (Drioli and Macedonio, 2008). Moreover, the UF membrane can remove multiple water-soluble organic matter and microorganisms through a single process. Thus, it displays a high potential to alternate the conventional treatment chain (Peter-Varbanets et al., 2009). Traditional water treatment plants usually include several processes such as media filtration, coagulation, flocculation, flotation, and adsorption; however, the application of UF in the water treatment process can simplify it and replace multiple steps of the process due to the advantages mentioned earlier. UF has been extensively implemented previously to treat and reuse secondary effluent from wastewater treatment plants (WWTPs) (Huang et al., 2012a; Zheng et al., 2011). Overall, the UF membrane technology is mostly used to treat drinking water (60%), and then for extensive industrial water treatment (18%), and wastewater treatment (15%) (Figure S1). However, the lack of high-performance UF membranes is still a long-term challenge, although the past decades have witnessed dramatic advances in membrane materials and technologies.

Recently, there are various review articles published in a similar field such as (Samavati et al., 2022; Damiri et al., 2022; Qalyoubi et al., 2021). Samavati et al. (2022) discuss the removal of heavy metals by nanofiltration and Damiri et al. (2022) reviewed the development of nanocomposite adsorptive membranes for heavy metals, and Qalyoubi et al. (2021) discussed the recent progress and challenges of adsorptive membranes for different pollutants. However, there is no recent review that focuses on both the toxic metal rejection and anti-fouling strategies for UF membranes together. In this review, the organic, and inorganic materials as well as their composites used for the improvement of UF performance are reviewed and discussed in detail. Based on that discussion, the comparison between different materials through set criteria was done to determine the most important materials for future research and implementation. Furthermore, the major challenges and limitations are combined together in this review to set the future direction of research in the field of UF membrane development and their applications.

2. Bibliometric analysis

The bibliometric analysis was carried out to investigate the recent research trends on UF membranes. The main areas of research targeted were the removal of heavy metals (potentially toxic elements), antibacterial application by UF membranes, and UF membrane fouling by natural organic matter (NOM). The major data source for this bibliometric analysis was the Web of Science (WOS) database to extract as many related articles as possible. Besides, manual searches and checks were performed to avoid any missing articles. The data relating to the removal of potentially toxic elements by UF was extracted using the terms "UF", and "heavy metal", while the data relating to the UF membrane fouling was obtained using the terms "UF", and "fouling". These terms were selected based on the preliminary analysis as they provided the most relevant results. Also, an additional search was carried out to ensure no related article is missed from the bibliometric analysis.

The total count of publications comprising research papers and review papers related to the removal of potentially toxic elements was 787, the UF biofouling was 1130, and the related to UF membrane fouling by NOM was 1361. Fig. 1a, b, and c are constructed to show the number of articles published each year related to the UF applications for the removal of potentially toxic elements (Fig. 1a), and related to the improvement in anti-biofouling (Fig. 1b), and anti-organic (NOM) fouling (Fig. 1c) performance. It is evident from these Figures that UF membrane technology has gained huge attention recently from the continuous growth in the number of publications over time. Therefore, during the past ten years (2012–2022), the number of articles published related to heavy metal removal accounts for 63% which is more than half of the total publications found (Fig. 1a). Moreover, the R^2 value of 0.942 showed that there is a linear increase in the number of publications each year and it is expected to continue considering the problems related to water shortages and wastewater discharge.

Similarly, Figs. 1b and 1c shows a rapid increase in the research related to the improvement in the anti-fouling performance of UF membranes over the last few decades. As seen in Fig. 1, the number of published papers has dramatically increased from 1 paper in 1982 to 1206 in 2022. During the past 10 years (2012–2022), the number of publications accounts for over 55% of the total publications. The elevation in publications number started in 2007 which means in that period this topic started to gain more attention than before, and the total number of publications starting from that period accounted for 83% of the total percentage of publications. The R^2 value of 0.943 (Fig. 1c) showed that there is a linear increase in the number of publications each year which is expected to continue in the upcoming years.

3. Advanced and novel UF membranes for potentially toxic elements removal

Potentially toxic elements also referred to as heavy metals or trace elements, are elements with atomic density higher than 6 g/cm^3, including mercury (Hg), chromium (Cr), nickel (Ni), copper (Cu), cadmium (Cd), cobalt (Co), and arsenic (As) (Li et al., 2016). The presence of high concentrations of potentially toxic elements in water bodies generates serious environmental issues and health problems and leads to a dramatic increase in wastewater treatment costs (Fernandez and Olalla, 2000; Ogoyi et al., 2011). The occurrence of potentially toxic elements in the environment could be naturally by rocks leaching, forest fires, and airborne dust. However, human activities such as agricultural, industrial, and domestic applications, directly and indirectly, influence the presence and accumulation of these metals in waterbodies (Gardea-Torresdey et al., 2005). Potentially toxic elements are naturally highly soluble, and non-degradable, so they are abundant in wastewater (Jaishankar et al., 2014). To protect the environment, most countries have enacted regulations that limit the discharge of effluents containing potentially toxic elements. For example, in Canada, the Canadian Metal Mining Effluent Regulations set limits for metals' concentration in effluents to be <1 mg/L Moreover, the European Community applied strict regulations on potable water in which the maximum nickel concentration accepted in potable water is 50 μg/L (Danis and Aydiner, 2009). Therefore, it is imperative to develop low-cost and efficient technologies for removing potentially toxic elements from wastewater.

Adsorption and chemical precipitation are the most common and widely used methods to eliminate potentially toxic elements from wastewater. However, these methods are low-selective processes and are very expensive (Rana et al., 2014). Membrane processes are advanced techniques and relatively cheaper and viable solutions in the treatment of wastewater that contains metals since membranes can be inserted as a retrofit of existing plants (Chen et al., 2014). Nanofiltration (NF) and Reverse osmosis (RO) are effective processes for potentially toxic elements removal from aqueous solution; however, more energy consumption and higher operating pressure are the downfalls, particularly for treating large volumes of wastewater with low levels of potentially toxic elements (Yang et al., 2007).

UF has some advantages over other membranes as it requires low pressure to perform. The UF membrane, however, separates contaminants by size exclusion mechanism, resulting in poor decontamination performance for potentially toxic elements. Moreover, the large volume of sludge, high energy demand, lack of selectivity, and incomplete removal of contaminants may occur when a large portion of water is treated. (Landaburu-Aguirre et al., 2009). Therefore, researchers are still looking for developing new types of UF membranes that will make them more effective for potentially toxic elements removal.

Several researchers have developed advanced and novel UF membranes for better removal of potentially toxic elements. Some studies have reported the use of various organic, inorganic, carbon-based, and composite-based nanomaterials (Table 1) such as zeolites, metal–organic frameworks (MOFs), graphene oxide (GO), and Fe–Mn binary oxides (FMBO) which could be incorporated into polymeric matrixes to form new types of membranes (Ghaemi et al., 2015). These types of membranes also called mixed matrix membranes (MMM's) have shown better metal removal capacities due to the beneficial effects offered by the additives such as high surface area, and high selectivity (Abdulkarim et al., 2021). Zeolite has a remarkable benefit as it has hydrophilic properties that enhance the water permeation properties of MMM's. As an ion-exchange material, zeolite contains cations like potassium, calcium, and sodium that can be exchanged with other metal cations present in solutions such as zinc, manganese, and cadmium (Xiao et al., 2021). Zeolites are also popular as an essential microporous material composed of $[SiO_4]_4-$ and $[AlO_4]_5-$ tetrahedral to create an open system of channels and pores (Al-Jubouri et al., 2018). Therefore, this porous system consists of easily exchangeable cations which are important to perform the separation process by ion exchange or adsorption and other catalysis processes (Al-Jubouri, 2020). A study conducted by Alfalahy and Al-Jubouri (2022) demonstrated the efficiency of zeolite in the removal of lead from aqueous solutions as described in Fig. 2. They used hydrothermal synthesis to prepare and incorporate NaX zeolite

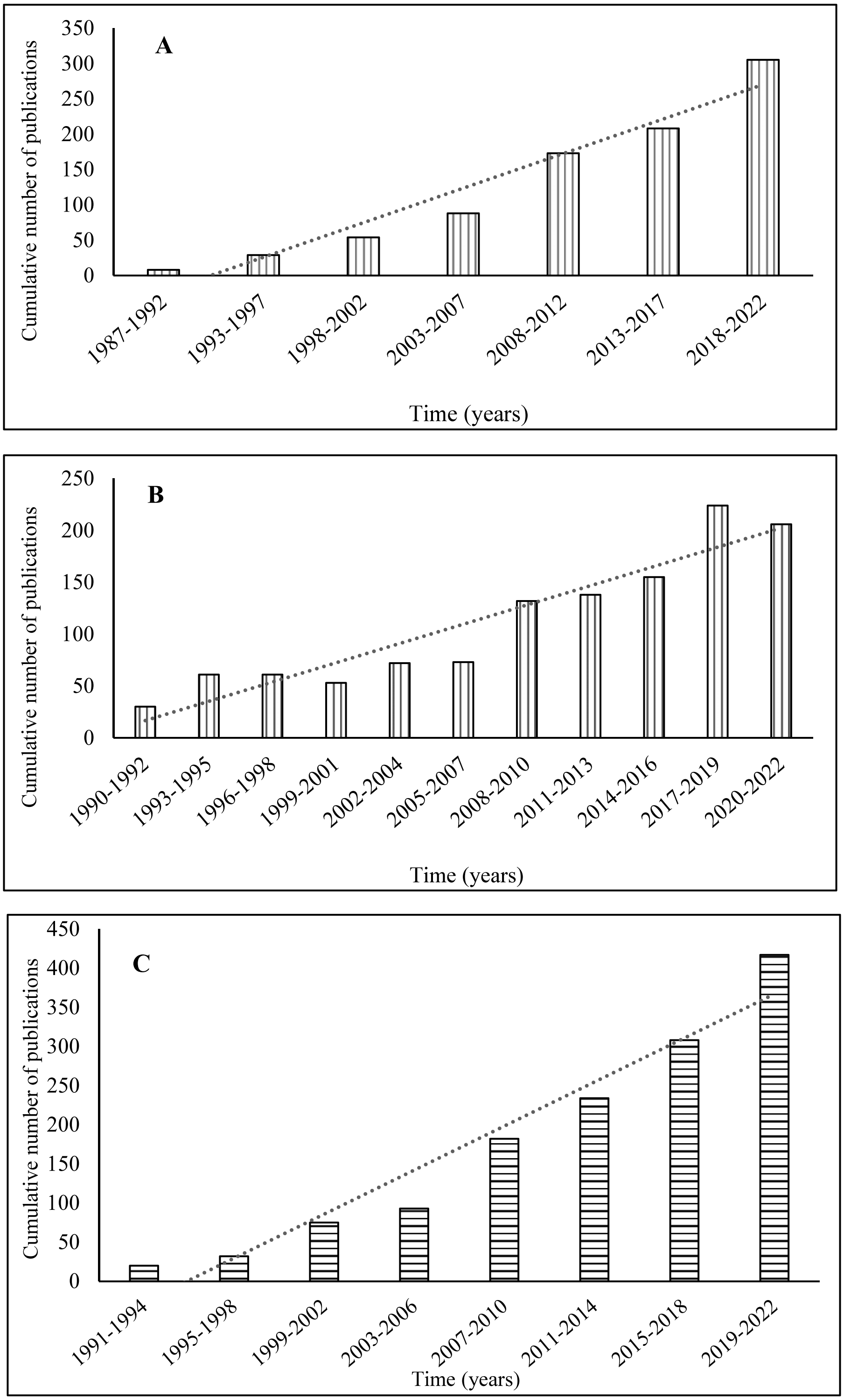


Fig. 1. (A) Number of papers published each year related to the removal of potentially toxic elements (heavy metals) by UF membranes. **(B)** Number of papers published each year related to the anti-biofouling by UF membranes. **(C)** Number of papers published each year related to UF membrane fouling by NOM.

Table 1

Summary of studies related to the development of UF membranes for heavy metal removal.

No.	Technique used	Metals studied	% Removal achieved	Adsorption capacity (mg/g)	Remarks	References
1	Clay-based UF membrane supported on natural zeolite	Chromium	Membrane with six layers Sm/Z6 = 89	–	UF composite membranes were obtained via layer-by-layer technique and four different membranes were prepared namely, Sm/Z4, Sm/Z5, Sm/Z6, Sm/Z7	Aloulou et al. (2020)
2	Extracellular polymer substances-enhanced UF (EPS-UF)	Lead Copper Cadmium	94.8 88.9 89.2	–	EPS solution formed cake on the UF membrane which was then used to filter contaminated water	Cao et al. (2020)
3	Polyelectrolyte-enhanced UF followed by dithionite-based chemical reduction	Copper	94	–	Polyethylenimine (PEI) achieved the highest removal at pH 3 and MWCO= 60 KDa	Chou et al. (2018)
4	Internal pore decoration with polydopamine nanoparticles in polymeric UF membrane	Lead Cadmium Copper	92.2	20.24 17.01 10.42	Dopamine solution penetrated PES/UF membrane from the reverse direction (PES/PDA-R) and exhibited favorable adsorption performance	Fang et al. (2017)
5	A mixed matrix membrane of UF impregnated with GO	Lead Copper Cadmium Chromium	90–96	79 75 68 154	The best adsorption capacity was observed at pH 10	Mukherjee et al. (2016)
6	Polyethersulfone/hydrous manganese dioxide UF mixed matrix membrane	Lead	99	204.1	Complete removal was achieved at pH 8	Gohari et al. (2013)
7	Bis-aminosilane cross-linked multiwall carbon nanotube UF membrane (Surface modification)	Lead Nickel Copper Zinc	89.53 90.42 91.43 91.86	–	The membrane was prepared by incorporating cross-linked quaternary ammonium carbon nanotube (CQACNT) nanomaterial into a poly-ether sulfone polymer matrix	Zheng et al. (2018)
8	PES membrane incorporated with nickel-bentonite nanoparticles	Zinc Copper Lead	98.62 97.88 97.03	–	Membrane with 0.5 wt.% NBNPs/PES exhibited the maximum metal rejection	Dadari et al. (2022)
9	UiO-66 incorporated UF membrane	Strontium Lead Cadmium Chromium	>93%	–	Membranes with MOF-0.6 wt.% possessed the maximum potentially toxic elements removal.	Wang et al. (2022)
10	α-zirconium-phosphate-nanoparticle/polyacrylonitrile (α-ZrP-NP/PAN) mixed matrix membranes (MMMs)	Lead	94.82	–	α-ZrP-NP contributed to enhancing the membrane's hydrophilicity and negativity in pH 3–10	Guo et al. (2022)
11	PES membrane decorated by Mil-125(Ti)/chitosan nanocomposite	Lead	95	–	1% wt% nanocomposite was the optimum load that led to increasing water flux and reduced contact angle and high separation efficiency	Khosravi et al. (2022)
12	Polysulfone amine-functionalized nanocomposite membrane	Copper Lead Nickle Cadmium	99.7 98.6 98.4 98.5	–	Organic/inorganic 3D nanonetwork was formed by intercalating amino group-functionalized carbon nanotubes and sodium styrene-maleic anhydride copolymer.	Dong et al. (2022)
13	Sulfonated polyethersulfone self-assembled with Amine functionalized GO/MnO_2 nanohybrid	Nickle Copper Zinc	81.1 64 67.4	–	The loading of 4wt% of GO/MnO_2 achieved the optimum metal removal and enhanced the flux by 118%.	Ibrahim et al. (2020)
14	Amine-functionalized MCM-41 modified UF membrane	Chromium Copper	86.8 87.1	2.8 3.7	The porous nanoparticles formed a uniform and hydrophilic thin film on the membrane's surface that increased its affinity to heavy metals.	Bao et al. (2015)

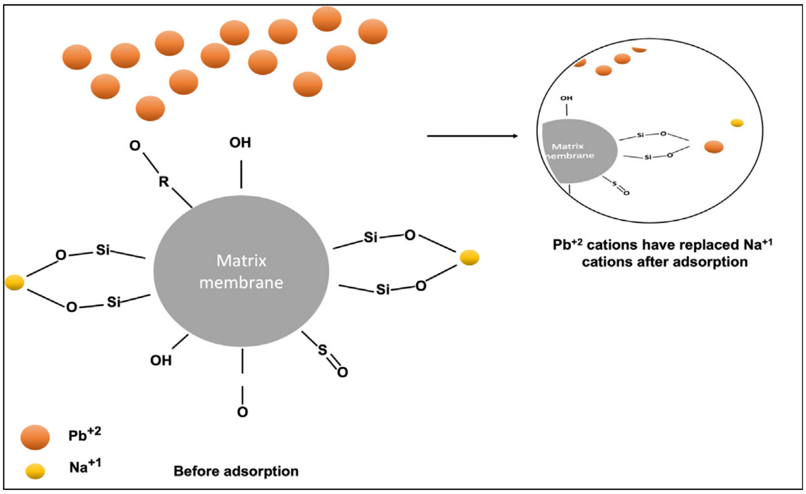


Fig. 2. Mechanism of zeolite adsorption embedded in PES membranes for Pb (II) ions removal. Zeolite contains cations such as sodium that are exchangeable with other positive cations which are Pb (II) in this case (Alfalahy and Al-Jubouri, 2022).

crystals into polyethersulfone membranes through phase inversion methods. The membranes showed excellent results in removing lead from aqueous solutions containing 0.9% weight per volume of NaX zeolite at the studied experimental conditions (pH 6, pressure 1.6 bar, temperature 25 degreeC, and initial lead concentration 50 ppm).

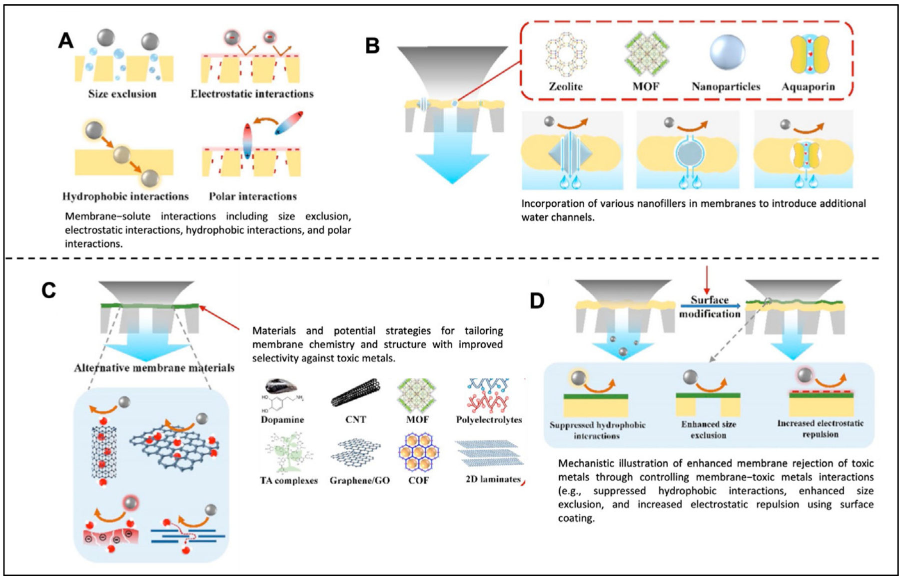
Similarly, Alawady et al. (2020) incorporated multiwall carbon nanotubes (CNTs) into the chitosan (CHIT) biopolymer matrix to enhance the separation performance and water permeability of an ion-selective membrane. The researchers functionalized carboxylic acid groups into multiwall carbon nanotubes (CNTS-COOH) to form a thin layer on top of polysulfone (PS), which served as a selective wall for the formation of COOH/CHIT/PS membranes. The researchers found that these membranes had enhanced metal ion rejection properties, particularly in terms of copper, nickel, cadmium, and lead ions. The results showed 99% rejection for all investigated metal ions at pH 10 and concluded that the incorporation of carboxylic acid groups causes polymer chains to extend significantly further and expose more to adsorption sites to bind with metal ions.

The researchers also used graphene oxide extensively to develop novel and advanced MMM's. Kaleekkal et al. (2017) synthesized carboxylated graphene oxide (c-GO), rich in oxygen-containing functional groups, from pristine graphite. The researchers blended polyethyleneimine polymer into the solution, which binds to metal ions in the feed solution and increases the molecular weight of Ni^{2+} and Cd^{2+} ions to increase binding efficiency. At pH 7, they were able to remove >90% of potentially toxic elements using this technique, and higher binding efficiency was recognized at a 5% loading ratio for c-GO. The efficiency of different membranes incorporated with inorganic fillers is shown in Figure S2.

In a study conducted by Fang et al. (2017), the authors designed a new adsorptive UF membrane for potentially toxic elements removal. The membranes were made by self-polymerizing dopamine solution through UF membranes. The membranes, polydopamine nanoparticles on polymeric UF membrane labeled as PES/PDA-R, were prepared by the penetration of self-polymerized dopamine solution through UF membranes from the reverse direction (R). Another type of membrane, labeled as PES/PDA-F, was prepared by forwarding filtration (F) to compare them with each other. The adsorption capacities for Pb, Cd, and Cu on PES/PDA-R membranes were 2.23, 17.01, and 10.42 mg/g respectively which are 1.69, 2.25, and 1.91 times greater than that of the PES/PDA-F membranes.

Wang et al. (2022) fabricated a UF membrane using UiO-66 polysulfone as filler for heavy metal removal. The separation performance of the fabricated membrane was compared to pure PSF membranes, and results revealed that the fabricated membrane has higher mechanical strength with an 88% flux recovery ratio compared to the blank membrane, which was 34%. Furthermore, it was found that when the pH of the solution increases, the rejection rate of the metals studied (Sr^{2+}, Pb^{2+}, Cd^{2+}, Cr^{6+}) increases. High metal rejection properties showed the potential of UiO-66 incorporated membranes for use in water and wastewater treatment. Table 1 lists some of the successful membrane modifications to remove potentially toxic elements from aqueous solutions. Fig. 3 shows the schematic diagram of the types of interactions that occurs between membranes and solutes, incorporating nanofiller to enhance the water permeability and the removal of toxic metals, potential modification for membrane tailoring to enhance selectivity and alternate their structure and chemistry, and rejection mechanisms of toxic metals by the membrane (modified from Guo et al., 2022).

The advantages and disadvantages of different types of materials are summarized in Table 2. It is evident that each material whether organic or inorganic or a composite has its own pros and cons. While composite materials have the superior advantage of combining the properties and functionalities of both organic and inorganic materials, their preparation procedures are often complex and costly. On the other hand, carbon-based nanomaterials like graphene oxide and carbon nanotubes have demonstrated excellent performance in the removal of toxic elements from water, but their preparation, and application at a larger scale are not yet tested.



Membrane–solute interactions including size exclusion, electrostatic interactions, hydrophobic interactions, and polar interactions.

Incorporation of various nanofillers in membranes to introduce additional water channels.

Materials and potential strategies for tailoring membrane chemistry and structure with improved selectivity against toxic metals.

Mechanistic illustration of enhanced membrane rejection of toxic metals through controlling membrane–toxic metals interactions (e.g., suppressed hydrophobic interactions, enhanced size exclusion, and increased electrostatic repulsion using surface coating.

Fig. 3. **(A)** Schematic diagram of the types of interactions that occurs between membranes and solutes. **(B)** Incorporating nanofiller to enhance the water permeability and the removal of toxic metals. **(C)** Potential modification for membrane tailoring to enhance selectivity and alternate their structure and chemistry **(D)** Rejection mechanisms of toxic metals by the membrane.
Source: Modified from Guo et al. (2022).

Table 2
Advantages and disadvantages of the four types of nanomaterials used for surface modification of polymeric UF membranes.

Material type	Examples	Advantages	Disadvantages	References
Inorganic fillers	Include different metal and metal oxide nanomaterials such as Ag, Au, Al, Fe, $MgAl_2O_4$ TiO_2 Fe_2O_3 SiO_2 Fe_3O_4	Large surface area, controllable structures, diverse surface chemistry, and unique optical and physical properties	High possibility of leaching and high load is required to achieve desired properties for the membrane can agglomerates which makes it difficult to achieve uniform dispersion. Unknown toxicity toward the environment	Yu et al. (2022), Moslehyani et al. (2015), Gohari et al. (2013)
Organic based nanomaterials	Any organic materials excluding carbon materials such as dendrimers, cyclodextrin, liposome and micelle chitosan N-halamine compounds, polymers biomolecules	This type of membrane is preferred over the inorganics as they have more functional groups, which makes them more adaptable and capable to attach cations and small-molecule organics to the substrate through molecular interactions.	Poor compatibility with hydrophobic polymer matrix and low thermal stability	Yu et al. (2022), Al Aani et al. (2020), Oliveira et al. (2018)
Carbon-based nanomaterials	Graphene, single-walled carbon nanotube, multiwalled carbon nanotube, carbon fiber, activated carbon	These materials have a large specific surface area, strong chemical stability in acid/alkaline conditions, enhanced mechanical and thermal stability, and high porosity	Minimal functionalization and inadequate dispersibility in aqueous conditions, leading to low removal	Yu et al. (2022), Awad et al. (2021)
Composite-based nanomaterials	Any combination of metal-based metal-oxide based, carbon-based, and organic-based nanomaterials and have complicated structures like MOF	Combines the basic properties of organic and inorganic materials and creates functional material with desired properties while offering specific advantages to meet the harsh requirements in water and wastewater treatment processes.	The preparation method with this type of nanomaterials is not simple	Damiri et al. (2022), Yu et al. (2022)

4. UF membrane fouling

Membrane fouling is a process in which the particles and substances present in feedwater get deposited on the surface of the membrane, partially or completely blocking the membrane pores. This results in the decline of permeate flux and salt rejection of the membranes, requiring frequent cleaning or replacements of the membranes (Ashfaq et al., 2022).

Adsorption and accumulation of material on the surface of a membrane and through its pores are consequences of mass transport, in which chemical and physical interactions occur between the components of the membrane surface and those of feed water (Guo et al., 2012).

Generally, membrane fouling can be classified as reversible or irreversible, depending on the system's operating conditions and how it gets cleaned (Chang et al., 2012). Reversible fouling happens at the membrane rejection surface, and this type of fouling can be restored through physical washing if the membrane is back washable, and if not, fouling can be eliminated by chemical cleaning. However, irreversible fouling can happen by pore plugging and chemisorption mechanisms. To restore the membrane's TMP, the membrane either gets cleaned extensively by chemicals or gets replaced (Bennett, 2005).

In the context of this review, all of which relate to UF fouling control, the rapid decline of permeate flux resulting from membrane fouling is considered a key problem in UF membrane application in water treatment. The reduction of membrane flux happens mainly due to two main reasons; the first one is concentration polarization which is a reversible phenomenon that does not impact the membrane's characteristics. These molecules that accumulated near the membrane's surface are hardly reversible in the bulk solution, and their concentration might be 50 times higher than their concentration in the bulk solution, resulting in a decrease in the solvent flow through the membrane and reducing its membrane's permeability (Humbert et al., 2007). Secondly, fouling in UF could be a consequence of the deposition of solutes from feed solution on the porous membrane structure (also called internal fouling) and membrane's surface (also named external fouling) (Zydney, 1997).

4.1. Mechanisms of UF membrane fouling

The mechanisms of membrane fouling in the case of UF membranes are mainly pore blocking, gel formation, cake formation, and adsorption. However, more than one fouling mechanism can be involved at the same time (Humbert et al., 2007). Fouling resulting from pore-blocking occurs when the particles and colloids settle on the membrane surface, fully or partially blocking the membrane's pores. This mechanism is dominant during the initial stages of the filtration process when the particles are in direct contact with the pores (Hermia, 1982). The second fouling mechanism that may occur on the UF membrane is a gel-layer formation which is a dense layer of solutes significantly blocking the membrane pores and reducing permeate flux. Cake formation is a third possible fouling mechanism, described as an accumulation of particles layer by layer on the membrane's outer surface, which prevents fluid flow through it (Hughes et al., 2006). Fouling reversibility is determined by the interaction between the membrane surface and cake layer, while its morphology dictates the flux decline. Fouling resulting from cake formation may lead to over-clogging and become more irreversible when the interstices of the formed cake are filled with small macromolecules that give rise to more significant hydraulic resistance.

4.2. Foulants responsible for UF membrane fouling

To control UF membrane fouling, it is imperative to identify the foulants responsible for membrane fouling in UF membranes. The foulants that are mainly responsible for UF membrane fouling are divided into four categories i.e., biological substances, macromolecules, particulates, and ions. Among these, biological substances and natural organic matter (NOM) are the most common foulants that have severely impacted the performance of UF membranes (Humbert et al., 2007). Macromolecule foulants are characterized by the presence of functional groups and a molecular weight ranging from a few thousand to one million daltons therefore, these characteristics determine their interaction with the surface of the membrane. While particulate foulants have rigid shapes and their sizes range from 1 nm to 1 μm. Depending on the size of these particulates, they may partially or completely clog the membrane pores and form a cake layer (Shi et al., 2014). Moreover, UF membrane systems are often get affected by protein foulants, especially in food and therapeutical industries. Protein adsorption on the surface of the membrane causes it to become thick and opaque, resulting in irreversible fouling (Bacchin and Aimar, 2006).

Bio-fouling or microbiological fouling is one of the biggest challenges that is faced by UF membranes. The process of biofouling is initiated by the adherence of cells to the surface of the membrane. Afterward, these cells produce exo-polymers (extracellular polysaccharide substances) that bind with the membrane and make it difficult to remove. After days of cell attachment, these cells form multiple layers of microcolonies called mature biofilms (Ashfaq et al., 2022; Shahkaramipour et al., 2017). In general, three methods of biofouling control are used: physical and chemical washing after a specific time of membrane performance, injection of a biocide into the system, and development of biofouling resistance membranes (Cheng et al., 2018). Although chemical and hydraulic cleaning can restore the membrane flux, the membrane is also exposed to damage through this step, and filtration rates could get severely affected and drop throughout cleaning. Moreover, the UF membrane is also exposed to breakage during aeration and backwashing by the shearing forces of water as well as requiring large input of energy and labor as well as toxic by-products that threatens the environment (Guo and Liu, 2012). Furthermore, it was found by Wang and Lin (2017) that the substances used through back-washings, such as acid and alkaline substances, oxidants, and contaminants in wastewater, can influence the membrane performance and decreases its lifespan. Therefore, modifying the membrane to deliver superior performance, such as operating high fluxes with low fouling and high rejection rates, which decreases energy consumption, becomes a goal. Various properties could be modified to enhance the antifouling performance of the membrane, such as decreasing the membrane's surface roughness, altering the surface charge, and increasing the hydrophilicity of the membrane (Cheng et al., 2018; Ji et al., 2020). Membrane hydrophilicity can be improved by using various methods such as coating or grafting the hydrophilic polymers, oligomers, and nanoparticles (Wenten et al., 2020; Younas et al., 2019; Plisko et al., 2018). Modification of the membrane surface is considered an effective technique and results in enhanced antifouling performance and better separation performance due to the location of the agent on the selective layer surface and pore walls and the possibility of the modification agent contacting the feed solution directly.

4.3. Techniques to control UF membrane fouling

There are various techniques used to control membrane fouling in UF membranes. These include membrane surface modification, changes in operating conditions, development of membrane modules, and spacers, application of pre-treatment techniques, and other techniques like in-situ fouling characterization techniques and membrane cleaning methods. In this review, techniques used for membrane surface modification are discussed in detail, while an overview of the other methods is also provided. For further details about these methods, additional information can be found in other recent reviews (Ilyas and Vankelecom, 2023; Peters et al., 2021).

4.3.1. Effect of operating conditions

The operating conditions especially crossflow velocity, permeate flux, and transmembrane pressure play a significantly important role in membrane fouling mitigation as these parameters determine the hydrodynamic conditions of the feed flow and drag, and lift forces acting on the foulants near the membrane surface. When the system is operated at a high permeate flux, the drag forces acting on the foulants bring more foulants toward the membrane surface causing more severe membrane fouling (Peters et al., 2021). Therefore, it is generally recommended to measure the critical flux of the membrane and operate the system below the critical flux (Bacchin and Aimar, 2006). Moreover, high transmembrane pressure also causes compaction of the gel layer/fouling layer on the membrane resulting in a more severe reduction in permeate flux and pores blockage (Taheri et al., 2019; Sioutopoulos et al., 2019). On the other hand, crossflow velocities act differently because high crossflow velocities cause more turbulence in the feedwater flow and enhance lift forces which do not allow foulants to settle down on the membrane surface and also detached the settled foulants from the surface (Du et al., 2017) (Table 3). Therefore, high crossflow velocities are considered favorable for fouling control. However, some other researchers have also shown that the high crossflow velocities can cause the breakage of bacterial flocs, or the breakage of microbial cells causing the release of polysaccharides and other organics which ultimately intensify the membrane fouling (Liu et al., 2014). Therefore, the composition of feedwater is one of the important factors before determining the operating conditions for UF systems (Peters et al., 2021).

4.3.2. Membrane module and spacers design and development

A module is used to physically seal and isolate the feed and permeate stream. The most used module designs for UF membranes are hollow fiber modules (HFMs), and flat-sheet plate and frame modules (PFMs). Researchers have studied the effect of novel spacers with different designs, angles, shapes, and types (non-woven or woven) on membrane fouling since the spacers not only provide mechanical strength to the membrane but also influence hydrodynamics of the feed flow (Ilyas and Vankelecom, 2023). Hence, the novel spacers are designed to enhance flow turbulence resulting in less pressure drop (or dead zones) and less accumulation of foulants on the surface. For example, vibrating spacers and turbo-spacers were developed to improve the performance of UF membranes and reduce their fouling propensity. In the PFMs, the turbo-spacers helped to improve the flow turbulence by using the kinetic energy of the feed flow. The experimental results showed less accumulation on membranes with turbo-spacers as compared to the conventional spacers because the conventional ones had very less impact on flow unsteadiness (Ali et al., 2020). Similarly, another research reported the development of 3D printed spacers based on triply periodic minimal surfaces (TPMS). TPMS are without self-intersecting and folding surfaces. Such designs helped to reduce the biofouling of membranes and improved the flux by 38% in UF (Sreedhar et al., 2018).

In addition, various turbulence promoters are used to improve the fluid hydrodynamics near the membrane surface. However, most of these turbulence promotion techniques are used for membrane bioreactors (MBRs), and RO. In UF, a turbulence promoter called a spinning basked membrane (SPM) module was used. It was like the rotating disk module which rotates flat sheet membranes around a hollow shaft. The use of the novel turbulence promoter reduced the fouling propensity significantly as only a 5% reduction in flux was noted during the experimental duration (Sarkar et al., 2012).

4.3.3. Pre-treatment techniques

Various pre-treatment techniques are used to develop Hybrid membrane processes (HMP) and Integrated membrane processes (IMP). HMPs/IMPs are used to reduce a load of foulants (organics, inorganics, and bacteria) in the feedwater that will help to control membrane fouling and improve its performance. HMP combines conventional pretreatment techniques (physical, chemical, and biological) with the membrane unit, while IMP integrates two or more than two membrane units. Physical techniques allow the removal of substances by increasing the size of the foulants and sedimentation or by filtration using a microfiltration membrane. Using chemical techniques, the feedwater chemistry is changed to increase

Table 3
Various techniques to control UF membrane fouling.

1	Operating conditions	
	1.1	Higher flux brings more foulants toward the surface causing more severe fouling. Therefore, it is recommended to operate UF systems at constant flux conditions instead of constant transmembrane pressure (Bacchin and Aimar, 2006; Taheri et al., 2019; Sioutopoulos et al., 2019).
	1.2	Also, researchers have shown that it is important to operate UF systems at low transmembrane pressure to reduce the drag forces which will compact the fouling layer causing a significant reduction in membrane permeability (Voutchkov, 2018).
	1.3	High cross-flow velocities enhance shear forces in the membrane system and cause flow turbulences which allow continuous detachment of foulants settling on the membrane surface (Peters et al., 2021). In addition, other techniques like gas sparging have also been shown to enhance shear forces and reduce UF fouling (Chan et al., 2011)
2	Membrane modules and spacer designs	
	2.1	Various novel spacers were developed such as turbo-spacers and vibrating spacers that cause flow turbulence and discouraged the settling of foulants on the UF membrane (Ali et al., 2020)
	2.2	The novel 3D printed spacers based on TPMS were developed which helped to reduce the biofouling of membranes and improved the flux by 38% in UF (Sreedhar et al., 2018).
	2.3	A spinning basked membrane (SPM) module was developed and used as a turbulence promoter which helped to control membrane fouling (Sarkar et al., 2012).
3	Pre-treatment techniques	
	3.1	Chemical coagulants used to control UF fouling are Aluminum salt (Wang and Wang, 2006), iron salt (Guigui et al., 2002), poly-aluminum chloride (Park et al., 2002), and titanium salt (Huang et al., 2016). The degree of fouling controlled by using these coagulants was found to be from 0.42 to 4.0 at dosages ranging from 2.2 mg/L–80 mg/L. Among these, titanium salt was shown to control the least degree of fouling (i.e., 0.42–1.0) at dosages as high as 10–80 mg/L. While aluminum salt showed the highest degree of fouling control (3.2–4.0) at a dosage of 3.5 mg/L (as Al).
	3.2	Adsorption by activated carbon such as powdered AC (Kang and Choo, 2010), Granular AC (Kim et al., 2009), and biochar (Shankar et al., 2017) has been used. Among these, granular AC demonstrated the highest degree of fouling control (i.e., 3.0–5.5) at a dosage of 1.5 g/L.
	3.3	Chemical oxidation is done using chlorine (Ha et al., 2004), ozone (Song et al., 2010), chlorine dioxide (You and Tsai, 2011), and permanganate (Lin et al., 2012). The highest degree of fouling control was found to be achieved with ozone (1.0–1.7) at concentrations of 0.5–3.0 mg/L.
	3.4	Ion exchange methods such as fluidized ion exchange (Cornelissen et al., 2009) and magnetic ion exchange (Humbert et al., 2007) have been used as pre-treatment techniques to reduce fouling and have been reported to control about 1.0 degrees of fouling.
4	Membrane Cleaning techniques	
	4.1	Physical cleaning techniques involve backwashing, and forward flushing which help to remove the foulants from the membrane and remove the major portion of the irreversible fouling. More than 80% flux recovery has been shown to be achieved using the physical cleaning method (Waterman et al., 2016; Liang et al., 2008).
	4.2	Chemical cleaning is done using acids (like 0.1 M HCl) and base (0.1 M NaOH) for a time duration of 30 min to 24 h. The flux recovery of about 70% has also been achieved using chemical cleaning methods (Lim and Bai, 2003; Lee et al., 2001)
5	In-situ fouling characterization techniques	
	5.1	Optical coherence tomography (OCT) is an image-based non-invasive characterization technique that can be used to monitor the cake layer formation on UF membranes (Liu et al., 2020; Han et al., 2018; Li et al., 2016)
	5.2	The In-situ Raman spectroscopy technique can be used to monitor the quality as well as quantity of membrane fouling. Previous research showed that its use in UF fouling studies helped to predict the onset of fouling even before it was even evident in the flux decline (Tang et al., 2020).
	5.3	Ultrasonic time-domain reflectometry (UTDR) is one of those techniques already in use at the industrial scale. This technique can be adopted for different membrane modules but it can only provide information about the fouling layer thickness and density and not on the composition of the layer (Rudolph et al., 2019).

the foulant's size and reduce its affinity toward the membrane. While biological techniques mainly involve disinfection methods that are used to kill bacteria to reduce biofouling potential. In Table 3, we have discussed the most commonly used pre-treatment techniques for UF membranes, and the degree of fouling controlled by the technologies. The degree of fouling control was estimated from the ratio of the membrane permeability with the fouling control technique to the permeability without it (Cui and Choo, 2014; Peters et al., 2021). Through comparison, it can be deduced that the adsorption by biochar was the most effective pre-treatment technique as the maximum degree of fouling control was estimated to be 5.5. However, it is worth noting that the efficiency of the pre-treatment technique to control fouling strongly depends on the type of technique, the dosage of agents, the composition of feedwater, and membrane surface properties (Gao et al., 2011).

4.3.4. Other techniques

Other methods that aim to mitigate membrane fouling are membrane cleaning techniques and membrane characterization techniques. These techniques do not prevent membrane fouling directly. Instead, cleaning techniques help to remove the fouling layer from the membrane surface so that its performance can be restored. And the novel characterization techniques allow in-situ monitoring of the fouling phenomenon and process so that on-time measures can be adopted, and cleaning procedures can be implemented. These methods are summarized in Table 3.

4.3.5. Novel and advanced uf membranes to prevent membrane fouling

4.3.5.1. Membrane surface modification for biofouling mitigation. It has been found that UF can eliminate 3 nm or smaller substances compared to MF membranes which can only achieve the removal of 50 nm or bigger pollutants. Therefore, the UF membrane can achieve higher bacterial removal and is also a low-cost method compared to other membranes (Vrouwenvelder et al., 2011). However, this makes the UF membrane highly susceptible to biofouling which decreases the permeation rates considerably during operation, limiting the application of UF membranes in the water filtration process. Therefore, the researchers have dedicated their efforts to studying various membrane modifications to increase fouling resistance and enhance the performance of the membrane. Based on the findings of different studies, two membrane techniques could be developed to prevent biofouling. The first method involves the incorporating of additives within the membrane matrix or on its surface for instance nanocomposite structures are also named mix matrix membranes (MMM) (Hu et al., 2019). The MMM has been found to be highly effective in improving the performance of the membranes and a variety of nanoparticles have been used such as Al_2O_3, Cu, SiO_2, and TiO_2 (Mollahosseini et al., 2012). The second method modifies the bulk and surface properties of membranes by changing their chemical structure. This review focuses mainly on the modification of the membrane surface which impacts the membrane's physio-chemical properties.

Surface membrane modification is considered one of the most successful techniques for antibacterial applications; to elevate the membrane bacteriostasis performance, overcome flux retention dilemmas, and reduce bio-fouling issues (Guo et al., 2012). Moslehyani et al. (2015) reported that when an inlet gets in contact with the incorporated membranes' surface, the antibacterial agents release and alter the cell wall of the bacteria, and both disinfection and filtration happen simultaneously. Previous reports have studied the antibacterial rate of different modified UF membranes. For example, Zodrow et al. (2009) studied the effect of silver nanoparticles impregnated with polysulfone UF membrane on removing bacteria. This study showed that this modified membrane is highly effective in removing *E. coli* K12 and *P. mendocina* KR1. Moreover, the modified membrane not only removed bacteria through filtration but prevented the binding of these strains to the membrane surface, which reduced the membrane's biofouling. A disadvantage of such a technique is that nanomaterials could be depleted rapidly during the long-term performance. Therefore, it was highly recommended to explore other methods focusing on the fixation of antimicrobial agents on the membrane's surface to slow the release of these agents.

More advanced methods are found promising in resolving the leaching issue through using an "immobilize agent" on inorganic or organic fillers that are entirely compatible with the polymer matrix. For example, a study conducted by Moslehyani et al. (2015) investigated the effectiveness of silver lactate halloysite nanotube (HNT) clay nano-filler attached to a PVDF polymer matrix as a bacterial separator. The main aim of this study was to fabricate membranes that are resistant to fouling and have high permeation flux. The purpose of using HNTs was to overcome the leaching of antibacterial agents from the membrane's surface, which is silver lactate (SL) in this study. To achieve immobilization, HNT was enhanced by N-β-(aminoethyl)-γ-aminopropyl tri methoxy silane (AEAPTMS). The experimental results illustrated 99% rejection of the two examined bacterial strains used in wastewater i.e., *Salmonella* and *Enterobacter aerogenes*, improvements in permeation flux, and reduction of SL leaching from the membrane's surface.

Geng et al. (2021) fabricated a PVDF UF membrane functionalized with imidazole graphene oxide by one-step grafting (Im-GO as an antibacterial agent). Previous studies confirmed that imidazole groups retain great antibacterial potential by releasing autolytic enzymes that destroy the cell wall of bacterial cells, which causes a leak of the components and cell death. GO nanosheets were integrated into PVDF as a hydrophilic modifier. This study showed that the bacterial adhesion on Im-GO/PVDF membrane decreases with the increase of Im-GO concentrations. Im-GO 0.2 wt% had the highest antibacterial efficiency against E. coli colonies, which was 96.4%, which damaged the cell membrane's integrity and released intracellular substances.

Ahmad et al. (2020) have fabricated a polyethersulfone (PES) membrane incorporated with zeolitic imidazole framework-8 (which is zinc attached to methylimidazole ligands) decorated with graphene oxide as an antibacterial agent by the direct post-modification method. Based on the literature, metal–organic framework (MOF) materials have many advantages over other antibacterial agents, such as high surface area, tailored functionality, structural diversity, and tunable properties, promoting sustainable antibacterial impact (Wyszogrodzka et al., 2016; Quirós et al., 2015). GO here was used due to its hydrophilic nature and large surface area; it is also effective and has antibacterial activity against various species of bacteria. The researchers have tested the antibacterial activity of the modified membrane against *E. coli* and *S. aureus*, and the results showed that the ZGO-NH membrane with 1.0 wt% showed an optimum performance, and colonies showed large area distribution in comparison to the unmodified membrane, the antibacterial efficiency was 81.1% and 85.7% against *E. coli* and *S. aureus*, respectively.

Cheng et al. (2018) modified the porous PVDF/UF membranes to enhance their antibacterial and antifouling properties, as well as their perm-selectivity. The modifications were achieved using "micromolecular zwitterionic materials" (DMAPAPS), which are defined as materials with equivalent sites of cations and anions. In addition, one-step co-deposition of DMAPAPS and dopamine (DA) was used to modify the hydrophobic PVDF/UF membrane with oxidation of $CuSO_4/H_2O_2$ for assistance. The modified membrane showed spectacular fouling resistance of 95% and superior antibacterial activities for *E. coli* and *S. aureus*. Also, the modified membrane showed excellent stability performance even under harsh pH, and with the assistance of oxidation, the modification of the membrane was done in only 40 minutes, and the hydrophilicity of the membrane was improved as the water contact angle was found to be 33°. Table 4 discusses different materials tested to control biofouling in UF membranes.

4.3.5.2. Membrane surface modification to mitigate NOM fouling. According to Peters et al. (2021), the presence of NOM is undesirable for the following reasons: their abundance influences the properties of the water, such as color, odor, and taste. Also, NOM can react with disinfectants and produce carcinogenic disinfection by-products. They also can act as a carbon source for microorganisms which enhances their growth in distribution networks. Moreover, NOM such as fulvic acids and humic acids can form organometallic complexes with potentially toxic elements and increase their transport, toxicity, and bioavailability.

NOM with low molecular weight particles can be challenging for UF membranes as these foulants can pass through the membrane's pores without any obstacles. However, a small percentage of NOM with low molecular weight can get adsorbed to the membrane's pores, resulting in pore blocking and flow obstructions (Li et al., 2018). Moreover, UF can reject NOM with a larger size than the membrane's pores. Therefore, these NOM with immense molecular weight can cause cruel membrane fouling (Vrouwenvelder et al., 2011). For example, Ding et al. (2021) evaluated the performance of holey graphene oxide (HGO) modified UF membrane in its ability to separate pollutants, purify water, and mitigate the membrane fouling caused by natural organic matter. The study outcomes showed that the water permeability was twice as higher in HGO-modified membranes compared to membranes coated with GO only. Moreover, it was also reported that HGO modified membrane with an HGO coating amount of 0.04 g/m^2 has an increase in rejection rates for NOM-like bovine serum albumin (BSA) from 55 to 85%, for sodium alginate (SA) from 29% to 72%, and for humic acid (HA) from 58 to 92%. This was mainly due to the increased membrane hydrophilicity as the water contact angle decreased from 71° to 35° after surface modification. The study also revealed that coating the PES membrane with HGO significantly enhances the antifouling performance compared to the unmodified PES membrane and inhibits the cake layer formation due to the negative charge of HGO and high hydrophilicity. Also, as shown in Fig. 4, as the amount of HGO coating increases, the antifouling ability of the membrane improves. Table 5 summarizes various studies conducted related to the improvement in antifouling properties of UF membrane.

Like any other membrane process, the properties of the membrane are considered excellent if have the following: high rejection rate, high flux, and low fouling tendency under mechanical and chemical stress over a sustained period of time (Yu et al., 2022). Based on the findings of this review, the approach of modifying the membrane with composite-based nanomaterials has been found undoubtedly feasible and convenient in addressing the toxic metal removal and the antifouling problem of UF membrane. The related studies that have been extracted from the literature have shown that using this approach contributes to the enhancement of hydrophily, and flux recovery, as well as these modifications have antibacterial properties that also significantly contribute to the fouling resistance of the membrane. In Table 6, the reported materials were evaluated from different aspects including separation performance and antifouling, leaching and toxicity, preparation simplicity, and cost (Table 6).

5. Challenges and limitations

Various studies have shown the development of UF membranes modified with organic, inorganic, and hybrid materials for the removal of toxic elements. Nevertheless, there are certain challenges in the widespread applications of these UF membranes. Fig. 5 shows 12 major challenges and limitations toward the application of UF membranes for the removal of toxic elements. The first 3 challenges are related to the materials (nanoparticles, inorganic filters, nanocomposites) used in the development of modified membranes. First of all, there is limited data available on the toxicity assessments of these materials. In case of their detachment or leaching from the membranes, they may become part of the product water. Therefore, it is suggested to obligate the toxicity studies for the materials being tested. Also, the agglomeration of nanoparticles with time can also lead to the deterioration of the membrane performance, and therefore, it is one of the major limitations of their application in membranes. Future studies should also focus on the development of easy to scalable and cost-effective membrane modification procedures. Often, it is noted that the membrane development procedure is a multi-step process that requires large volumes of chemicals, reagents, etc. as well as expert skills which makes the overall process more complicated and difficult for industrial-scale implementation.

From Fig. 5, the challenges and limitations mentioned in numbers 4–6 are related to the membranes. The loss of permeability (and sometimes selectivity) after modification is commonly noticed which will jeopardize the operating

Table 4
Surface modification of UF membranes to mitigate biofouling.

No.	Technique	Bacteria studied	Flux recovery ration (FRR)%	Antibacterial rate %	Remarks	References
1	Fabricated photocatalytic membrane by functionalizing polyvinylidene fluoride (PVDF) (UF) membrane with titanium oxide (TiO2) nanoparticles	Antibiotic resistance bacteria (ARB)	–	98%	Modification of UF membrane by TiO2 showed excellent antifouling properties and reached 98% after exposure to UV radiation	Ren et al. (2018)
2	Silver lactate-holloysite nanotube clay nano-filler embedded into polyvinylidene fluoride polymer matrix (UF nanocomposite membranes)	*Salmonella, Enterobacter aerogenes*	–	99%	The modified UF showed a small amount of silver leaching and string antibacterial performance as confirmed by the inhibition zone formed around the membrane.	Moslehyani et al. (2015)
3	Polysulfone UF membranes impregnated with silver nanoparticles (nAg)	*E. coli* K12 *Pseudomonas mendocina* KR1	–	99%	Antimicrobial activity was primarily due to the release of Ag+ ions. The modified membrane showed a significant improvement in virus removal	Zodrow et al. (2009)
4	PVDF UF membrane modified by dopamine and zwitterion (DMAPAPS)	*E. coli* *S. aureus*	–	100%	Antibacterial activity was excellent due to the copper ions and ammonium groups that existed in PDA-DMAPAPS coating and the water contact angle decreased to 33°	Cheng et al. (2018)
5	amine-functionalized ZIF-8-decorated GO for UF membrane	*E. coli* *S. aureus*	84.4	81.1% 85.7%	Modified membrane with 1.0% ZGO-NH has superior antibacterial performance against Gram-positive and Gram-negative bacteria	Ahmad et al. (2020)
6	Fabricated polyethersulfone silver composite (silver nitrate AgNO3) UF membrane	*E. coli* *S. aureus*	–	100%	Membrane with 0.5 wt.% of Ag exhibited the best performance against gram-negative bacteria	Basri et al. (2011)
7	Graphene oxide (GO) modified by hyperbranched polyethyleneimine (HPEI) blended into polyethersulfone (PES)	*E. coli* *S. aureus*	92.1	74.88%	The membrane was prepared by dispersing HPEI-GO nanosheets in the PES casting solution	Yu et al. (2013a,b)
8	Carbon nanotubes (CNTs) were functionalized by sodium lignosulfonate (SLS) and blended with polyethersulfone (PES)	*E. coli*	95	100%	SLS-CNT/PES UF membrane showed excellent antibacterial performance with low voltage electric field	Wang et al. (2018)
9	SiO2@N-Halamine/polyethersulfone UF membrane	*E. coli*	96	60.22%	Membranes with 5% of SiO_2@N-Halamine showed the optimum performance in the antibacterial process	Yu et al. (2013a,b)
10	Zwitterionic poly(aryl ether oxadiazole) UF membrane	*E. coli*	97.6	98%	Zwitterionic membranes exhibit high resistance to bacterial adhesion and biofilm formation	Guo et al. (2020)
11	Fabricated Imidazole-functionalized graphene oxide PVDF UF membrane	*E. coli*	80.8	96.4%	The antibacterial efficiency reached the optimum in membrane 0.2 wt% of Im-GO	Geng et al. (2021)
12	Copper (II)-chelated polyacrylonitrile UF membrane	*E. coli*	91	71.5%	PAN-PEI-Cu membrane exhibits better antibacterial performance compared to unmodified PAN-PEI membrane which is 14.5%	Xu et al. (2012)
13	Macrovoid-free polyethersulfone/sulfonated polysulfone (PES.SPSf) UF membrane	*E. coli*	>90	99%	The highest O-MWCNT loading the better antibacterial performance	Gumbi et al. (2018)

cost of the membrane system. In addition, most of the studies focus on the development of modified membranes for the removal of toxic elements does not investigate membrane fouling. Although, membrane fouling is one of the major drawbacks of the use of membrane technology. Therefore, future research should focus on the development of multi-functional membranes with high rejection capabilities and high resistance to membrane fouling. The next 3 limitations

Table 5
Various UF-based membranes reported for improved antifouling performance (protein foulants).

No.	Technique used	Optimum concentration (wt%)	Flux recovery ratio (%)	Remarks	References
1	Carboxylic polyethersulfone blend hollow fiber UF (CPES/PES-UF)	12	86	The incorporation of CPES increases the liquid-liquid phase separation and permeation flux by 300%.	Heidari et al. (2021)
2	Blended polydopamine-coated graphene oxide nanosheets with sulfonated poly(ether sulfone) (PDAGO/SPES)	10	93.4	The membrane had a flux of 1138.7 L $m^{-2}h^{-1}$ and the irreversible fouling ratio of the membrane was 6.9% compared to 32.7% for the pristine membrane	Kumar et al. (2022)
3	Cellulose nanocrystals incorporated into polyethersulfone UF membrane (PES/CNC)	5	97	The flux recovery rate increased from 51% to 90% when the CNC concentration increased from 0 to 5 wt%	Zhang et al. (2018)
4	Troger's base polymer modified by zwitterion and blended with UF membrane (TB/ZBT)	6.9	44.4–52.3	The adsorption capacity of BSA dropped from 90.6 $\mu g/cm^{2}$ in TB membrane to 46.8 $\mu g/cm^{2}$ in TB/ZTB membrane	Huang et al. (2012b)
5	Hydrous manganese dioxide nanoparticles incorporated into polyethersulfone (PES/HMO MMM)	1.5	96.15	The highest BSA rejection was 98% and it is the membrane that has the highest HMO loading	(Gohari et al., 2013)
6	Al_2O_3-PVDF nanocomposite tubular UF membrane	1	100	the best performance was achieved at pH 10 and the flux of the modified membrane is 138.53 L/m^2 h compared to 66.88 L/m^2 h of the unmodified membrane	Yan et al. (2009)
7	Polyethylene glycol/polyethyleneimine UF membrane (PEG-g-PEI/UF)	–	98.26	After fouling for 30 min, only decreased to 95.30% at 60 min, and was still as high as 92.20% after fouling for 90 min.	Wei et al. (2021)
8	Sulfonated polyethersulfone/sulfopropyl methacrylate membranes impregnated with polysulfopropyl acrylate coated ZnO nanoparticles (ZnO-g-PSPA/SPES UF)	5	99	The enhancement improved the porosity and the hydrophilicity of the membrane which resulted in a high permeate flux of 99% and an excellent flux recovery ratio of up to 99% in the antifouling measurement.	Alosaimi et al. (2022)
9	Polyethersulfone hybrid membrane incorporated with sulfonated polydopamine	4	92.3	The incorporation of negatively charged SO_3 improved the membrane properties	Kallem et al. (2021)
10	Fabricated UF membrane with integrated zwitterionic and TiO2	–	100	Protein rejection and fouling removal efficiency improved remarkably compared to the unmodified PVDF membrane by about 100%	Zhao and Shen (2020)

Table 6
Comparison of various parameters of the four types of nanomaterials used for surface modification of polymeric UF membranes.

Parameters	Inorganic fillers	Organic based nanomaterials	Carbon-based nanomaterials	Composite-based nanomaterials
Separation performance and antifouling	✓✓	✓✓	✓✓	✓✓✓
Leaching and toxicity	✓	✓✓✓	✓✓	✓✓
Preparation simplicity	✓✓	✓✓	✓✓	✓
Popularity in UF modification	✓✓	✓	✓✓	✓✓✓
Cost	✓✓✓	✓	✓✓	✓✓
Overall rating (out of 15)	10	9	10	11

✓✓✓means satisfactory, ✓✓means intermediate, ✓means unsatisfactory.

are related to the scope of the research works. The data related to the performance of modified UF membranes while treating real wastewater is scarce. Also, most studies lack a transformation perspective in which research work done at the lab scale should be implemented at the pilot scale over a longer time duration. This will help to collect additional information on the sustainability and durability of the membranes and it will also bridge the gap between lab-scale work and industrial applications. In the end, it is also important to perform cost estimations for the modified membranes and their applications, investigate strategies for their regeneration and their disposal after use, and any technical requirements for the industrial scale applications of the modified membranes.

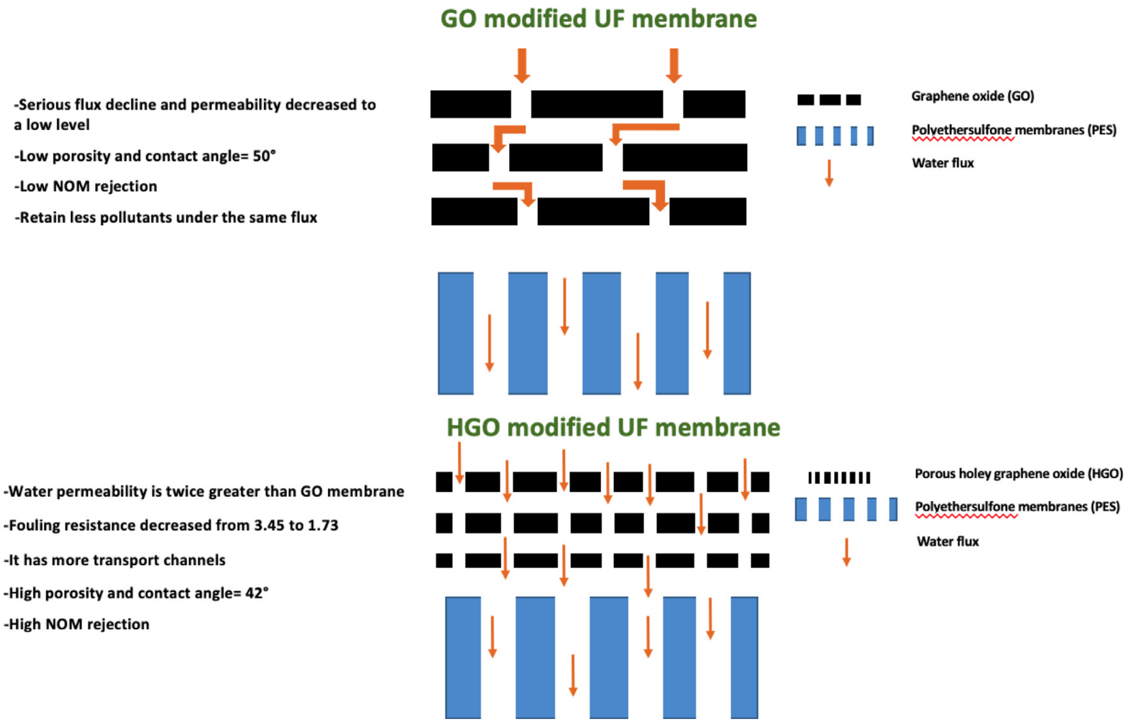


Fig. 4. Demonstration of filtration process through GO and HGO modified UF membrane, information retrieved from Ding et al. (2021).

Fig. 5. Challenges and limitations in using UF for the removal of toxic elements.

6. Conclusion

UF is a low-pressure-driven membrane process that has been extensively used in different industrial applications and water treatment processes. However, the UF membrane has poor performance in eliminating potentially toxic elements and is susceptible to fouling by bacteria and NOM. These are the most common foulants that severely impacted the performance of UF membranes. Therefore, this review comprehends the recent research progress on the development of UF membranes for superior heavy metal removal capacity and better anti-fouling performance. The bibliometric analysis conducted showed steadily increasing trends in the number of publications in the studied subject areas. Several researchers have developed advanced and novel UF membranes for better performance including incorporating different types of materials like organic, inorganic, carbon-based, and composite-based substances into the polymeric UF membrane to enhance the separation performance and control fouling. Examples of these materials include zeolites, metal–organic frameworks (MOFs), graphene oxide (GO), and zinc-doped aluminum oxide ($ZnAl_2O_3$). Functionalizing the membrane's surface with these materials showed good separation performance for different heavy metal ions and promising results in terms of controlling membrane fouling. However, based on the overall critique done in this review, composite-based nanomaterials showed exceptional performance in different aspects like separation performance and antifouling, leaching, research popularity, and cost efficiency which make them more favorable than other materials. However, most of the researchers aim to develop and test membranes at only laboratory scale conditions and there is a serious lack of literature related to the application of these membranes at a larger/pilot scale. Also, there is a need to develop a membrane modification/fabrication process that is more scalable, cost-efficient, and environmentally friendly for faster and easier application at the industrial level.

CRediT authorship contribution statement

Rouzan Shoshaa: Data curation, Investigation, Writing – original draft. **Mohammad Y. Ashfaq:** Data curation, Investigation, Writing – original draft. **Mohammad A. Al-Ghouti:** Data curation, Funding acquisition, Project administration, Conceptualization, Investigation, Supervision, Writing – review & editing.

Declaration of competing interest

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

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Appendix A. Supplementary data

Supplementary material related to this article can be found online at https://doi.org/10.1016/j.eti.2023.103162.

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