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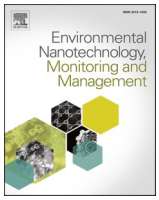

The impact of nanoparticle leach on sustainable performance of the membranes – A critical review

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ABSTRACT

Fouling remains a major challenge in membrane-based water treatment technologies. As a result, contemporary research is geared towards membrane surface modification techniques to reduce fouling. Mitigation strategies involving incorporation of nanoparticles (NPs) into polymeric membranes has gained a remarkable interest. However, NPs leach is eminent, particularly with poor support on membranes resulting in secondary pollution. Consequently, aquatic life is threatened depending on the level of toxicity of the leached NPS. Also, these NPs present toxicological effects to other water consumers. Therefore, this work reviews contemporary literature on membrane surface modification techniques paying attention to incorporation of NPs in the membrane polymer matrices. Various factors governing NPs leach are concisely presented. Special attention was focused on stability of the NPs immobilization on the polymeric membrane due to thermodynamic interactions. Similarly, the effects of NPs leach on membrane physicochemical properties and the NPS ecotoxicity are discussed in detail based on literature reports. Different approaches presenting improvement on NP stability in the polymer matrix are discussed. Lastly casting of future perspectives and the impact of NP leach on sustainable performance of the membranes and ecotoxicity is presented.

1. Introduction

Despite the current water scarcity and the provision of high-quality potable water challenge globally, the demand for water supply is estimated to increase in the coming years (Saraswathi et al., 2019). These challenges are further aggravated by continued pollution of water sources due to a myriad of contaminants including salts, trace metals (Edokpayi et al., 2017), dyes, emerging trace organic compounds (aus der Beek et al., 2016) and pathogens (Gitis and Hankins, 2018) detected in various water sources. These solutes originate from different sources including human and industrial activities. For instance, dyes are generated from the textile, rubber, and printing industries (Wang et al., 2018). Similarly, trace organic compounds are discharged from the production site, uncontrolled release of surplus drugs from households, leakage from sewer pipes, animals ejection, and farms effluents (Fauzan et al., 2018). Some of these contaminants are environmentally intolerable. Among other water contaminants, methylene blue dye impairs functionality of the kidneys, brain, central nervous system and the liver (Rajabi et al., 2017). Notably, similar consequences are experienced from exposure to trace organic compounds (Li et al., 2021). Some of these contaminants are endocrine disruptors, hence possess deleterious effects on human health at trace concentrations, thus necessitating urgent remedial attention.

Different water treatment methods are used to reduce the risk of human vulnerability to harmful chemicals. This involves preventative measures towards the discharge of poor-quality wastewater effluent. Remedial techniques include desalination using high retention membranes (Adel et al., 2022; Ruiz-García et al., 2019), electrocoagulation (Song et al., 2017), advanced oxidation processes (Fatta-kassinos et al., 2011; Feng et al., 228 (2013)), biological treatment (membrane bioreactors) (Cecconet et al., 2017), adsorption (Kyzas et al., 2015), ozonation (Gomes et al., 2017), microbial fuel cell (Nayak and Ghosh, 2019), wetlands (Ingrao et al., 2020), activated sludge (Modin et al., 2016), and ion exchange processes (Mazur et al., 2018). Though these

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processes are selective to the target compounds, they do not completely remove the pollutants. They require additional post-treatment steps, thus increasing capital and operational expenditure (CAPEX and OPEX). Further, they require large spaced setup (Mahlangu et al., 2022).

On the other hand, membranes processes have grown a reputation of high-quality water production due to their efficient removal of different classes of contaminants (Misdan et al., 2012; Nthunya et al., 2024; Ali et al., 2024) Membranes are selective barriers allowing the passage of fluids (gases or liquids) and retain solutes. They are classified according to their molecular weight cut-off (MWCO) as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO). In water treatment, the feed water contains various pollutants rejected by the membranes according to their size, charge and hydrophobicity. Membranes have been used to produce clean streams from contaminated water. In seawater and brackish water desalination, RO membranes achieve 99 % removal of salts (Park et al., 2012), trace organic compounds (Alturki et al., 2010) and microbes (Yang et al., 2019).

Despite the prolific performance of membranes, their application in water treatment is limited to developed and medium economy countries possibly due to the availability of membrane producing companies, stable power generation capacity and rare to non-existent skills set in the other economic regions though the water quality remain a critical challenge. Consequently, there is an urgent need to develop high-performance membranes with excellent water permeability and good solute retention properties. This served as motivation to modify membranes through various methods. Similarly, application of membranes is limited to fouling reducing water flux (due to additional hydraulic resistance by the cake layer) and solutes rejection. Fouling is the deposition and accumulation of micro- and macro- molecules on the membrane surface leading to the formation of a cake layer. Based on different types of foulants, fouling is categorized as organic, colloidal, inorganic and biofouling. Due to its potential alteration of the membrane properties, fouling is undesirable. To restore decayed flux, the following approaches are available, 1) increase in applied pressure, 2) cleaning of fouled membranes using various methods classified as physical and chemical cleaning strategies and 3) pretreatment of the feed, all of which increase the CAPEX and OPEX despite minimizing the membrane fouling (Mahlangu et al., 2023). Further, some cleaning methods degrade polymeric membranes leading to shorter lifespan. Therefore, recent studies focused on surface modification to increase membrane resistance towards foulant interactions and/or biofilm growth as well as improving their integrity and process performance.

Mostly, membranes are modified using hydroxyl and carboxylated additives to improve surface hydrophilicity. In recent years, research focused on NPs-modification of the membranes to improve process performance and minimize fouling (Tjale et al., 2022; Mpala et al., 2022; Nthunya et al., 2019; Usman et al., 2020; Chimanlal et al., 2023). However, NPs leach from the polymer membrane cause secondary pollution. Further, the toxicity of different NPs has been demonstrated. Therefore, this review explores NP leach from polymeric membranes and strategies to improve their stability. To the best of our knowledge, there is no reported review exploring the NPs leach and improvement strategies ensuring sustainable attachment on membrane surfaces. This study brings awareness of NP leach and toxicity. This will open innovative research methodologies towards improving the stability of NPs in mixed matrix membranes.

2. General surface modification of the membranes

This study reviews the impact of NP leach on sustainable performance of polymeric membranes. Although the study focuses on NPs, other surface modification techniques including the incorporation of copolymers are briefly discussed to present various options towards improvement of membrane functional, mechanical and filtration properties. Also, these copolymers improve stability of the NPs immobilization on the polymeric membrane due to thermodynamic interactions. Further, these polymeric additives are briefly discussed to acknowledge the research progress and/or advancements towards improvement of membrane properties.

Several materials with varying size, charge, and functional groups are used to modify membrane surfaces. These include zwitterionic, amphiphilic, quaternary ammonium, hydrophilic polymers, biopolymers, polyelectrolytes, polydopamine, inorganic salts, and nanomaterials (NMs) (Rana et al., 2006; Rana et al., 2014; Choudhury et al., 2018). These additives are added onto membranes through various mechanisms including surface coating (Shafi et al., 2015) where supplements with adhesive properties (through chemical or physical adsorption) are deposited onto membrane surface (Miller et al., 2017), grafting – immobilization of additives onto membrane surfaces where the functional groups play a key role in forming specific chemical bonds (Liu et al., 2015) and covalent coupling where small molecular weight reagents are attached to the membrane surface through reactive functional groups (Huang et al., 2016). Also, hydrophilic surface modifying macromolecules (LSMMs) have been extensively evaluated to improve the hydrophilicity of various polymeric membranes (Rana et al., 2006; Kim et al., 2009). These LSMMs are prepared from several copolymers including poly(ethylene oxide) (PEO), poly(propylene oxide) (PPO), poly(vinyl alcohol) (PVA), PEG and MDI (Kim et al., 2009). The LSMMs are characterised by different degree of hydrophilicity and charges. Upon modification, the LSMMs render varying properties to polymeric membranes including resistance to fouling and improved process performance. Rana et al. (2005) prepared a hydrophilic surface modifiers composed of MDI and PEG for use to improve hydrophilicity of PES membranes (Rana et al., 2005). The modified PES membranes presented long-term fouling resistance during treatment of a highly polluted river feed water (Rana et al., 2005). In another study, Kim et al. (2009) evaluated the impact of various surface modifying macromolecules (SMM) on hydrophilicity of PES membranes (Kim et al., 2009). The membrane became more hydrophobic upon blending the PES with hydrophobic SMM. However, charged SMM rendered the membrane hydrophilic (Kim et al., 2009).

Each modification approach is processed via different techniques. For example, grafting is achieved through free radical, chemical vapour deposition and atom transfer radical polymerization. Table 1 presents materials used to enhance membrane surface and filtration properties. Oxygen-rich materials are added to improve membrane hydrophilicity and flux. During this process, reduction of foulant attachment and microbial inactivation ensure sustainable performance of the membranes. Similarly, additives are incorporated to improve membrane properties. For example, the addition of silver (Ag) and titanium dioxide (TiO_2) NPs improves photocatalytic and antimicrobial properties of membranes.

3. Nanoparticles: Nature and applications in various industries

The NPs are defined as materials with diameter $\leq$100 nm. They are characterized by high surface area to volume ratio, thus making them useful in various applications. To improve their resistance aggregation, their surface chemistry is modified through functionalization. The structure of the NPs comprises of the surface, shell and core. The shell is formed by a layer of materials that are chemically different from the core material (the centre of the NM). The overall physicochemical properties of the NPs are dominated by the properties of the core, thus determining their toxicity. The core may be complex due to crystal defects, twinning and packing phenomena (Ghosh Chaudhuri and Paria, 2012).

The NPs are characterized by different morphologies including nominally isotropic, rod or wire, tetrapod, tear drop, dumbbell and dendrite structures (Ung et al., 2007). The shapes and structures of metal NPs range from sphere, rod, octagonal, hexagonal, triangle, flower-like, cube, tetrahedron, and nanotubes (Ghosh Chaudhuri and Paria, 2012). They are grouped into various categories namely carbon-based NPs, metal NPs, ceramic NPs, lipid-based NPs, semiconductor NPs, and polymeric NPs. Carbon-based NPs such as carbon nanotubes (CNTs)

Table 1
Membrane surface modification techniques and additives.

Classification	Additive	Base membrane	Modification methods	Foulant/solute	Key features	Ref.
Hydrophilic polymers	Polyvinyl alcohol (PVA)	LE-400 RO membrane	Surface coating	Pseudomonas aeruginosa (*P. aeruginosa*) bacteria, Escherichia coli (*E. coli*) and Bacillus subtilis (*B. subtilis*)	Formation of superhydrophilic and smooth surfaces. The membranes presented anti-adhesion and antimicrobial properties	(Nikkola et al., 2013)
	Hydrophilic surface modifying macromolecules (LSMM) composed of −Methylene bis (phenyl isocyanate) (4,4-diphenylmethane diisocyanate and PEG	Commercial UF polysulfone (PS) membrane	Inter-facial polymerization (IFP)	*E. coli*, humic acid (SH), silica particles (SPs), and chloroform (CF)	The surface of the thin film membranes became more hydrophilic. The membrane was resistant to flux decay due to improved antifouling properties	(Rana et al., 2011)
	Poly-(methacrylic acid) (PMA)	UF polyethersulfone (PES) membranes	Radical grafting	Bovine serum albumin (BSA)	The membranes retained permeability	(Belfer et al., 2000)
	Poly(acrylic acid) (PAA)–silane	Polyamide (PA) thin-film composite RO membrane	Free-radical graft polymerization	Gypsum, alginic acid	Improved wetting properties. resistance to flux and salt rejection decay and gypsum scaling.	(Lin et al., 2010)
	Poly(ethylene glycol) (PEG)	Cellulose acetate (CA) phthalate–polyacrylonitrile (PAN) blend UF membrane	Phase inversion	Bovine serum albumin (BSA)	Resisted deposition of BSA and presented 97 % flux recovery ratio.	(Mondal and De, 2015)
	Sericin	PA thin-film composite RO membrane	Dip-coating followed by in situ cross-linking	BSA	Excellent long-term performance. Rejection and flux were maintained after modification	(Yu et al., 2013)
	Chitosan	Thin-film composite RO membrane	Interfacial polymerization	–	Improved hydrophilicity, smooth surface morphology and enhanced negative surface charge	(Raval et al., 2015)
Dendritic polymers	Polyglycol	Pressure retarded osmosis (PRO) hollow fiber membrane	Surface grafting	BSA, *E. coli* and *S. aureus*	Improved fouling resistance to proteins, microbes, and high flux recovery ratio	(Li et al., 2016)
	Hyperbranched polyglycerol	Thin film composite (TFC) PA membranes	Surface grafting	BSA	Hydrophilicity improvement with high repulsion to protein fouling. Stable water permeability	(Liu et al., 2017)
	Polyethylenimine (PEI)	TFC (TFC) NF membrane	Interfacial polymerization	BSA	Improved selectivity of cations, 90 % flux recovery ratio and formation of smooth membrane surfaces	(Bera and Jewrajka, 2016)
	Poly(amidoamine) (PAMAM)	TFC-RO membrane	Interfacial polymerisation onto and surface coating	BSA	20–25 % increase in water permeability, and resisted protein adsorption	(Nikolaeva et al., 2015)
Quaternary ammonium (QA) polymers	3-allyl-5,5-dimethylhydantoin (ADMH)	Aromatic PA thin-film composite RO membrane	Sequential surface treatment	BSA	Improved surface hydrophilicity, reduced surface negative charge, slightly increased surface roughness, and enhanced salt rejection and water flux. Antifouling properties Improvement	(Zhang et al., 2013)
	Poly (methylacryloxyethyldimethyl benzyl ammonium chloride-r-acryl- amide-r-2-hydroxyethyl methacrylate) (P(MDBAC-r-Am-r-HEMA))	PA-TFC-RO membrane	Free radical polymerization	BSA, *E. coli*	Improved resistance to fouling by BSA and *E. Coli*	(Ni et al., 2014)
Amphiphilic polymers	Polyacrylonitrile-graft-polyethylene oxide (PAN-g-PEO)	Polyacrylonitrile (PAN) UF membrane	Free radical polymerization	*E. coli*	Modification prevented bacteria adhesion. Cleaning at high crossflow velocity	(Adout et al., 2010)

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Table 1 (*continued*)

Classification	Additive	Base membrane	Modification methods	Foulant/solute	Key features	Ref.
					removed adhered bacteria	
	Hydrophilic HEMA and hydrophobic per-fluorodecylacrylate (PFDA) repeating units (HEMA-co-PFDA)		Chemical vapor deposition	BSA	Reduced protein adhesion on the thin films	(Baxamusa and Gleason, 2009)
	Hydrophilic hydroxyethylmethacrylate and hydrophobic perfluorodecylacrylate (PFA)	Thin-film composite PA RO membrane	Chemical vapor deposition	*E. coli*	Increased membrane surface roughness and hydrophobicity. Reduced bacterial adhesion	(Ozaydin-Ince et al., 2013)
	Hydroxyl poly(oxyethylene) methacrylate homopolymer (HPOEM) and zwitterionic carboxylated polyethyeleneimine (carboxylated PEI)	TFC-RO membrane	Surface coating	BSA and alginic acid	Improved antifouling properties during seawater desalination	(Choi et al., 2015)
	Methyl methacrylate-hydroxy poly (oxyethylene) methacrylate (MMA- HPOEM)	Thin film composite (TFC) RO membrane	free radical polymerization and surface coating	BSA and *E. coli*	Resisted fouling by BSA and *E. coli* in seawater	(Choi et al., 2012)
	Triethylenetetramine (TETA) and 2,2,3,4,4,4-hexafluorobutyl methacrylate (HFBM)	PES-UF membranes	Surface grafting	BSA, humic acid, and oil/water emulsion	Improved antifouling performance against organic foulants. Presented 100 % flux recovery	(Zhang et al., 2016)
Thermo-responsive polymers	Polyurethane (TPU) and Poly(n-isopropylacrylamide) (PNIPAM)	Microfiber membrane	Surface coating	1 % oil-in-water emulsion	Switchable superhydrophilicity and super-hydrophobicity as the temperature changes. 99 % oil-in-water emulsion separation efficiency	(Ou et al., 2016)
	Poly(N-isopropylacrylamide-co-acrylamide) (PNIPAM-co-AM) and N-isopropylacrylamide-co-acrylic acid copolymers (PNIPAM-co-AA	Commercial TFC PA RO membranes	Surface coating	BSA	Increased hydrophilicity and reduced fouling compared to pristine membrane	(Wu et al., 2010)
	N-isopropylacrylamide (NIPAm) and acrylic acid (AA)	Thin-film composite aromatic PA RO membranes.	Graft polymerization	BSA	Improved membrane hydrophilicity, negative charge, fouling resistance and salt rejection	(Cheng et al., 2013)
Zwitterionic polymers	Poly(4-vinylpyridine-co-ethylene glycol diacrylate) (p(4-VP-co-EGDA))	TFC-HR RO membrane	Chemical vapor deposition	BSA) and humic acid (HA)	Improved biofouling resistance	(Shafi et al., 2015)
	Poly[2-(dimethylamino)ethyl methacrylate-coethylene glycol methacrylate]	Commercial RO membranes	Chemical vapor deposition and surface coating	*E. coli*	Improved antibacterial properties	(Yang et al., 2011)
	Poly(4-(2-sulfoethyl)-1-(4-vinylbenzyl) pyridinium betaine) (PSVBP)	SW30-XLE (RO membrane)		BSA	Surface hydrophilicity and rejection improved. Resistance to fouling during short-term studies	(Meng et al., 2014)
	Poly[2-[(methacryloyloxy)ethyl] dimethyl-(3-sulfopropyl) ammonium hydroxide]	ESPA-1 RO membrane	UV-assisted radical grafting	Bacteria	90 % decrease in bacteria adhesion	(Marré Tirado et al., 2016)
	Poly(sulfobetaine methacrylate) (pSBMA)	Polysulfone (PSf) support membrane	Surface grafting	BSA	97 % reduction in protein adsorption and 65 % improvement in water flux	(Zhang et al., 2013)
	3-(3,4-dihydroxyphenyl)-l-alanine (l-DOPA)	SW 30 XLE RO membrane	Surface coating	BSA and alginic acid	Improvement in surface hydrophilicity and water flux. Reduction in foulant adsorption	(Azari and Zou, 2012)
	Polysulfobetaine	SW 30 HR and AD RO membranes	Surface coating	Bacteria	80 % reduction in microbial adhesion. No change in salt rejection and water permeability	(Ginic-Markovic et al., 2015)
Biomimetic adhesive agent: polydopamine	Polydopamine	XLE RO membrane, NF90 NF membrane, PSf A1 UF membrane and PP and PTFE MF membranes	Surface coating	Oil/water emulsion	Intrinsic pure water properties were retained, while fouling resistance improved	(McCloskey et al., 2012)
	Polydopamine	XLE RO membranes	Surface coating	Oil/water emulsion	Improved flux and resistance to fouling	(Kasemset et al., 2013)
Fluoropolymers, polydimethyl	Fluorinated polyamine	PA NF membrane	Surface coating	BSA	Reduction in membrane surface energy, low	(Dizaj et al., 2014)

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Table 1 (*continued*)

Classification	Additive	Base membrane	Modification methods	Foulant/solute	Key features	Ref.
	siloxane and ionic liquid				fouling and 98.5 % flux recovery ratio	
	1-butyl-3-methylimidazolium chloride ([BMIM]Cl) showed	Aromatic PA membranes	Surface coating	Colloids and organic matter	20 % increase in flux, while salt rejection decreased slightly. Decrease in membrane roughness and improved fouling resistance	(Meng et al., 2014)
Metal and metal oxide NPs	Copper (Cu) NPs	PA-TFC-RO membrane	Layer by layer assembly	*E. faecalis* and *E. coli*	Minor flux decline upon modification. 94–100 % bacteria inactivation.	(Ma et al., 2016)
	Ag NPs	SW 30 XLE	*In situ* formation	*E. coli*	Slight reduction in water permeability while salt selectivity, and hydrophilicity improved. 75 % antimicrobial efficiency	(Ben-Sasson et al., 2014)
	Tannic acid–ferric ion–polyethylenimine (TA–Fe–PEI) and Ag NPs	PA RO membrane	Chemical deposition and in situ formation	*E. coli* and *B. subtilis*	Increase in membrane hydrophilicity, salt rejection and flux. 100 % mortality of microbes. The NPs were stable	(Dong et al., 2017)
	TiO_2 NPs	BW30 RO membrane	UV-irradiation surface coating	Reactive red 261 dye and BSA	Enhanced flux and dye removal. Improved fouling resistance to BSA	(Ngo et al., 2016)
	TiO_2 (Polydopamine used as binder)	PA TFC membrane	Self-assembly		Polydopamine increased membrane hydrophobicity and reduced flux.	(Zhang et al., 2013)
Mineralizing inorganic salts	$Cu(OH)_2$ NPs	ES-20 RO membrane	Dip-coating (adsorption)	*E. coli* and *P. putida*	Good antibacterial activity after modification	(Karkhanechi et al., 2013)
	$Zn(OH)_2$ NPs	PSf-UF membranes	Polymerization and surface coating	BSA	Smooth and hydrophilic membranes with improved flux and antifouling properties were formed.	(Wu et al., 2016)
Carbon based NPs	Graphene oxide (GO) nanosheets	PA- TFC membrane	Layer-by-layer assembly	BSA	Improvement in fouling resistance due to membranes becoming smoother and more hydrophilic	(Choi et al., 2013)
	Azide functionalized graphene oxide (AGO)	PA XLE RO membrane	Surface grafting	*E. coli*	Water permeability and salt rejection was maintained. Membranes became smoot and more hydrophilic. 17-fold reduction in biofouling after 24 h of exposure to *E. coli*	(Huang et al., 2016)
	GO nanosheets	TFC-PA membranes	Surface functionalization	*E. coli*	65 % bacterial inactivation after 1 h of exposure. No change in membrane intrinsic properties after modification	(Perreault et al., 2013)
Polymer/NP hybrid materials	GO nanosheets, gold nanostars (AuNS), and hydrophilic polyethylene glycol (PEG)	BW30 RO membrane	Interfacial polymerization	$CaCO_3$, $CaSO_4$, HA and *E. coli*	Reduced scaling, organic fouling, and biofouling after membrane modification.	(Ray et al., 2015)
	Ag and PEG	TFC membrane	Interfacial polymerization	*E. coli* and BSA	Reduced foulant attachment after modification	(Zhang et al., 2013)
	PA–Cu–GO complex	TFC membranes	Chelation and surface reaction	BSA	Improved water flux, salt rejection and chlorine attack resistance	(Ali et al., 2016)
	GO–poly L-lysine (PLL)	PA-TFC-FO membrane	Layer-by-layer assembly	Bacteria	Smooth and hydrophilic membranes were formed with antibacterial properties. Flux restoration by water cleaning	(Hegab et al., 2015)
	Polydopamine–Zn complex	The PSf-UF membranes	Polymerization and surface coating	BSA	High water flux and antifouling properties	(Wu et al., 2016)

have gained interest in various applications due to their electrical conductivity, high strength, electron affinity as well as adaptability. They are classified as single-walled, double-walled or multi-walled. Metal NPs such as Cu, Au and Ag are purely derived and prepared from metals and ions and they have a wide adsorption band in the visible region (Khan et al., 2019). Ceramic NPs are made from inorganic, non-metallic materials and they possess excellent heat resistance and durable characteristics. Ceramic NPs are mainly used as coatings, catalysts, and batteries (Morsi et al., 2022). Semiconductor NPs possess properties like non-metals and metals. Due to their absorption and emission properties, they are used to make electronic devices, solar cells, in cancer therapy, and bio imaging (Nguyen et al., 2013).

The NPs play different roles in various applications due to their diverse characteristics. For these reasons, they are used in different industries such as, medical, environmental, agricultural, food, electronics, automotive and defence industries. Table 2 presents detailed applications of NPs in the various industries.

4. Nanoparticle-surface modification of membranes

As previously highlighted, membrane surface modification is performed to improve water permeability, solute rejection efficiency and fouling resistance. In some cases (e.g., the addition of NPs), membranes are engineered to induce other functionalities such as photocatalytic and antimicrobial activity.

4.1. Membrane polymers and polymer-nanoparticle compatibility

The most widely used polymers for membrane fabrication are slightly-hydrophobic in nature, typically due to good chemical stability. However, hydrophobic membranes present poor filtration performance. These polymers include polyvinylidene fluoride (PVDF) (Lin et al., 2022; Chen et al., 2020; He et al., 2020), polysulfone (PSf) (Zhang et al., 2019; Rodrigues et al., 2019; Mamah et al., 2021); polyethersulfone (PES) (Kim et al., 2010; Giwa and Hasan, 2020; Mansor et al., 2021; Johari et al., 2021) polyacrylonitrile (PAN) (Valamohammadi et al., 2020; Bai et al., 2022; Jang et al., 2020), polypropylene (PP) (Shao et al., 2019; Li et al., 2020; Alammar et al., 2020); cellulose acetate (CA) (Vetrivel et al., 2019; Zamel et al., 2019; Ounifi et al., 2022), PA (Huang et al., 2021; Cheng et al., 2021; Liu et al., 2019), polydimethylsiloxane (PDMS) (Khairkar et al., 2020; Ariati et al., 2021; Li et al., 2019), and polytetrafluoroethylene (PTFE) (Yadav et al., 2022; Cao et al., 2022; Zhang et al., 2020). Membranes prepared from these polymers exhibit variations in permeability, selectivity, and stability in water sources due to their differences in chemical characteristics and structure. Therefore, modification is carried out to enhance performance and selectivity.

During preparation of mixed matrix membranes, it is important to establish excellent compatibility between the polymer and NPs. However, polymer-inorganic filler miscibility poses a serious challenge preventing the performance and stability of nanoparticle-enhanced membranes. To improve polymer and NPs compatibility and/or miscibility, specific interactions between the different components should be established. However, some studies reported miscible systems in the absence of specific interactions (Rana et al., 2000). This is achieved through a blend of different polymers resulting in a net exothermic combination. Under these circumstances, similar and dissimilar contact energies are nearly equivalent, thus neglecting the impact of van der Waals interaction energies and the heat of mixing (Rana et al., 1996). Furthermore, the equation-of-state effect is small, thus outweighed by the combinatorial entropy when the components have approximate similarities in chemical structure and functional groups (Bhattacharya et al., 1989). Also, the intra- and inter-molecular interactions increase compatibility, thus strengthening van der Waals interactions when the polymers are in contact (Roland et al., 1993). Affinity parameters for a mix of copolymer X constituting of monomer residues A and B with a homopolymer Y comprising monomer residue C can be predicted from

Table 2
Industrial applications of various nanoparticles.

Industry	Category	Function
Environment	Bioremediation	AgNPs, CNTs, nanoscale zeolites, FeNPs and metal oxides are used to pollutants from the soil (Das et al., 2019). Similarly, nanoclays emerged as effective adsorbents towards environmental remediation of pesticides (Lousada et al., 2023).
	Sensors	Au NPs (Theron et al., 2010) are used to improve water quality and environmental clean-up (Pradeep, 2009), and detect microbial pathogens and biological compounds (Yadav et al., 2010).
	Catalysts	Pt NPs (Lam and Luong, 2014) and Hg NPs (Kora and Rastogi, 2018) are used as catalysts in chemical reactions including biomass conversion into fuels (Lam and Luong, 2014).
Medicine	Drug delivery	Au NPs (Siddique and Chow, 2020), ZnO NPs (Anjum et al., 2021), Cu NPs (Yuan et al., 2018) and Ag NPs (Schröfel et al., 2014) are used as drug delivers towards treatment of various diseases such as cancer and bacterial infection (Siddique and Chow, 2020).
	Diagnosis	Au NPs (Siddique and Chow, 2020) and Fe_3O_4 NPs (Nguyen et al., 2013) are used as imaging agents to visualize specific body areas.
	Tissue engineering	TiO_2 NPs (Kim et al., 2014) are used to stimulate tissue and organ growth and repair.
	Antimicrobials	Cu NPs and Ag NPs (Hoseinzadeh et al., 2017) are applied in wound dressing and medical devices due to their antimicrobial properties
Agriculture	Pesticides and herbicides	Ag NPs and Cu NPs (Hoseinzadeh et al., 2017; Kapinder and Dangi, 2020) are used as pesticides and herbicides towards targeted treatment (Khan et al., 2019).
	Fertilizers	Ag NPs (Jaskulski et al., 2022), Zn NPs (Song and Kim, 2020), Cu NPs, Au NPs, Al NPs and Fe NPs (Khan et al., 2021) improve plant mineral nutrition and delivery of fertilizer more efficiently.
	Food safety	Various NPs are used to detect and eliminate pathogens from food products (Das et al., 2019)
	Water purification	Various NPs such as Cu, Fe, Ag are used in water purification systems (Das et al., 2019)
Food industry	Food processing, preservation, and packaging	Ag NPs (Kapinder and Dangi, 2020) are used to improve performance of food processing operations e.g., grinding, mixing, and drying. Due to their cytotoxic effects, CuNPs are used in food packaging (Nthunya et al., 2023).
	Food fortification	Cu NPs and Fe_2O_3 NPs (Khan et al., 2021) deliver essential nutrients (vitamins and minerals) to food products.
	Sensors	Various NPs such as Pt and CNTs are used in detection of NO and food spoilage respectively (Yadav et al., 2010).

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Table 2 (*continued*)

Industry	Category	Function
Electronics and automotive	Display and storage	Ag NPs and Au NPs (Gwynne, 2020) are used to improve performance electronics such as liquid crystal display (LCD) (Bahadur et al., 2021).
	Data storage	Fe NPs, Co NPs and Ni NPs (Ahmad et al., 2021) improve capacity and speed of data storage devices (Ahmad et al., 2021).
	Chemical processing and catalysis	Pt NPs (Bhavani et al., 2021), Fe NPs (Jiang and Xu, 2011) and Ni NPs (Salem and Fouda, 2021) act as catalysts in chemical reactions (e.g., fuel cell, hydrogenation, and oxidation reactions) (Lara and Philippot, 2014)
	Separation and purification	Fe_2O_3 NPs (Siddique and Chow, 2020), Ag NPs (Pradeep, 2009), Au NPs (Siddique and Chow, 2020) and Al NPs (Das et al., 2019) are used in separation and purification of gases and liquids (Hollamby et al., 2010).
Defense	Sensors	Curcumin NPs improve sensitivity and specificity of sensors used improve defense mechanisms by detecting threatening signals (Zheng et al., 2010).
	Protective coatings	Al NPs and Zn NPs (Rangel-Olivares et al., 2021) are used to improve performance and durability of protective coatings used in defense equipment.
	Weapons	Ceramic and metal NPs (Ye et al., 2020) are used to make weapons against microbes (bacteria and viruses) and development of armour and protective equipment (Ye et al., 2020)
	Manufacturing	Ceramic and metal NPs (Ye et al., 2020) improve the performance and durability of materials used in defense equipment.
	Energy storage	MnO_2, $LiCoO_2$ NPs and Fe_2O_3 NPs (Morsi et al., 2022) improve performance and efficiency of batteries and fuel cells used in defense systems (Morsi et al., 2022).

the following relationship (**Equation (1)**:

$$X_{blend} = X_{AC}\varnothing_A + X_{BC}\varnothing_B - X_{AB}\varnothing_A\varnothing_B \quad (1)$$

where $\varnothing_A$ and $\varnothing_B$ account for the volume fraction of the monomer residues A and B, respectively in the copolymer. X_{AC}, X_{BC} and X_{AB} are the interaction parameters between monomer residue A, B and C. When $X_{blend} < X_{critical}$, the mixing is compatible. For polymer–polymer systems, X_{blend} is approximately zero.

The miscibility and interactions between polymers and NPs has not been well reported. However, it is important to infer the compatibility of polymer−filler mixtures. In the case of NP-engineered membranes, these interactions determine the dispersion of NPs on the polymer matrix (thermodynamic compatibility).

4.2. Nanoparticle-enhanced polymeric membranes

Hydrophilic nanomaterials are incorporated into pristine membranes to enhance water uptake properties. For example, Mahlangu et al. (2017) reported an increase in water flux upon incorporation of GO-ZnO on PES membranes (Mahlangu et al., 2017). In another study, Ma et al. (2020) improved water flux of hydrophobic PVDF membranes through addition of GO and PEG (Ma et al., 2020). In both instances, the solute rejection was not affected. Similarly, the separation efficiency and selectivity of membranes is enhanced through modification of the membrane surface properties. Upon treatment of oil/water emulsions, super hydrophilic or super oleophilic properties are desired to improve separation efficiency. Qing et al. (2020) improved hydrophilic properties of polyvinyl alcohol (PVA) nanofibers membrane through incorporation of SiO_2NPs (Qing et al., 2020). The nanoengineered membranes presented excellent separation of oil and water mixtures. Further, separation of surfactant-stabilized oil-in-water emulsions was achieved under gravity. In a similar study, Tjale et al. (2022) incorporated SiO_2NPs into PSf/PP membrane, resulting in improved oil flux while the water was retained (Tjale et al., 2022). This was attributed to formation of oleophilic/hydrophobic upon addition of SiO_2 NPs. Oil permeation through the oleophilic membrane was ascribed to the negative intrusion pressure (Zhou et al., 2018).

Interestingly, the incorporation of NPs improves retention of trace organic compounds (Mahlangu et al., 2017; Patala et al., 2022) and salts due to introduction of efficient removal mechanisms (Bera and Jewrajka, 2016; Dong et al., 2017). For instance, photocatalytic NMs with improved degradation properties have been anchored onto membrane surfaces towards improved dyes (Phan et al., 2020; Kadhim et al., 2020; Nthunya et al., 2024) and organic compounds removal (Zheng et al., 2017; Krishnan et al., 2022). Photocatalytic membranes are used to perform filtration and photodegradation simultaneously. A PVDF membrane was modified with bismuth tungstate (BWO) for the degradation of pirimicarb (Krishnan et al., 2022). The membrane achieved 81 % photodegradation of the pesticide at pH 5. Numerous studies reported the impact of NPs modification towards reduced membrane fouling and improved antimicrobial properties (Nikkola et al., 2013; Zhang et al., 2013; Patala et al., 2022; Mpala et al., 2023; Sisay et al., 2023; Al-Ani et al., 2020). Alsalhy et al. (2020) reported a reduced cake layer build-up (10.42 μm thickness) on the surface of a polyvinyl chloride (PVC) membranes modified with ZnO nanoparticles compared to the pristine membrane (52.8 μm thickness) (Al-Ani et al., 2020). In another study, multiple nanoparticles (TiO_2, CNTs and $BiVO_4$) were used to improve fouling resistance of PVDF membranes during dairy wastewater treatment (Sisay et al., 2023). Fouling prevention was attributed to improved membrane surface smoothness and hydrophilicity. Hydrophilic properties induce formation of hydration layer, thus reducing membrane-foulant interactions (Stanley and Rau, 2011; Miao et al., 2017). Similarly, antibacterial NPs inhibit growth of bacteria on membrane surfaces (Kim et al., 2012). For this reason, NP enhanced membranes have been applied to purify hospital (Alsalhy et al., 2018), refinery (Al-Ani et al., 2020), dairy wastewaters (Sisay et al., 2023) and other various types of water with complex matrixes. Fig. 1 demonstrates improvement in membrane physical and filtration properties before and after NP modification.

Although surface modification is a viable solution, it possesses specific challenges. Nikkola et al. (2013) reported a decrease in membrane water permeability of TFC-RO membranes after polyvinylalcohol (PVA) modification (Nikkola et al., 2013). Also, fouling prevention is selective to specific foulants (Liu et al., 2017; Dizaj et al., 2014; Chimanlal et al., 2022). Technically, antimicrobial membranes are fouled by colloids and sparingly soluble salts through increased concentration polarization (CP). According to experimental results, modified membranes foul after long-term exposure to real water samples. Another challenge is trade-off between permeability and selectivity. As per Saraswathi et al. (2019), water flux and fouling resistance were enhanced through incorporation of hydrophilic polydopamine and antibacterial AgNPs on the membrane surface (Saraswathi et al., 2019). In this work, the additives formed a thin film on the membrane surface and functioned as a barrier layer. However, NPs leached from the membranes causing secondary contamination. In addition to leach from the polymer matrix, majority of reported NPs synthetic methods are environmentally unfriendly due to

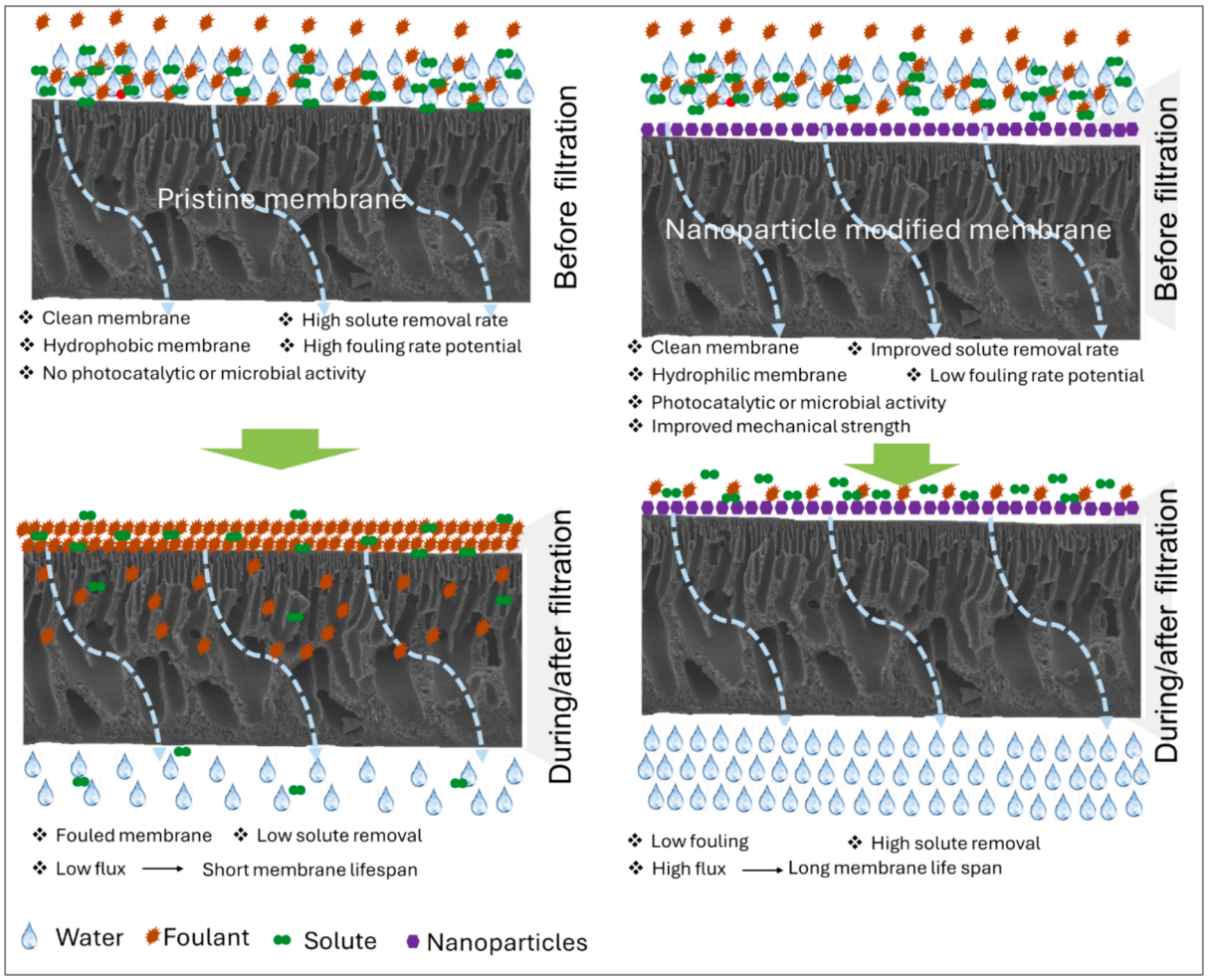

Fig. 1. Effects of NPs on membrane physical and filtration properties.

the usage of toxic chemicals such as strong acids (sulfuric acid) and bases (ammonium hydroxide).

5. Nanoparticle leach, measurement and influencing factors

The NPs are released to the environment during filtration and/or membrane cleaning. This is influenced by the abrasion on the membrane surface caused by the shear cross-sectional force, permeate drag and percolation.

5.1. Measurement of nanoparticle leach during fabrication, application and chemical cleaning

The rate of NPs leach is measured at various stages including preparation of the NP-modified membranes and application. The release of NPs during application has been studied in both short-term and long-term experiments (Ma et al., 2024; Jackson et al., 2024; Yasir et al., 2024; Kajau et al., 2021). In polymeric membranes, NPs are often added in small quantities <1 wt% to prevent NP agglomeration and membrane pore blocking at higher concentrations. For this reason, leaching of NPs at trace levels requires instrumental measurements such as inductively coupled plasma mass spectroscopy (ICP-MS) and/or inductively coupled plasms optical emission spectroscopy (ICP-OES) (Mpala et al., 2023). Also, atomic absorption spectroscopy (AAS) and UV–vis spectrometry have been actively used to measure the concentration of AgNPs leach from the membranes (Liu et al., 2013; Nthunya et al., 2019). Although NP leach has been a concern for years, there are still no standardized procedures for quantification and reporting guidelines. As a result, some studies have reported leaching rate (e.g., $\mu g \cdot m^{-2} \cdot h^{-1}$) (Kajau et al., 2021), while others have reported the total concentration of leached NPs (e.g., $\mu g \cdot L^{-1}$) (George and Kumar, 2023).

Upon membrane preparation, NPs are released from the membrane matrix to the non-solvent coagulation bath. To demonstrate NP leach during preparation, Huang et al. (2014) measured the concentration of AgNPs in the water coagulation bath using ICP-MS (Huang et al., 2014). The casting solution was immersed into a coagulation bath with known volume of ultrapure water. The cast membrane was removed from the coagulation bath and the NP leach was measured from the coagulation water bath. The NP loss rate was estimated based on the ratio of the NP concentration in the coagulation bath to the total NP amount in the casting solution used for membrane preparation. Zero release of AgNPs was reported due to the dense layer of the membrane and high affinity between the Ag and the SiO_2. In another study, Samavati et al. (2024) evaluated the release of AgNPs during membrane fabrication, where the residue from the coagulation bath was analyzed for the presence of AgNPs using ICP-MS (Samavati et al., 2024). The amount of AgNPs released from the membrane was 0.003 ppm.

In other studies, NP leach was reported during membrane filtration (Samavati et al., 2024; Andrade et al., 2015). In this case, test samples were collected from the permeate for ICP measurement. For example, ultrapure water was filtered through Ag-SiO_2 modified membranes in a dead-end filtration cell and the permeate samples were collected at predetermined time intervals over a 3 h period (Huang et al., 2014). ICP-MS analysis presented membrane resistance to NP leach during

filtration. Similarly, Makhetha and Moutloali (2021) studied NP release (AgNPs and CuNPs) from polymeric polyethersulfone membrane by performing dead-end filtration for 240 min (Makhetha and Moutloali, 2021). The highest amounts of AgNPs and CuNPs released from the membranes were 0.573 and 0.134 mg respectively. In another study, NP leach was investigated during membrane application in a cross-flow system over a 72 h period (Damavandi et al., 2023). The leaching of TiO_2 nanoparticles was ascribed to the weak physical adhesion of the NPs. Evaluation of the NPs leach from the permeate indicate the possible toxicity of the treated water to the end user. However, NPs can leach into the concentrate which often needs further treatment.

The investigation of NP leach has also been conducted in batch method where known dimensions of membrane coupons are submerged in the test liquid (often deionized water). This is followed by shaking and collection of samples at selected time intervals (Iqbal et al., 2022; Haider et al., 2016). Using this immersion technique, Makhetha and Moutloali (2021) demonstrated leaching of AgNPs and CuNPs from polyethersulfone membranes (Makhetha and Moutloali, 2021). The highest amounts of leached AgNPs and CuNPs were 0.579 and 0.192 mg respectively. Liu et al. (2013) used a unique approach (quartz crystal microbalance with dissipation, QCM-D) to study the kinetics of Ag leach (Liu et al., 2013). The QCM-D experiments were conducted with a cross-flow rate of 100 $\mu L \cdot min^{-1}$ and deionized water was used. Although QCM-D approach provides evidence of NP release, this method does not represent the actual dissolution scenario during membrane filtration because water flows only on the membrane surface in QCM-D setup.

Despite fouling prevention being one of the reasons for incorporating NPs into membranes, the membranes eventually foul (although at a lower rate compared to the pristine membrane) and require cleaning. Depending on the fouling type, chemical cleaning is used to restore membrane flux. However, membrane degradation caused by harsh chemicals or different pH values has been documented (Kajau et al., 2021; Banerjee et al., 2024). Therefore, recent studies have evaluated NP leach due to exposure to different membrane cleaning chemicals. Jackson et al. (2024) studied the effects of membrane exposure to $NaHCO_3$ at pH 3 on the stability of Ag NPs (Jackson et al., 2024). Reportedly, The NPs leached due to prolonged exposure to $NaHCO_3$. Similarly, Kajau et al. (2021) demonstrated leaching of CuO NPs from PES membranes due to prolonged exposure (840 h) to different wash solutions (Kajau et al., 2021). Based on ICP-MS results, sodium chloride, hydrochloric acid, sodium hydroxide and sodium hypochlorite induced the NP leaching rates of 29, 218, 72, and 90 $\mu g \bullet m^{-2} \cdot h^{-1}$ respectively.

5.2. Nanoparticle leach from polymeric membranes

The research focus has shifted to investigating the release of NPs from polymeric membranes. This is mainly due to environmental concerns of NP leach causing secondary pollution. These concerns involve toxicity of NPs requiring in-depth investigation. Based on previous studies, NPs are introduced into polymeric membranes to improve their filtration and mechanical properties (Nthunya et al., 2019; Saleem and Zaidi, 2020; Nthunya et al., 2020). However, their leach to the environment introduces pollutants of emerging concern. Herein, the studies reporting on NP leach are summarized.

Saraswathi et al. (2019) reported leaching of Ag NPs from UF membranes due to high hydraulic pressure during water treatment (Saraswathi et al., 2019). However, the concentration of the leached NPs was lower than the maximum acceptable limit of silver contamination in water. The NPs leach during membrane cleaning is accelerated by the susceptibility of polymers to the harsh cleaning conditions. Kajau et al. (2021) exposed CuO-modified PES-UF membranes to different cleaning solutions comprising of sodium chloride (NaCl), hydrochloric acid (HCl) and sodium hypochlorite (NaOCl) (Kajau et al., 2021). After 840 h of exposure, the NPs leached, thus compromising the membrane integrity. Another study monitored a daily leach of AgNPs from modified PES membranes under different filtration conditions (Ghalamchi et al., 2019). Based on dissolution, desorption, passive diffusion, and polymer matrix composite degradation mechanisms, the concentration of leached NPs increased as a function of time. Due to degradation of polymeric membranes in extreme conditions, microplastics and nanoplastics are detected in the water bodies at high concentrations (Kajau et al., 2021). According to Ding et al. (2020), nanoplastics and microplastics are released from membrane systems due to physical flushing, chemical agents, mechanical stress, aging, wear and tear (Ding et al., 2020). Detailed leaching of NPs from the membranes is presented in Table 3.

5.3. Factors influencing nanoparticle leach from the membranes

The NPs leach from membranes is influenced by various factors including capping materials (e.g. polymers), nanoparticle loading concentration, operational conditions and chemistry of the cleaning solution. Based on size (NPs smaller than 1.33 nm^3), leaching is influenced by diffusion within the polymer matrix (Ghalamchi et al., 2019). Also, the concentration of NPs influences their leaching rate. According to Saraswathi et al. (2019), membranes coated with 10 mM $AgNO_3$ presented higher initial Ag leach rate (2.5×10^{-2} mg/L) compared to similar membranes coated with 5 mM $AgNO_3$ (Saraswathi et al., 2019). The initial leach rate of the latter was 1.2×10^{-2} mg/L. Other studies reported considerable leaching of Ag caused by high NPs loading (Basri et al., 2010; Basri et al., 2011; Yu et al., 2003; Zodrow et al., 2009; Cao and Liu, 2010; Chou et al., 2005). On one hand, high loading promotes poor support of NPs on the membrane, thus increasing initial rate of leaching, while on the other, material compatibility affects NPs stability. Due their swelling behaviours, hydrophilic additives coated on hydrophobic materials present certain level of leaching (Li et al., 2014). Also, oxygen promotes NPs leach due to formation of weakly bound metal oxides (Ghalamchi et al., 2019).

During filtration, freely bound NPs are dislodged from the membrane surface and macro-void channel due to the shear stress and hydraulic pressure caused by the water flow (Saraswathi et al., 2019; Mpala et al., 2023). In adsorption studies, NPs are discharged into the solution due to the agitation to promote solution homogeneity, mass transfer and the pulling force toward solute molecules (Phan et al., 2020). This pulling effect loosens the adherence between the particles and the polymeric membranes. Likewise, the mode of membrane application plays a role on NPs leach. Bi et al. (2018) subjected PA-RO membranes incorporated with AgNPs under different leaching tests; namely batch immersion, dead-end filtration, cross-flow filtration and water jetting (Bi et al., 2018). Batch immersion presented highest amount of Ag leach (1.54 $\mu g/cm^2$). Likewise, dead-end filtration presented the lowest amount of Ag leach (0.044 $\mu g/cm^2$). The cross-flow and water jetting presented 0.34 $\mu g/cm^2$ and 0.92 $\mu g/cm^2$ AgNP leach, respectively. The other parameters influencing NPs leach include properties of the host substrate (i.e., the presence of functional groups on the membrane surface) and characteristic NP-polymer interactions. Ng et al. (2020) compared the impact of different polymers on leaching of GO from film nanocomposite (TFN) membranes (Ng et al., 2020). Reportedly, PA-TFN membranes presented lower GO leach compared to PSf-TFN membranes. Nonetheless, leaching stopped after 2 days for all membranes. This was linked to a strong interaction between the remaining GO and TFN membranes.

5.4. Effect of nanoparticle leach on membrane performance

The leaching of NPs compromises the physical, chemical and performance characteristics of the membrane. Upon exposing CuONPs-modified PES-UF membranes to 0.1 M NaCl, 0.1 M HCl, 0.1 M NaOH and 0.5 % NaClO, NPs leach enlarged membrane microvoids, thus increasing membrane surface roughness and hydrophobicity (Kajau et al., 2021) (Fig. 2). The findings alluded to the effect of chemical cleaning on the mechanical properties of the polymer. During cleaning, the selective layer swells resulting in the relaxation of undulated surface

Table 3
Nanoparticle leach from polymeric membranes.

Membrane and additive	Description	Leaching behavior	Ref.
Poly (ether imide) UF membranes coated with Ag and polydopamine	The modified membranes were immersed in 2 M HCl for 10 min under ultrasonication	2.5 x10^{-2} mg/L Ag leached from the membrane. The leaching rate decreased as a function of time	(Saraswathi et al., 2019)
Chitosan-sodium alginate multilayer membrane modified with $FeWO_3$NPs	The modified membranes were soaked in distilled water at 25 °C for 14 d.	56.8 μg/L iron leached	(Kazemi et al., 2018)
PES membranes embedded with Ag_3PO_4/g-C_3N_4 NPs	The membranes were placed in 250 mL deionized water at 45 °C for 3 weeks. Ag leach was monitored daily	The Ag leach increased in the first days and stabilized at 11 % after 4 days	(Ghalamchi et al., 2019)
The PES membrane modification using sulfonated poly (aryl ether sulfone) and CuNPs blend	NP leach was evaluated through filtration experiments. The Cu leach was verified by measuring its presence in the permeate	160 μg/L Cu was measured in permeate of the UF membranes at the start of the experiment. The leaching decreased as a function of time presenting 70 μg/L Cu after 12 h of filtration	(Zhang et al., 2018)
PVDF nanofibre membrane modified with AgNPs, SiO_2NPs and MWCNTs	AgNPs leach was evaluated from the permeate solution of the membrane distillation processed water	AgNPs are tightly bound on the PVDF nanofibre membranes with minimal leaching rate, and thus demonstrating their stability.	(Nthunya et al., 2019)
PA-RO membrane modified with AgNPs	Leaching was investigated through 72 h batch immersion, 4 h dead-end filtration, 50 h cross-flow filtration and 1 h water jetting	Ag NPs leached from the membranes as follows: 1.54 μg/cm^2 from batch immersion, 0.044 μg/cm^2 from dead-end filtration, 0.34 μg/cm^2 from cross-flow filtration and 0.92 μg/cm^2 from water jetting	(Bi et al., 2018)
The PVDF membranes incorporated with Fe_3O_4NPs	The membrane was immersed in a beaker containing 12 M HCl for 5 min and deionized water for 3 weeks	NPs leaching resulted in structural changes of the membrane	(Agbaje et al., 2018)
The PVDF membranes were modified with perfluorosulfonic acid (PFSA) and SiO_2NPs	Leaching of NPs was investigated by exposing the membranes in distilled water under stirring	SiO_2NPs easily leached from PVDF/PEG/SiO_2 hybrid membrane under stirring conditions	(Zhou et al., 2018)
Zwitterion-Ag nanocomposite modified thin-film FO membrane (ZTFC-Ag)	The Ag leaching experiments were performed in batch mode over 12 days	The leach rate of Ag ions was rapid during the first 4 days (~2.1 μg/cm^2/day) and thereafter slowed down to reach a steady value	(Qiu and He, 2018)
PSf-UF membranes were modified with AgNPs via dip-coating approach	Leaching of Ag ions was measured during a dead-end filtration at 1.0 bar for 24 h.	The Ag leach was 2.7 μg/L at the start of the experiment. This declined rapidly 0.6 μg/L after 5 h and thereafter stabilized at 0.4 μg/L	(Qi et al., 2018)
Poly(vinyl alcohol-co-ethylene) nanofibrous membrane decorated with AgNPs	The Ag leach was performed by filtering 1.3 L deionized water through the membrane for 30 min at 0.10 MPa pressure. Also, the membrane was immersing into 100 mL of pure water under 100 rpm agitation 7 days.	13 μg/L Ag leached from the membrane polymer matrix after 30 min of filtration. Similarly, agitation caused leaching of 21 μg/L Ag	(Cheng et al., 2020)
Hydrophilic microporous PES UF membrane containing CuONPs	The membranes were immersed in milli-Q water for 120 h	5 ng/L CuONPs leached from the polymeric membrane	(Nasrollahi et al., 2019)
PVDF and PES membranes incorporated with AgNPs	Leaching of AgNPs was investigated during 7 d membrane filtration	Ag NPs leached from the PES membrane in the concentrations of 130.9, 77.8, and 36.8 μg/L on day 1, 3, and 7, respectively. Similarly, leaching from PVDF membranes, 66.18, 44.4 and 22.4 μg/L AgPs on similar days	(Amouamouha and Badalians Gholikandi, 2018)
The PVDF membranes modified with $CuAl_2O_4$ spinel and CuONPs	The membrane was soaked into 20 mL of deionized water at 25 °C followed by agitation at 50 rpm for 24 h	The Cu^{2+} ion concentrations were 19.83 ± 0.27 mg/L and 1407.06 ± 5.44 mg/L in the $CuAl_2O_4$ spinel and CuO leachates respectively	(Wang et al., 2019)
Nylon membranes coated with MnOOHNPs	The membranes were immersed in deionized water for 3 d	0.02 mg/L MnOOHNPs leached	(Zhang et al., 2021)
Ultrafiltration membrane with polyampholyte hydrogel and GONPs	The modified membrane coupons were submerged in 3 mL of salt solution (8.5 mM NaCl and 1.5 mM $CaCl_2$) and agitated at 100 rpm for 7 days	Leaching of GO was exceptionally low, with over 98 % of the GO remaining on the membrane surface after 7 d	(Zhang et al., 2018)
Pd/FeNPs integrated poly (methacrylic acid) (PMAA) PVDF membranes	Contaminated groundwater samples (200 mL) were pressurized through the membranes at 1 bar and the permeate was	2 % Fe and Pd were leached	(Wan et al., 2020)

(continued on next page)

Table 3 (*continued*)

Membrane and additive	Description	Leaching behavior	Ref.
	assessed for Pd and Fe nanoparticles		
AgNPs-incorporated thin film nanocomposite (Ag-TFN) membranes	The Ag-TFN membrane was immersed in deionized water (20 mL) and stirred at 100 rpm	Ag NPs leaching rate decreased sharply 0.2 μg/cm^2/d to a steady rate of 0.008 μg/cm^2/d after day 4	(Jeon and Lee, 2020)
PES membrane modified with Ag_3PO_4/ZnAlCu nanolayered double hydroxide (NLDH)	The composite membrane was socked in 100 mL of deionize water under stirring condition	The leach rates of Ag and Cu were 0.4 mg/L and 0.7 μg/L respectively	(Ghalamchi et al., 2019)

topological features. In the same manner, hydrophilic membranes have high water uptake resulting in swelling effect. This leads to structural rearrangements and changes in the surface roughness of the membranes (Zhang et al., 2020; Guo et al., 2021). The NP leach increases membrane hydrophobicity due to the removal of hydrophilic additives (Kajau et al., 2021). Other studies reported an decrease in membrane resistance and mechanical strength caused by increased pore size and porosity due to hypochlorite aging (Rabuni et al., 2015; Abdullah and Bérubé, 2013). Reduced membrane integrity promotes NPs leach.

Apart from physical and chemical changes, NPs leach increases water permeability due to deformation of the membrane microvoids, increase in membrane porosity and roughness (Puspitasari et al., 2010). Further, NPs leach reduces solute removal due to changes in size exclusion factor caused by increase in membrane microvoids (Kajau et al., 2021; Abdullah and Bérubé, 2013). In other instances, NPs leach decrease membrane performance due to reduction in fouling resistance, photocatalytic and antimicrobial activities. These effects require extensive research to ascertain their impact on process performance.

6. Toxicity of nanoparticles

Although unique properties of NPs enable their usage in textiles, biomedical, food packaging and water treatment (Tang et al., 2011; Chaloupka et al., 2010), their leach increases toxicity to the environment due to their high chemical reactivity. The NPs react with biological systems causing various toxicological effects (Choi and Hu, 2008; Hussain et al., 2005). When exposed to oxidative conditions, NPs form reactive oxygen species (ROS) causing unwanted physiological outcomes; namely genotoxicity, apoptosis, necrosis, inflammation, fibrosis, metaplasia, hypertrophy, and carcinogenesis (AshaRani et al., 2012; de Lima et al., 2012). Further, NPs induce oxidative stress, inflammation and damage to proteins, cell membrane and deoxyribonucleic acid (DNA) (Fard et al., 2015).

The biological toxicity of NPs is caused by excessive production of ROS, namely hydrogen peroxide (H_2O_2), superoxide anion radicals (O_2^-), and hydroxyl radicals (•OH) (Boonstra and Post, 2004). Normally, the ROS are produced under physiological conditions as a response to oxygen uptake. However, exaggerated production in the presence of NPs is detrimental to the cells (Akter et al., 2018). The general mechanism of NPs toxicity involves their initial attachment on the surface of a cell. Subsequently, NPs enter the cell and initiate the production of ROS, thus causing traumatic stress (Ahmed et al., 2018). Similarly, the ROS attack biomolecules needed for the survival of the cell (Appierot et al., 2009; Ma et al., 2013). Fig. 3 presents various effects of NPs on biological cells. These effects involves 1) destruction of lipid peroxidation and cell membrane, 2) lysosome leak after the membrane destruction, 3) destruction of the mitochondrial membrane with a subsequent reduction in production of adenosine triphosphate or (ATP), 4) production of ROS interfering with ribonucleic acid (RNA) translation, 5) attachment inhibition of transfer RNA (tRNA) to the ribosome, 6) polymerization of proteins and deoxyribonucleic acid (DNA) induced by high ROS, 7) DNA mutation and finally 8) damage of the nuclear membrane causing cell death.

Various nanomaterials (NMs) including carbon nanotubes/nanodots,

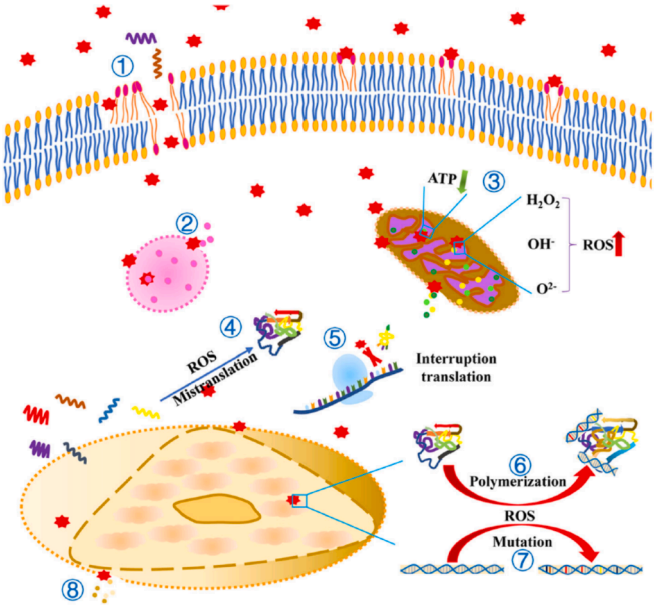

Fig. 3. Effects of NPs on cell destruction (Yu et al., 2020).

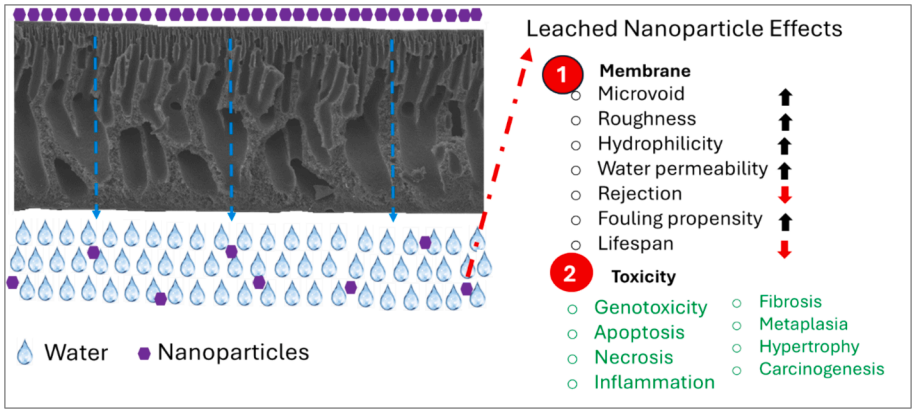

Fig. 2. Effects of NP leach on membrane filtration performance and toxicological impacts.

graphene, other small-sized fullerenes and metallic NPs have been integrated into polymeric membranes to improve their performance. However, these NMs present toxicological effects to the environment (Table 3). Chen et al. (2007) evaluated the impact of carbon nanotubes (CNTs) on the embryos of zebrafish (*danio rerio*) (Heng et al., 2007). 120 mg/L single walled CNTs and 240 mg/L multiwalled CNTs exposure caused a delayed hatching. However, hatching efficiency was 99 % upon embryos exposure to all CNTs (Heng et al., 2007). Although the test was based on carbon nanomaterials, the hatching delays were induced by Co and Ni catalysts remaining as impurities during production of CNTs. Metallic NPs cause functional or morphological changes of the biological species. Their cytotoxic effect is influenced by various factors including their solubility, charge at membrane surface and binding capacities. According to the reported acute toxicity of graphene oxide on *Daphnia magna*, the NPs altered normal physiology (heart rate) and feeding activity of the planktonic crustacean (Fekete-Kertész et al., 2020). This was associated with increased risk of predation and decline in reproduction. For this reason, the environmental health effects of NPs require constant monitoring. In another study, the toxicity of TiO_2 was related to the particle size of the NPs (16, 36, 89 and 3264 nm) (Fekete-Kertész et al., 2017). Reportedly, the nanoscale TiO_2 presented a negative impact on the aquatic ecosystem, and this was strongly influenced by their large surface area and wide distribution in water. Some contributing factors relating to NP toxicity include their concentration, duration and mechanism of exposure (Fekete-Kertész et al., 2016). Various studies evaluated ecotoxicity of NPs on terrestrial species including wildlife, plants, and invertebrates (Table 4). Ling and Xing (2007) evaluated the effects of various NPs on seed germination and growth of higher plants species (radish, rape, ryegrass, lettuce, corn, and cucumber). Mild seed germination resistance was reported with severe impact on roots growth.

Due to their high reactivity, silver (Ag) NPs cause brown skin colourization. The exposure to high Ag concentrations cause argyria, typically a skin colour change to blue or blue-grey. When released to the environment, AgNPs, undergo transformation processes resulting in their physicochemical modification, environmental fate and toxicity. Levard et al. (2012) evaluated environmental transformation of AgNPs, considering their stability and fate. Upon sulfidation, the study reported a slow environmental release due to low water solubility of Ag_2SNPs (Levard et al., 2012). However, sulfidation of AgNPs creates a long-term environmental impact. When released to the soil, long-term effects involves slow autotrophic oxidation of ammonia (nitrification) due to destruction of bacterial phyla (Grün and Emmerling, 2018). Upon discharge to the wastewater treatment plants, AgNPs reduce the proliferation of microorganisms required for degradation of organic matter. Similarly, aquatic biota is affected by AgNPs release into the receiving bodies. Among others, the life marine species is threatened by the high levels of AgNPs. Katsumiti et al. (2015) investigated the mechanistic toxicity of AgNPs on mussels haemocytes and gill cells (Katsumiti et al., 2015). Small sized AgNPs ($\leq$20 nm) presented adverse effects on cell viability. The reported mechanistic action of AgNP involved genotoxicity, oxidative stress, destruction of actin cytoskeleton and inactivation of haemocytes phagocytosis stimulus (Katsumiti et al., 2015). The ecotoxicity of various nanoparticles is summarised in Table 4.

7. Improvement of nanoparticle stability on the membranes

Due to the leaching of loosely bound NPs, contemporary research is geared towards strategic enhancement of their stability in polymeric membranes (Bi et al., 2018; Mofokeng et al., 2020; Nthunya et al., 2016) Phan et al. (2020) reported uncontrolled leaching of ZnO NPs from polyacrylonitrile nanofibers during dye adsorption studies (Phan et al., 2020). However, the addition of hinokitiol (serving as a gluing agent) improved their stability with improved resistance to leaching. Also, addition of hinokitiol improved antibacterial activities and dye removal. In another study, Ng et al. (2020) improved stability of GO on TFN PA membranes supported on PSf through addition of binding PVA (Ng et al., 2020). The enhanced stability was attributed to the excellent interaction of GO with PVA due to intermolecular hydrogen bonding. The bonding was enabled by the presence of hydroxyl (–OH) functional groups from PVA and carboxylic (–COOH), hydroxyl (–OH) and oxygen (–O–) functional groups from GO (Kashyap et al., 2016). Other studies explored addition of glutaric dialdehyde (GA) as the cross-linking agent between the membrane polymers and the NPs (Tao et al., 2017). Also, copolymers are used as adhesive agents to effectively retain NPs in the

Table 4
Ecotoxicity of nanoparticles on various ecological species.

Nanoparticle and size	Test dose	Test species	Cytotoxic results	Ref.
AgNPs (50 nm)	50 μg/L	Juvenile rainbow trout (*Oncorhynchus mykiss*)	The AgNPs presented high genotoxicity through gene oxidative stress, protein denaturation, impaired DNA synthesis and decreased differentiation into the acetylcholine phenotype	(Auclair et al., 2019)
AgNPs (20 nm)	0.03–0.09 mg/L	Carp fish	Bioaccumulation of Ag causing histopathological alterations including gill tissue damage and necrosis	(Kakakhel et al., 2021)
AgNPs (50 nm)	25–1000 mg/kg	*Labeo rihota* fish	Damage of tissue, primary lamella, and blood vessels. Also, formation of vacuolation in liver and muscles was reported	(Thirumurugan et al., 2016)
CNTs	120–240 mg/L	Zebrafish (*Danio rerio*)	The presence of NP catalysts impurities from the CNTs caused delayed hatching of zebrafish	(Heng et al., 2007)
Zn (35 nm)ZnO (20 nm)	20–2000 mg/L	Ryegrass and corn	Both NPs inhibited seed germination. The roots of germinated seeds were resistant to growth	(Lin and Xing, 2007)
CeO_2NPs	0.05–0.5 g/kg	Soybean (*Glycine* max)	The NPs caused the plant leave stress and damage.	(Priester et al., 2017)
AgNPs (20–73 nm)	100 mg/L	Broad bean (*Vicia faba*)	Leaf accumulation of AgNPs affected plant photosynthesis	(Falco et al., 2020)
CuONPs (30 nm)	0–1000 mg/L	Bt-transgenic cotton	The NPs aggregated on the epidermis and intracellular spaces of cotton plant, causing nutrient uptake and growth decay	(Le Van et al., 2016)
AuNPs (14 nm)	16–33 mg/kg	Bacteria (*S. rimosus*)	The AuNPs presented no toxicity on *S. rimosus* and therefore classified as harmless on soil microbes	(Maliszewska, 2016)
AgNPs (30–75 nm)	1 mg/g	Bacterial rich sediments	The NPs affected the sediment microbial biomass and enzyme activity.	(Bao et al., 2016)
PdNPs	9.5 mg/L	Marine bacteria (*V. fisheri*)	The NPs were deemed non-toxic to marine microbes.	(Nuzzo et al., 2017)
AuNPs (5–40 nm		Marine bivalve *Scrobicularia plana*	The NPs accumulated in the tissues of the invertebrate impairing its burrowing behaviour.	(Pan et al., 2012)
CuONPs (25–55 nm)	0–100 mg/L	Rotifer (*B. plicatilis*), shrimp (*A. fransiscana*)Copepod (*T. fulvus*)	The NPs presented various lethal doses ranging from 12.4, 16.9, 64.6 mg/L against *T. fulvus, B. plicatilis, A. fransiscana* respectively.	(Rotini et al., 2018)

membrane polymer. Yu et al. (2021) improved AgNPs leach resistance from PA-MF membranes through mussel-inspired immobilization of zwitterionic poly(carboxybetaine acrylate-co-dopamine methacryamide) (PCBDA) (Yu et al., 2021). Bonds formation between NPs and substrate (polymer) increases stability and resistance to leaching. According to Farid et al. (2019), formation of hydrogen-bonding and strong van der Waals forces influenced by carbon nanotubes (CNT) coating on polyvinylpyrrolidone (PVP) increased NPs stability with minimal leaching during hydraulic flushing in cross-flow filtration at 1 bar for 6 h (Farid et al., 2019). The NPs were anchored onto the PVDF membrane via vacuum filtration. Similarly, Ding et al. (2019) chemically bound SiO_2 and copper ferrocyanide (CuFC) layers onto PVDF membranes for the removal of cesium (Ding et al., 2019). The CuFC layer was rigidly attached on the surface of SiO_2NPs and the membrane, thus minimising leaching propensity. In another study, Abdulkarem et al. (2021) prepared PVDF-α-zirconium phosphate NPs mixed matrix membranes to remove toxic metals from water bodies (Abdulkarem et al., 2021). The NPs were immobilized within the membrane matrix through the formation of hydrogen bonds between C–H groups of PVDF chains and O–H moieties of α-zirconium phosphate. Their findings were consistent with related studies (Ouda et al., 2020; Ibrahim et al., 2020).

In other instances, the stability of NPs is improved through a smart choice of material compatibility and synthesis alteration. The in-situ method to generating yttrium-PES composite adsorptive membranes demonstrated resistance of NP leach during fluoride and arsenic adsorption studies (He et al., 2019). This was associated with stable embedment of NPs within the membrane matrix due to the good phase compatibility. Also, the coating process improves NP resistance from leaching. Basri et al. (2010) reported improved stability of AgNPs upon the addition of PVP to the casting solutions, preventing their leach from the polymeric matrix (Basri et al., 2010). The PVP-coating of NPs improved their dispersion with minimal agglomeration on the polymer matrix (Figueiredo et al., 2015). Further, PVP increased the solvent-non-solvent demixing rate during the phase inversion process, thus increasing the membrane porosity and interactions between the polymer and additive (Purnawan et al., 2021). In another study, Beisl et al. (2019) synthesized CA-AgNPs and CA-Ag ion exchange zeolites nanofiltration membranes (Beisl et al., 2019). The PVP-coating of the NPs improved resistance to leaching. The negligible loss of NPs was dependant on the solution chemistry including pH, redox potential and other ions or molecules.

8. Future perspectives

In certain circumstances, the synthesis of NPs adopts the use of harmful chemicals such as strong bases and acids (Mahlangu et al., 2017). Furthermore, these procedures are lengthy and costly. However, recent studies are focusing on greener and fast methods towards synthesis of NPs such as plant extracts-microwave-assisted methods (Li et al., 2013; Abu et al., 2020), are aimed at minimization of environment negative impacts. Nonetheless, the mass production of NPs following these green techniques has not yet been demonstrated (Kajau et al., 2021). There is a need for further investigation for potential commercialization of mixed matrix membranes while cognizant of the intensifying concern of secondary pollution from leaching of the incorporated NPs. Therefore, the use of environmentally friendly methods would be in vain if leaching toxic NPs occurs.

With the latest literature presenting the impact of nano- and microplastics from membrane systems in drinking water treatment plants, incorporation of NPs requires extensive assessment (Ding et al., 2020). Another factor needing attention is the disposal of worn-out membranes as these present serious challenges due to environment pollution. Also, commercial membranes have been successfully applied in different water matrices including harsh conditions. Also, these membranes have been used over extended periods (years) with high clear protocols of operational and cleaning procedures. Contrary, nanoengineered membranes have not been evaluated over long-term. For this reason, the effect of different cleaning strategies on the stability of NPs has not been demonstrated. This is a key requirement for commercialization, mostly because some cleaning solutions are capable of degrading polymeric membranes (Kajau et al., 2021). Therefore, the current study paves a way to two aspects of nano-engineered membranes: i) Membrane fabrication/preparation (knowledge of the essential nanoparticle-polymer interactions and compatibility) and ii) fundamental knowledge long-term application of the future ideal (next generation) membranes potentially breaking the performance boundaries of current commercial membranes (nano particle stability).

9. Conclusions

The current study demonstrated improvement in membrane performance (removal of solutes) and reduction in fouling upon NPs addition. However, the challenges associated with NP-modification of the membrane require research attention. These include potential leaching of toxic NMs causing secondary pollution. Also, the use of harmful chemicals during NPs synthesis remains the environmental critical threat. Furthermore, disposal of nanoengineered membranes should be adequately established and clearly indicated before commercialization. Besides adoptions of other membrane modification technologies, incorporation of dendritic, hydrophilic and zwitterionic polymers, NPs could be firmly anchored onto the polymer matrix through the formation of hydrogen bonds and/or selection of compatible (miscible) blends. Despite proven opportunities to use NPs-enhanced membranes in water treatment, their commercialization is far from realization. This is augmented by research requirements of nano-enhanced membranes towards commercialization. Critical aspects needing improvement are presented in the current study, with a special focus on leach minimization and environmental toxicity. Irrespective of the challenges, mixed matrix membranes present opportunities for application in water treatment targeting the removal of a wide range of emerging pollutants. Nonetheless, research development of nano-enhanced membranes should be coupled with the analysis of the NP toxicity.

CRediT authorship contribution statement

Oranso T. Mahlangu: Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Software, Resources, Project administration, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Mxolisi M. Motsa:** Writing – review & editing, Writing – original draft, Data curation. **Heidi Richards:** Supervision, Resources, Project administration, Funding acquisition. **Bhekie B. Mamba:** Resources, Project administration, Funding acquisition. **Mosotho J. George:** Writing – review & editing, Validation, Formal analysis, Data curation. **Lebea N. Nthunya:** Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Software, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization.

Declaration of competing interest

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

Data availability

Data will be made available on request.

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