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Removal of heavy metals from wastewater using reverse osmosis

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This study presents an overview of and innovations in reverse osmosis (RO) membrane processes for rejecting charged metal ions in wastewater in relation to the main problems associated with purification methods. It also explains the emergence of nanomaterials and the different methods applied for RO membrane modification to improve performance. Membrane regeneration and retentate management are also considered. The study concludes with an economic feasibility study for the industrial scale-up of the methodology.

KEYWORDS

reverse osmosis, heavy metal, thin-film composite, thin-film nanocomposite, wastewater

1 Introduction

Both the environment and human health are under serious pressure from inorganic and organic micropollutants due to their non-biodegradable nature, persistence, toxicity, bioaccumulation in the food chain, and biomagnification in higher organisms (Feng et al., 2021; Peng and Bartzas, 2021). Micropollutants are contaminating flora and fauna, and the air, atmosphere, water, and soil; they have become progressively more pronounced over the past decade as a result of global population growth (Peng and Bartzas, 2021).

In ecological terms, any metal or metalloid that causes environmental pollution or has no biological interest in the organism can be considered a heavy metal (HM) (Herrera-Estrella and Guevara-Garcia, 2001). Some of these metals are micronutrients necessary for plant growth (e.g., Zn, Cu, Mn, Ni, and Co), while others have unknown biological functions and are toxic (e.g., Cd, Pb, and Hg) According to Feng et al. (2021) and Herrera-Estrella and Guevara-Garcia (2001), a "heavy metal" is a metal or metalloid element that causes environmental pollution, has no vital function, and is toxic at low concentrations (such as Pb and Hg), or has a vital function but is harmful to organisms at high concentrations (such as Cu and Mo). Heavy metals have been categorized as toxic metals (Hg, Cr, Zn, Cu, Ni, Cd, As, Co, Sn, etc.), precious metals (Pd, Pt, Ag, Au, Ru, etc.), and radionuclides (U, Th,

Abbreviations: BOD_5, biological oxygen demand; ICP, inductively coupled plasma; Init. conc., initial concentration; CA, cellulose acetate; CNTs, carbon nanotubes; COD, chemical oxygen demand; CS, chitosan; GO, graphene oxide; HM, heavy metal; LPRO, low-pressure reverse osmosis; MF, microfiltration; MMMs, mixed-matrix membranes; Na_2EDTA, disodium salt of ethylenediaminetetraacetic acid; NA, not available; NF, nanofiltration; ND, no data; PA, polyamide; PAA, polyacrylic acid; RO, reverse osmosis; SRB, sulfate-reducing bacteria; SDS, sodium dodecyl sulfate; SWRO, seawater reverse osmosis; TFC, thin-film composite; TDS, total dissolved solids; UF, ultrafiltration; WHO, World Health Organization; WW, wastewater.

Ra, Am, etc.) (Wang and Chen, 2009). The most common are Pb, Zn, Hg, Ni, Cd, Cu, Cr, and As, which are always dangerous, even when detected at trace levels. Table 1 below lists the main sources of heavy metals, their health effects, and their permissible drinking water standard. Other metals frequently present in wastewater, such as Ag, Fe, Mn, Mo, B, Ca, Sb, and Co, must be completely eliminated (Qasem et al., 2021).

Heavy metals must be treated due to their non-biodegradable properties (Fu and Wang, 2011). Trace heavy metals can be toxic through the process of biomagnification, which can increase their concentration to a point where they become toxic (Khan et al., 2008). Several processes are used to remove heavy metals, including chemical precipitation and electrochemical treatment, but these are not effective for concentrated ions and produce excessive amounts of sludge. Ion selectivity is high in ion exchange treatment, but the cost of the resins is too high (Bashir et al., 2019). Adsorption uses either inorganic adsorbents—natural minerals, ores, clays and industrial solid waste such as bauxite red mud, slag, ash, water treatment sludge (alum), and seawater-neutralized red mud—or organic adsorbents—waste organic matter from plants or animals (Khan et al., 2008; Zhu et al., 2019). Sulfate-reducing bacteria (SRB) are used to biologically remove heavy metals, producing metal sulfide precipitates on a large scale (Perales-Vela et al., 2006). This treatment has weaknesses such as long residence times and the need for continuous feeding substrates and larger bioreactors (White et al., 1997; Perales-Vela et al., 2006). Moreover, microalgae are limited and do not purify effluent (Monteiro et al., 2012).

It has become imperative to look for new technologies to remove heavy metals, such as membrane technology. The right technology needs to be scalable, applicable to field conditions, economical, and capable of eliminating heavy metal concentrations to the established standard (Sheng et al., 2004). Membrane technologies are therefore of immediate interest for the quality of treated water. Table 2 below summarizes the strengths and weaknesses of biological, chemical precipitation, electrochemical, and pressure filtration processes.

Many studies have reviewed the application of reverse osmosis (RO) membranes. For example, Abdullah et al. (2019) conducted a literature review of the use of membrane technologies for heavy metal removal. They discussed the performance and capabilities of different membrane processes and their advantages and disadvantages. Guo et al. (2022) studied the scale inhibition mechanism and modification strategy, which influences the biofouling of RO membranes. Xiang et al. (2022) reviewed the latest developments, discoveries, and prospective applications related to ultrafiltration (UF), nanofiltration, reverse osmosis, and electrodialysis, with an in-depth focus on heavy metal removal. They presented perspectives on opportunities and challenges in the field of membrane filtration. Saleh et al. (2022) evaluated removal processes by chemical precipitation, photocatalysis, flotation, ion exchange, remediation, electrochemical treatment, adsorption,

TABLE 1 Typical heavy metals present in wastewater (WHO, 2017; Qasem et al., 2021).

Heavy metal	Main sources Krishna Kumar et al. (2015); WHO, (2017); Demiral et al. (2021)	Main organ and system affected Ngah and fatinathan, (2008); Fu and Wang, (2011); Owalude and Tella, (2016); Marciniak et al. (2019); Duan et al. (2020)	Permitted amounts (µg) Demiral et al. (2021)
Pb	Lead-based batteries, solder, alloys, cable sheath pigments, rust inhibitors, ammunition, enamels, and plastic stabilizers	Bones, liver, kidneys, brain, lungs, spleen, immunological system, hematological system, cardiovascular system, and reproductive system	10
As	Electronics and glass production	Skin, lungs, brain, kidneys, metabolic system, cardiovascular system, immunological system, and endocrine	10
Cu	Corroded plumbing systems, electronics, and cables industry	Liver, brain, kidneys, cornea, gastrointestinal system, lungs, immunological system, and hematological system	2000
Zn	Brass coating, rubber products, some cosmetics, and aerosol deodorants	Stomach cramps, skin irritations, vomiting, nausea, anemia, and convulsions	3,000
Cr	Steel and pulp mills and tanneries	Skin, lungs, kidneys, liver, brain, pancreas, tastes, gastrointestinal system, and reproductive system	50
Cd	Batteries, paints, steel industry, plastic industries, metal refineries, and corroded galvanized pipes	Bones, liver, kidneys, lungs, testes, brain, immunological system, and cardiovascular system	3
Hg	Electrolytic production of chlorine and caustic soda, runoff from landfill and agriculture, electrical appliances, industrial and control instruments, laboratory apparatus, and refineries	Brain, lungs, kidneys, liver, immunological system, cardiovascular system, endocrine, and reproductive system	6
Sb	A rare naturally occurring element; its presence in aquatic ecosystems, soils, and the atmosphere is justified by volcanic activity, the weathering of rocks, and the use of products containing antimony	According to the NCBI (National Center for Biotechnology Information), antimony poisoning can cause gastrointestinal distress, nausea, and even ulcers. Inhalation exposure to antimony causes pneumonitis, and long-term consumption leads to cancer	6
Ni	Stainless steel and nickel alloy production	Lung, kidney, gastrointestinal distress, pulmonary fibrosis, and skin	70

TABLE 2 Strengths and weaknesses of biological and pressure filtration processes (Wang and Chen, 2009; Chen et al., 2018; Perpetuo et al., 2011; Monteiro et al., 2012; Bashir et al., 2019).

Type of process		Strength	Weakness
Biology	Bioremediation [microorganisms, sulfate-reducing bacteria (SRB)]	- Low investment costs	- Ineffective for highly concentrated ions in solution (Jiang et al., 2018)
	Phytoremediation (plants)		- Long adsorption residence time
			- Does not purify the effluent and requires space
		- Effective for low concentrations of HMs in solution	- Accumulation depends on the following parameters: i) metal ion properties (atomic weight and valence);
		- Energy-saving	ii) biotic and abiotic parameters (pH, temperature, ionic strength, contact time, and biomass concentration); iii) biosorbance type (may determine differences in selectivity and affinity for metal ions)
			- Lack of awareness regarding metal–microbe interactions remain unexploited and at times is indecipherable
Membrane filtration and physical–chemical treatment	- Reverse osmosis	- Less cumbersome	- High capital cost
		- High removal rate and purifies the effluent	
		- Simple scale-up	- Energy consumption
		- Treating large amount of industrial effluent	
	- Chemical precipitation	- Used for low concentrations	- Treatments are ineffective
	- Electrochemical treatment		- Produces large quantity of sludge
	- Ion exchange	- Higher ion selectivity	- Expensive
		- Reusability of ion-exchange material	

membrane technologies, and coagulation/flocculation. Qasem et al. (2021) exhaustively and critically reviewed and discussed methods in terms of the agents/adsorbents used, metal ion removal efficiency, operating conditions, and the advantages and disadvantages of each method. In this study, we focus specifically on the different materials and specific methods for synthesizing RO membranes, as well as different wastewater treatment methods. The information available in this review will allow researchers to understand the available work and then consider the synthesis of new innovative RO membranes. It provides researchers with comprehensive data on metal and metalloid discharges and describes the performance of different membranes in terms of permeate flux and methods of heavy metal removal from wastewater, as well as the necessary information on chemical membrane cleaning and end-of-life membrane management.

This review also addresses the technical challenges of the existing membrane process and recommends future research for further improving membrane performance to make it the best alternative for treating water laden with heavy metals. This work is not the first on the subject to eliminate heavy metals. A great deal of work has been done to eliminate metal ions from water. For example, Dompé and Ahoulé, (2016) conducted a study on borehole water intended for consumption that was contaminated with arsenic. The removal of As (V) by reverse osmosis membrane (TW30) was 97.6% effective. A synthetic tannery effluent was first treated with commercial reverse osmosis membranes (BW30 and SW30). Next, a CS membrane on a polyethersulfone (PES) support (cs-PES MFO22) was prepared. The cs-PES MFO22 membrane was very effective at removing >99% of chromium. Removal of Cr, Ca, Mg, K, and Na on a BW 30 membrane ranged from 80%–90%, 85%–98%, 80%–97%, 60%–80%, and 60%–80%, respectively, and, on a SW30 membrane, 80%–98%, 80%–98%, 80%–98%, 78%–96%, and 78%–96%, respectively (Zakmout et al., 2020). Conidi et al. (2018) conducted a study to reduce the salinity of flue gas desulphurization (FGD) wastewater. After softening with Na_2CO_3 H_2O and ultrafiltration, the wastewater was filtered through two commercial thin-film composite polyamide RO membranes (SWC-2540 and ESPA-2540 from Hydranautics). Experimental results indicated that the SWC-2540 membrane performed better in rejecting ions: Mg^{2+} ions were completely rejected, while the rejection of monovalent ions such as Na^{+} was approximately 95.5%. The ESPA-2540 membrane showed rejection of Ca^{2+} and Mg^{2+} higher than 86.5%, whilst the observed rejection of Na^{+} was 80%. For the SWC-2540 membrane, an increased rejection of Ca^{2+} and Na^{+} ions was observed by increasing the operating pressure in the range of 16–50 bar. Mg^{2+} ions were totally rejected independently by the operating pressure. MIL-101 (Cr) nanoparticles were doped into the dense layer of selective polyamide (PA) on the polysulfone (PS) ultrafiltration support to prepare a thin-film nanocomposite membrane for water desalination. sodium chloride (NaCl) removal on TFN-MIL-101 RO was greater than 99% (Xu et al., 2016).

2 Reverse osmosis membranes

2.1 Thin-film composite membrane

Membrane separation technology is at the cutting edge of water purification (Friess et al., 2021). Inorganic, organic, and pharmaceutical compounds and salts dissolved in water are easily removed by reverse osmosis (Foureaux et al., 2019), making it a key technology for alleviating the water crisis (Shannon et al., 2008; Greenlee et al., 2009; Elimelech and Phillip, 2011; Chuang and Dudeney, 2018). Reverse osmosis is more effective at eliminating micropollutants and purifying effluent and is easy to scale-up. Cellulose acetate (CA) and thin-film composite (TFC) membranes are attracting industry interest, and the TFC membrane is attractive for its ability to reject solutes. CAs and aromatic polyamides (APs) are the active polymer derivatives used for RO membrane coatings (Sagle and Freeman, 2004; Ahmed et al., 2015; Makisha, 2019; Alanood et al., 2021).

These membranes have some advantages and disadvantages; CA membrane has high pH sensitivity, lower tolerance to high temperature, and it is more resistant to chlorine than TFC membrane. However, TFC membranes are much more efficient and effective than CA membranes. Interfacial polymerization, composite coating, and multilayer composite molding are the procedures commonly used for membrane preparation (Jackson and Hillmyer, 2010; Lalia et al., 2013; Ahmed et al., 2015; Sandu et al., 2022). TFC is the principal membrane used in the RO process and is characterized by a very thin active layer of PA that is formed on a porous substrate (Ismail et al., 2015; Xu et al., 2016). The thickness of the thin film is <0.2 μm with an interstitial void size ≤0.5 nm between the polymer chains (Maruf et al., 2012; Ismail et al., 2015; Gan et al., 2020; Zhang et al., 2022). The main determinant of the selectivity and water flux of the polyamide membrane is the selective layer formed by the interfacial polymerization reaction of aqueous amine and acyl chloride (Porter, 1989; Chuang and Huang, 2018).

The first research on RO dates back to the 1970s. For example, Kremen et al. (1977) demonstrated the possibility of purifying wastewater of various metal ions with an integrated process containing RO and precipitation units (Algieri et al., 2021). Zn^{2+} and Cu^{2+} ions were removed by a low-pressure RO process in the presence of a chelating agent (EDTA) (Ujang and Anderson, 2000). They found that permeate flux varied as a function of pressure, EDTA concentration, and temperature. The evolution of RO performance was demonstrated for Cu^{2+} and Cd^{2+} removal (Qdais and Moussa, 2004). The experiments were performed with polyamide membranes characterized by a spirally wound configuration (Algieri et al., 2021). Harharah et al. (2022) showed that permeate flux and Cu (II) removal were directly proportional to operating pressure and feed temperature. Pressure increased from 10 to 40 bars and removal increased from 89.98% to 94.21% on the Dow Polyamide TFC BW30XFR membrane. Pretreatment of real industrial wastewater by electrocoagulation, followed by reverse osmosis, revealed 99.89% removal of Cr^{3+} ions (Rasha et al., 2020). Coagulation/flocculation pretreatment of industrial and mining wastewater was followed by reverse osmosis on polyamide membrane (DOW™ FILMTEC™ BW30-440i); turbidity, total dissolved solid (TDS) concentrations, antimony, arsenic, nickel, zinc, and iron were reduced by 85%, 96%, 95%, 66%, 82%, 48%, and 10%, respectively, in the permeate (Samaei et al., 2020). Agboolo et al. (2021) have shown that TFC membranes can maintain selectivity only at low water permeance between 1–20 $Lm^{-2}\,h^{-1}\,bar^{-1}$.

2.2 Thin-film nanocomposite membrane

Membranes mixed/coated with various emerging nanomaterials such as carbon nanotubes (Farahbakhsh et al., 2019), graphene oxides (Chu et al., 2017; Chu et al., 2017), MXenes (Ding et al., 2017; Han et al., 2017), and metal–organic frameworks (MOFs) (Denny et al., 2016; Basu and Balakrishnan, 2017) have been synthesized. These have attracted interest from the water industry due to their outstanding characteristics, including selectivity, high tunable porosities and large accessible surface areas (Lu et al., 2014), enhanced hydrophilicity and resistance to fouling, high ability to easily combine particular species/features without changing the topology of the structure (Evans et al., 2014), and diverse potential applications (Furukawa et al., 2013).

Zeolite imidazolate frameworks (ZIF-8) were specifically selected because they are porous crystalline materials consisting of well-ordered pores (Pan et al., 2011). They have a high specific surface area, and their high thermal, chemical, and hydrothermal stability (Bhattacharjee et al., 2014; Şahin et al., 2017) and durable synthesis at room temperature have attracted the attention of scientists (Abbasi et al., 2020; Li et al., 2021). The unique structure of ZIF-8 resists chemical and thermal attack (Zhang et al., 2012; Wang et al., 2019)

Mixed-matrix membranes (MMMs) combine the advantages of organic membranes and inorganic materials, increasing permeability and reducing fouling (Zheng et al., 2017; Bi et al., 2018; Jeon et al., 2018). Castro-Muňoz et al. (2021) showed that metal ions are removed by mixed-matrix membranes.

However, chitosan (CS) has a hydrophobic character, is an important renewable natural biomass, and is one of the cheapest natural polysaccharides with abundant groups (Agnihotri et al., 2004; LogithKumar et al., 2016; Yang et al., 2016; Liu et al., 2019). Its properties of antibacterial activity, biocompatibility, non-toxicity, and good film-forming have made it advantageous for use as a selective coating (Kumar and Ioan, 2016; Reza et al., 2019; Pishnamazi et al., 2020). CS is known to be an excellent metal ligand, forming stable complexes with many metal ions (Chui et al., 1996). Due to the presence of hydroxyl and amine groups, CS is widely used to remove HM ions from aqueous solutions (Bozorgi et al., 2018; Haripriyan et al., 2022). However, the poor mechanical properties of CS limit its potential (Upadhyay et al., 2021).

Other materials used for heavy metal rejection, such as incorporating UiO-66-NH_2 into PAN/chitosan nanofibers, have removed Pb (II), Cd (II), and Cr (VI) ions by 94%, 89%, and 85.5%, respectively (Jamshidifard et al., 2019). UiO-66-NH_2 nanoparticle incorporated into the polyvinylidene fluoride (PVDF)/CS nanofiber membrane showed 95.6% rejection of Cr

TABLE 3 List of key studies on the removal of HMs from wastewater by reverse osmosis.

Membrane	Element	Init. conc. (ppm)	Removal efficiency (%)	Condition	Permeate flux	Remarks	Reference
PAs (ES 20)	Cu^{2+}, Ni^{2+}, and Cr^{6+}	10–500	Synthetic feed: > 99.5, 99.9, and > 99.5. Industrial feed: 99.03, 99.37, and 98.75	1–5 bars, pH: 7–9	24.64 L/m^2.h	- Synthetic and industrial wastewaters were used	Ozaki et al. (2002)
						- Membrane effective area was 60 cm^2	
						- Feed solution was stored at 20°C	
					18.63 L/m^2.h	- pH value was varied between 3 and 9	
						- Permeate flux decreased with the increase in feed concentration	
						- Increasing the pH of the feed solution raised the heavy metal ion removal rate	
LPRO	Co	244–409	>95	3 bars, pH: 4–10	6.52 L/m^2.h	- To enhance LPRO membrane performance, polyacrylic acid (PAA) has been added to the membrane	Dang et al. (2016)
Membrane	Sr					- Low permeability was observed on the addition of PAA and had no effect on removal	
						- Therefore, the addition of PAA only caused fouling	
Thin-film membrane (TFM)	Ni^{2+}	44–169	99.3	pH: 6.5–7.5		- Synthetic WW was used	Ipek (2005)
						- Addition of 240 ppm concentration of EDTA enhanced zinc and nickel ion removal efficiencies from 98.9% to 99.6% and from 99.3% to 99.7%, respectively	
						- The operating pH was 4 to 8	
	Zn^{2+}	64–170	98.9			- Feed pH and conductivity did not greatly influence removal rates	
						- Increasing the added EDTA increased the feed conductivity and removal efficiency of metal ions	
TFC	Cu^{2+}	500	99.5	5.03 bar, pH: 7.8		- Synthetic WW was used	Mohsen-Nia et al. (2007)
						- Removal rate of copper ion was higher than that of nickel due to the greater ionic size of copper	
	Ni^{2+}					- Osmotic and applied pressures affected metal ion removal	
						- Addition of Na_2EDTA enhances metal ion removal efficiency	

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TABLE 3 (*Continued*) List of key studies on the removal of HMs from wastewater by reverse osmosis.

Membrane	Element	Init. conc. (ppm)	Removal efficiency (%)	Condition	Permeate flux	Remarks	Reference
Polyamide	Ca^{2+}	451	97	6–22 bar	29.3 L/m^2. h.	- Industrial wastewater was used	Lui et al., 2008
	Mg^{2+}	1,102					
	Na^{+}	82					
TFC PA	As^{3+}	<0.50	20–55	4–6 bars, pH: 4, 5, 6, 7	ND	- Synthetic wastewater was used	Chan and Dudeney (2008)
						- Operating pH ranged from 3 to 9	
						- The initial As^{5+} concentration and pH value ranging from 6 to 9 showed a slight influence on the retention efficiency of the membrane	
	As^{5+}		91–99			- RO removed > 90% of As^{5+} residual	
						- Due to the presence of arsenite as a neutral molecule, the RO membrane did not effectively remove As^{3+}	
Polyamide	Cu^{2+}	10–100	70–95	1–6 bars, pH: 7-8		- Synthetic wastewater was used	Zhang et al. (2009)
						- Addition of surfactant sodium dodecyl sulfate (SDS) improved the Cu^{2+} removal efficiency to 90%–99%	
						- Cu^{2+} removal of 59%–75% was achieved by applying the electro-reduction technology of the 3D electrode cell	
						- Electro-reduction process was slightly affected by the addition of SDS and EDTA	
						- Low-pressure RO recorded average removal efficiency of copper ion of 85%	
						- Initial pH of solution was 6.0	
	Cu^{2+}	0.012	100			Removal of suspended solids and reduction of COD and BOD_5 from the wastewater were performed by a series of biological treatment processes	Saad and Omar (2010)
	Co^{2+}	0.140	100				
	Zn^{2+}	0.162	90.74				
	Pb^{2+}	0.165	100				
	As^{5+}	0.972	100				
	Cd^{2+}	6.360	99.86				
	Cr^{6+}	0.149	87.92				

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TABLE 3 (*Continued*) List of key studies on the removal of HMs from wastewater by reverse osmosis.

Membrane	Element	Init. conc. (ppm)	Removal efficiency (%)	Condition	Permeate flux	Remarks	Reference
AD-SWRO, AG-BWRO, BW-30-BWRO, and AK-BWRO	B^{3+}	10–11	B>91.9	15–0 bars, pH: 8.6	94.7 L/m^2. h	- Geothermal water was used	Öner et al. (2011)
	Na^{+}	10.5–10.9				- Four types of RO membranes were used	
	Ca^{2+}	366	Mg^{2+}, Ca^{2+} 99.5			- All the tested membranes showed > 96.5% silica and salt removal efficiency	
	Mg^{2+}	26				- Increase in operating pressure showed favorable variations in both quantity and quality of permeate flux	
	K^{+}	3.7	Na^{+}, K^{+} >99.3			- Operating pressure and type of membrane greatly influenced both permeate flux and boron rejection efficiency. Silica rejection was over 96.5%	
		26					
TFC PA (TW-30 et XLE, Filmtec™)	B^{3+}	70	>99	5–20 bar, pH: 7–11		- Synthetic wastewater was used	Dydo et al. (2012)
						- RO removed boron in the presence of chelating polyols containing a 1,2-diol group	
						- Permeate flux boron removal was affected by feed pH, initial feed concentration, and operating pressure	
						- Increased boron removal was recorded with an increase in applied pressure	
						- Boron concentration in the permeate was found to be below 1 mg/L, even at high recovery	
						- Rapid membrane degradation or fouling was not detected	
	Cr^{6+}	50	>91 %	15–35 bar, pH: 1–6		-Synthetic wastewater was used	Çimen et al. (2014)
		100					
		500				- NaOH and 0.1 M HCl were added to the feed solution to adjust the pH; permeate recycling was the operating system	
		1,000					
TFC PA	Ni^{2+}		98	0.5 MPa		-Addition of EDTA to form complexes with ions	Pires da Silva et al. (2016)
	Cu^{2+}					- 99% elimination was achieved for one ion in solution and in ion mixtures	
PA	Ni^{2+}	50	98.5	1–4 bars, 10–40°C, pH: 2–5.5	10 to 56 L/m^2.h	-Synthetic wastewater was used	Algureiri and abdulmajeed (2016)
	Cu^{2+}	100	97.5				

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TABLE 3 (*Continued*) List of key studies on the removal of HMs from wastewater by reverse osmosis.

Membrane	Element	Init. conc. (ppm)	Removal efficiency (%)	Condition	Permeate flux	Remarks	Reference
	Pb^{2+}	150–200	96				
Graphene	Cu^{2+}	0.5–3 M	100.0 (for OH graphene), > 98.0 (for B- graphene), and 95 (for NH graphene)	50–300 MPa		- Synthetic wastewater was used	Li et al. (2017)
						- Three types of functionalized hydroxyl (OH), nitrogen (NH), and boron (B) group nanoporous graphene membranes were used as RO membranes	
						- Increasing membrane pore density increased the removal efficiency, while increasing ion concentration in the feed decreases the permeate flux	
						- OH, B, and NH graphene reached 100%, > 98%, and 95% Cu^{2+} removal efficiency, respectively	
TFC PA	Co^{2+}, Ni^{2+}, Mg^{2+}	39.4	98.6	41 bars, pH: 1.46	Permeate flux was 2.9 L/m2.h.bar	- Industrial wastewater was used	Ricci et al. (2017)
		214.9	98.1			- Both RO and NF were used to treat POX effluent generated from the gold ore mining process	
		2,429	98.6			- The operating range of pH for RO are 4 –11	
						- Under acidic conditions, the stability of the RO membrane was satisfactory	
PA	Zn,	150–500	99.49;	1–4 bars	48.44 L/m^2.h	- Feed solutions were prepared by $ZnCl_2$ $CuCl_2$ $.2H_2O$, $NiCl_2.6H_2O$, and $CrCl_3.6H_2O$	Al-Alawy and Salih (2017)
	Ni,		99.49;				
	Cu,		99.33;			- Operating temperature was 26–40 and the feed pH varied 4–7	
	Cr		99.93				
TFC PA	B^{3+}	5.688	34–48	21–76 bar, pH: 3–11	7.1–32.5 L/m^2. h.	- Industrial wastewater was used	Cingolani et al. (2018)
						- Triple-stage RO system was adopted	
						- Greater than 91% water recovery was achieved by a two-stage RO system	
						- TDS rejection factor was in a range of 91.1%–97.7%, while that for and COD was over 95%. Also > 94.0% was achieved for selenide removal	
						- Rejection factor for ammonia was in a range of 57.4%–77.3%	
						- Water recovery efficiency was > 91% for a two-stage RO system	

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TABLE 3 (*Continued*) List of key studies on the removal of HMs from wastewater by reverse osmosis.

Membrane	Element	Init. conc. (ppm)	Removal efficiency (%)	Condition	Permeate flux	Remarks	Reference
PA TFC IP/SA	NaCl	500	94.2	5 bars, 25°C, pH: 3–6,5	42.0 L/m^2.h	-Parallel installation of three identical 23.6 cm^2	Yu et al. (2018)
SW30	Cr	0.2–1.3	97	10–20 bar; pH: 3, 25±5°C	566–830 L/m^{2}h	-Wastewater produced by the aluminum industry	Atès and Uzal (2018)
	Ni	3.2–5.3	99			-RO membrane composed of polyamide with the non-porous active skin layer	
	Al		99				
	Pb^{2+}	0.034	< 1 ppb	1.76 MPa, pH: 6.12		-Pretreatment of wastewater with biological waste: cereals, rice pods, coffee waste, tea waste, sugar beet pulp, and mushrooms.	Thaçi and Gashi (2019)
	Zn^{2+}	0.153	< 0.002 ppb				
	Cd^{2+}	0.025	< 0.1 ppb				
	Co^{2+}	0.018	< 0.2 ppb				
	Mn^{2+}	1.146	0.006 ppb				
	Ni^{2+}	0.004	< 0.5 ppb				
Dow PA TFC BW30XFR	Cu^{2+}	25	89.98 to 94.21 %	10, 20, 30, and 50 bar; 25, 35, and 45°C; pH: 2–11	Effect of operating pressure	-Copper sulfate used to prepare stock solution	Harharah et al. (2022)
					44.28 to 124.98 kg/m^2·h;		
					Effect of feed temperature	-Temperature correction factor (TCF) was studied	
					46.12 to 67.0 (kg/m^2·h);	-Permeate flux and metal removal were calculated after every 10 minutes of sampling	
		50			Effect of feed concentration	- Total dissolved solids counter was used to analyze the samples	
		100			87.41 to 83.86 (kg/m^2·h).	-Permeate flux and metal ion removal increase with increasing pressure	
		150			Effect of feed flow rate	Permeate flux increases with rising temperature	
					82.2 to 84.3 (kg/m ^{2}.h)		
BW30XFR	Cr^{6+}	5	99.8	10, 30, and 45 bars; 25, 35, 45 and 55°C.	30 to 158 kg/m^2.h (25°C)	-Higher operating pressures and temperatures (10, 30, and 45 bar and 25, 35, 45, and 55°C)	Singhidi et al. (2022)
		30	94.3			-Metal ion removal is a function of feed concentration	
		100	77.2		70 to 226 kg/m^2.h (55°C)	-Permeate flux increases as temperature rises	
						-Increasing temperature and pressure affect polymer membranes with fouling	

(Continued on following page)

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TABLE 3 (*Continued*) List of key studies on the removal of HMs from wastewater by reverse osmosis.

Membrane	Element	Init. conc. (ppm)	Removal efficiency (%)	Condition	Permeate flux	Remarks	Reference
RO X-20™	Cr III	15, 26, and 56 ppm	99	50 bars		- Synthetic water and real water	Lumami et al. (2022)
	Ni II		98–99				
	Cd II		99				
	Pb II		99				
	Sb III		98–99			- MF sample pretreatment. Efficient removal of metal ions at different feed concentrations	
	As III		98–99				

(VI) (Pishnamazi et al., 2020). The incorporation of graphitic carbon nitride nanosheets (g-C_3N_4) into PSF membranes showed rejections of 95%, 80%, and 70% for lead, cadmium, and arsenic, respectively (Akshatha et al., 2021). The mixed-metal nanoparticle Al–Ti_2O_6 has been used in the polysulfone membrane to remove heavy metals. Al–Ti_2O_6 was prepared by the precipitation method, and the membranes were prepared by diffusion-induced phase separation method with different Al–Ti_2O_6 compositions. The membrane showed a rejection of approximately 96% for As, 98% for Cd, and 99% for Pb (Sunil et al., 2018). The Al–Ti_2O_6 mixed-metal nanoparticle increased metal ion removal in contrast to the other nanoparticles cited above. Thin-film nanocomposite membranes were prepared by the interfacial polymerization (IP) between PIP and trimesoyl chloride, followed by post-treatment with polyethyleneimine (PEI) or PEI-polyethylene glycol conjugate and then the immobilization of Ag NP. The IP was conducted on a polyethersulfone/poly (methyl methacrylate)-co-poly (vinyl pyrollidone)/silver nanoparticle (Ag NP) blend ultrafiltration membrane support. The TFNC membranes exhibited >99% rejection of Pb^{2+}, 91%–97% rejection of Cd^{2+}, 90%–96% rejection of Co^{2+}, and 95%–99% rejection of Cu^{2+} with permeate flux ~40 $Lm^{-2}\,h^{-1}$ at applied pressure 0.5 MPa (Bera et al., 2018).

Table 3 briefly explains the types of membranes and operating techniques applied in RO to remove heavy metals from wastewater.

It should be noted that the membrane operating techniques were conducted under differing laboratory conditions. However, Table 3 describes the variation in solute removal and permeability of the different membranes, depending mainly on the membrane synthesis and pore size, structure, and intensity. These results indicate that almost all PA TFC membranes succeeded in giving high rejection values of between 98%–100%, respectively. It should be noted that some PA TFC membranes such as BW30XFR, TW-30, and AD-SWRO showed rejection of B^{3+}, Cu^{2+}, and Cr^{6+} ions < 95% and were dependent on the feed concentration.

2.2.1 Types of RO membrane materials

RO membrane performance is mainly determined by the different types of materials used in its manufacture. Lee et al. (2012) reported that RO membrane is typically designed on a porous PSF support on which an ultra-layer, a thin barrier layer, is deposited on the top surface. They suggest that the emergence of nanomaterials could offer an interesting alternative to polymeric materials.

The emergence of nanotechnology to improve TFC RO membranes has been applied by several researchers. Cohen-Tanugi and Grossman et al. (2015) conducted theoretical research on the addition of graphene in reverse osmosis membrane. They reported that the performance of RO membranes was improved by the addition of graphene, which exhibits pore size uniformity of 0.40 ± 0.24 nm diameter and high permeability up to 3 L/m^2.h.bar. Chen et al. (2023) proposed improving the performance of TFC RO membranes by using a hydrophilic polycarboxylic acid–ZIF-8 hydrophobic nanoparticle bilayer interlayer on the PSF substrate prepared by the catechol autopolymerization reaction. The membrane exhibited 98.8% NaCl removal performance and the permeability was 4.2 L/m^{2}h.bar. Guan et al. (2023) prepared a polyketone membrane as a porous substrate using a standard NIPS procedure and another membrane on the PSF porous support, and proposed using MPD—the monomer for IP formation of the PA layer.

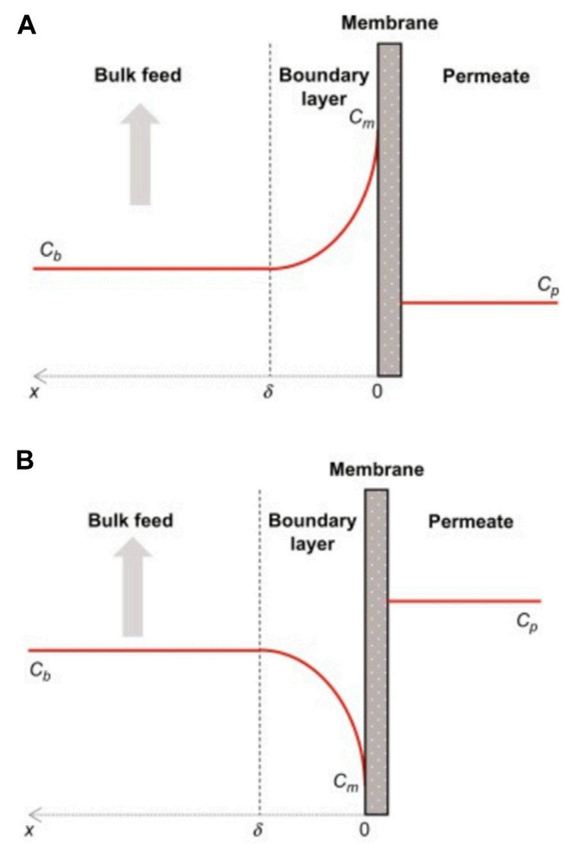

FIGURE 1
Concentration representation with concentration polarization: **(A)** mass transfer limited by the membrane and **(B)** mass transfer limited to the membrane surface (Luis, 2018). Transmembrane flux decreases as a result of concentration polarization, low retention of low molecular weight solutes, and can lead to higher retention for mixtures of macromolecular solutes.

The membranes showed NaCl rejection from 96.6% to 98.6% in a 2000 ppm feed solution, with permeability varying, respectively, from $0.23 \pm 0.13\,L\,m^{-2}\,h^{-1}\,bar^{-1}$ to $0.18 \pm 0.06\,L\,m^{-2}\,h^{-1}\,bar^{-1}$. The polyethersulfone material was modified by three hydrophilic molecules—thioglycolic acid, DL-cysteine hydrochloride, and 2-(dimethylamino) ethanethiol hydrochloride—to form a series of new hydrophilic polyethersulfone copolymers (HPES-TGA, HPES-CYSAH, and HPES-DMAET) that offer the best performance in terms of water permeability and water/salt selectivity (Zhang et al., 2023).

The addition of nanomaterials and modification of the RO membrane synthesis provide high performance, but a number of challenges remain in terms of the economics of industrial-scale application. The incorporation of nanoparticles or nanomaterials improved membrane performance, and this comparison was made between mixed-matrix membranes and TFC PA.

3 Performance of RO membranes

Numerous investigations have been made into the performance of the RO membranes in terms of pressure, flow rate (Dimitriou et al., 2017), concentration, and temperature (Ruiz-Gracía et al., 2020). Luis (2018) has shown that permeate flux is one of the most important factors in determining membrane performance, reflecting permeate quantity and species selectivity. The data from the other studies focused on permeate flux and solute release. The study by Armstrong et al. (2022) on the SWC4 seawater membrane and an ESPA3 brackish water membrane showed that membrane performance is evaluated on water-solute selectivity depending on the pressure applied during permeation tests. Qi et al. (2016) also found that water permeability and solute rejection are sensitive to temperature and applied pressure.

However, the introduction of graphitic carbon nitride (g-C_3N_4) nanosheets into PSF membranes improved the permeability and separation performance of PSF membranes at low transmembrane pressures ranging from 1 to 5 bar (Nadig et al., 2021).

The performance of the polysulfone composite membrane was improved by the addition of Al–Ti_2O_6 nanoparticles to remove heavy metal ions at low pressures of 200 kPa (Sunil et al., 2018).

It has been observed that the addition of nanomaterials to membranes enhances performance in terms of selectivity and water permeability at low operating pressures.

Many studies have demonstrated that the highwater flux is linked to better roughness (Al-Jeshi and Neville, 2006; Kong et al., 2010; Ma et al., 2018; Song et al., 2019a; Song et al., 2019b; Ma et al., 2019; Yan et al., 2019; Wu et al., 2020).

The selectivity of TFC membranes is excellent, but there is a trade-off with permeability. However, MMMs offer advantages such as low manufacturing costs, exceptional selectivity, and high packing density of polymeric materials with the long-term stability, high mechanical strength, and regenerative capacity of ceramic materials (Karan et al., 2015; Ayaz et al., 2019; Rezakazemi et al., 2019; Rozaini et al., 2019; Yang et al., 2019; Zhu et al., 2019). Table 3 also includes the performance of several RO membranes studied in the literature for metals removal.

4 Main shortcomings of RO membranes

4.1 Clogging

Pore clogging or solute adsorption on the membrane surface adversely affects membrane fouling (Van der Bruggen et al., 2003; Lee et al., 2011; Malaeb and Ayoub, 2011). Pretreatment is recommended to prolong the lifetime of the membrane. Ultrafiltration or microfiltration (MF), scaling control such as softening, and acidification for pH regulation constitute the pretreatments currently considered (Abba et al., 2023). Either microbial growth (Speth et al., 2000; Abba et al., 2023), scaling to organics, or particle matter can form cakes (Gabelich et al., 2005), and the membrane becomes vulnerable to fouling. Membrane fouling can be reduced to the best possible roughness and high hydrophilicity (Vrijenhoek et al., 2001; Van der Bruggen et al., 2003; Mondal and wickramasinghe, 2008).

4.2 Concentration polarization

The underpressure filtration process is constrained by the phenomenon of concentration polarization, which affects its efficiency (Koseoglu et al., 2018). During operation, the solute concentration on the membrane surface increases due to selective transport across the membrane (Luis, 2018). In the case of pressurized membrane processes, the solute is generally retained by the membrane, leading to a concentration profile similar to that shown in Figure 1A. During filtration, the membrane retains the solute, leading to a concentration profile similar to Figure 1B as the component permeates more rapidly through the membrane, being in the boundary layer where the transport is limited by diffusion (Luis, 2018). This phenomenon is similar to capacitive deionization in that it removes ions from the feed concentration. However, when pressure is applied across an ion permselective membrane, the ICP electrokinetic phenomenon occurs. This method is based on ion depletion and enrichment, which are dynamic changes in ion concentration near the membrane to maintain electroneutrality (Rabiee et al., 2019).

5 Fouling of reverse osmosis membranes

The pressurized membrane separation process is limited by the problem of fouling, which is unavoidable and reduces the performance of the process. The presence of inorganic and organic compounds in the water affects membrane fouling. Approximately 90% of suspended solids were eliminated in UF by Petrinic et al. (2015). To achieve this, Huang et al. suggested the need for a more advanced pretreatment process to avoid membrane fouling. RO membrane operation at low pressure with constant feed rate spares the membrane from vulnerability to fouling (Park and Kwon, 2018). Research by Lumami et al. (2022) has shown that changes in permeate flux depend on changes in pressure and affect membrane fouling.

Singhidi et al. (2021) reported that increased temperature and high pressure applied to the membrane alters the pore size, which leads to fouling. Li and Chen, (2010) cited membrane properties, feedwater quality, and ionic condition as the three factors influencing membrane fouling. Other parameters cited in the literature review include pressure, temperature, flow rate, and concentration—all of which influence membrane fouling.

6 Chemical cleaning of membranes

Fouling slows down wastewater treatment using membrane processes (Ang et al., 2011), leading to a drop in productivity. Membrane cleaning is extremely important for removing undesirable matter from the membrane surface and renewing its functionalization (Wilson et al., 2022). In the literature, very little research seems devoted to membrane cleaning (Porcelli and Judd, 2010).

Qi et al. (2016) showed that a solution of ethylenediaminetetraacetic acid (EDTA), sodium hydroxide (NaOH), and citric acid was better at cleaning the RO membrane and had good chemical stability. The chemical stability of the membrane was further confirmed by cleaning with a solution of citric acid, NaOH, and EDTA (Wilson et al., 2022). Ang et al. (2011) proposed that the addition of sodium chloride to solution sodium hydroxide (NaOH), ethylenediaminetetraacetic acid (EDTA), and sodium dodecyl sulfate (SDS) was ideal for cleaning membranes resulting from the treatment of alkaline solutions, metal chelating agents, surfactants, and salt. They concluded that cleaning performance increased with an increasing pH of the cleaning solution.

7 Management of heavy metal retentates

Reverse osmosis retentate can have a number of environmental impacts. However, rational management and reuse of retentate is recommended. Heavy metals are not generally recycled, but recycling by sector is recommended. The best organized channels are obviously those where the largest masses are treated in order to recover valuable compounds. This study proposes two retentate treatment options. The first is recycling for the use of heavy metals, which requires additional investment in additional treatment by a metallize electrolysis reactor. Metals can be recovered by a reactor that uses the principle of electrolysis to cause charged particles, such as heavy metals, to agglomerate on the cathode. The metal agglomerate can then be recovered and recycled (Lenntech). The second option considers the circular economy of the process. For example, the use of retentate as mixing water to produce calcio-sulfoaluminate cement bricks for construction is recommended (Valdés et al., 2021).

8 Management of end-of-life reverse osmosis membranes

The lifetime of the RO membrane depends on factors such as the RO membrane model, the type of pretreatment of the plant, the quality of the feedwater, the operating conditions, the location of the RO membrane, and the frequency of chemical cleaning (Kharraz et al., 2021). In view of the above, the average life has been estimated at between 5 and 10 years for pretreated feedwater (Kharraz et al., 2021); however, it is shorter at approximately 12 months when the feed solution is too loaded and if there is no pretreatment (Chang, 2006). At the end of RO membrane life, reuse in MF and NF is recommended by the literature. Khoo et al. (2021) converted RO membranes to end-of-life by modifying their microfiltration properties with $KMnO_4$ treatment and showed that the end-of-life of RO membranes should considerably increase in coming years. The membrane converted to MF had a NaCl rejection of approximately 80%, and the permeability to pure water was 172.6 L/m.h.bar. In addition, RO has been cited as the process that generates environmental impacts following the production of solid waste (Luisa et al., 2006).

Senán-Salinas et al. (2021) have estimated that, by 2025, two million membrane modules will be discarded worldwide by the desalination sector in developed countries. Kharraz et al. (2021) revealed that membranes consist of plastic parts and that their recycling will reduce landfill disposal.

In addition, these membranes are made from petroleum-based polymers and are responsible for greenhouse gas emissions (Senán-

Salinas et al., 2021). Rattanakul (2012) has shown that RO membranes contain 80% recycled components. However, plastics such as polyethylene terephthalate (PET), high-density polyethylene (HDPE), and polypropylene (PP) are recyclable and represent a step forward in reducing harmful impacts on the environment.

9 Conclusion

The RO membrane process is effective at removing organic and inorganic compounds from wastewater and in the simplicity of its scale-up. Removal of inorganic micropollutants varies between 95%–100%, depending on the type of polymer, materials, and operating conditions applied. However, the temperature rise applied in RO affects the membrane's vulnerability to fouling.

The application of CS and ZIF-8 in the process improved selectivity and water permeability. It was observed that metal oxides also perform best in terms of selectivity and permeability. The emergence of nanomaterials in the membrane process has improved operation at very low pressure, as in the case of nanofiltration, with low operating and maintenance costs.

Author contributions

VK: conceptualization, methodology, validation, visualization, and writing–original draft. PL: conceptualization, funding acquisition, methodology, project administration, supervision, validation, and writing–review and editing.

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