

Nanofiltration for drinking water treatment: a review

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Abstract In recent decades, nanofiltration (NF) is considered as a promising separation technique to produce drinking water from different types of water source. In this paper, we comprehensively reviewed the progress of NF-based drinking water treatment, through summarizing the development of materials/fabrication and applications of NF membranes in various scenarios including surface water treatment, groundwater treatment, water reuse, brackish water treatment, and point of use applications. We not only summarized the removal of target major pollutants (e.g., hardness, pathogen, and natural organic matter), but also paid attention to the removal of micropollutants of major concern (e.g., disinfection byproducts, per- and polyfluoroalkyl substances, and arsenic). We highlighted that, for different applications, fit-for-purpose design is needed to improve the separation capability for target compounds of NF membranes in addition to their removal of salts. Outlook and perspectives on membrane fouling control, chlorine resistance, integrity, and selectivity are also discussed to provide potential insights for future development of high-efficiency NF membranes for stable and reliable drinking water treatment.

Keywords nanofiltration, drinking water, disinfection byproducts, micropollutants, selectivity

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1 Introduction

Water scarcity and pollution at a global scale are grand challenges in the modern society [1,2]. According to the United Nations, 30% global population does not have the access to safe and reliable drinking water services, which drives United Nations to include the universal drinking water supply in their sustainable development goals [3]. Effective water treatment and management strategies are urgently called to achieve the goal through eliminating water pollution and producing clean drinking water. Membrane-based separation techniques such as reverse osmosis (RO) and nanofiltration (NF) are considered as promising candidates for reliable production of drinking water [4]. RO is mainly used for seawater desalination, which has rigorous criteria on the rejection of NaCl (e.g., normally $\geq 99\%$) [5]. Such high NaCl rejection generally requires a dense rejection layer who often comes at the expense of high energy consumption and low water permeability. In comparison, NF membranes have relatively loose structure to allow faster water production with lower energy requirement [6]. Meanwhile, NF membranes can be designed and optimized in a more flexible way to maximize their efficiency on drinking water production according to the different application scenarios (Fig. 1).

Since its first introduction at the late 1980s, NF membranes have been developed and optimized with improved separation performance through tuning their material compositions and structural properties [7,8]. Existing NF membranes often possess the state-of-the-art thin film composite (TFC) structure, consisting of a thin rejection layer (e.g., polyamide), a porous substrate layer,

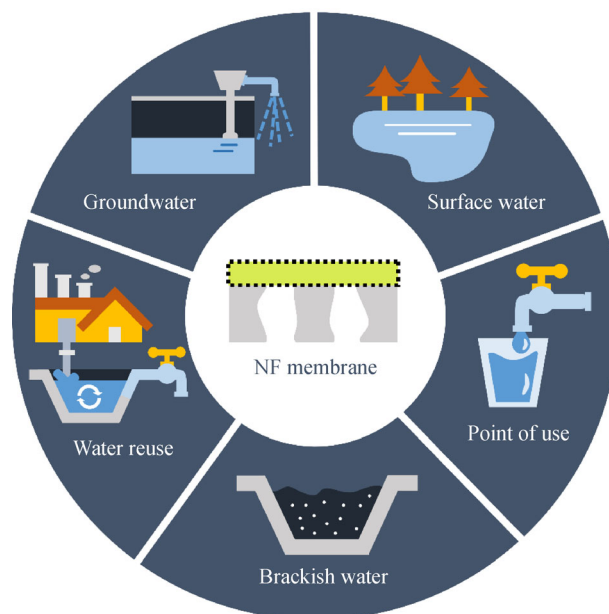


Fig. 1 Various application scenarios of NF technique, including surface water treatment, groundwater treatment, water reuse, brackish water treatment, and point of use applications.

and a non-woven fabric support [9]. The TFC structure allows independent optimization of rejection and support layer, which offers great flexibility for the design and fabrication of high-performance NF membranes [10]. In principal, the top thin rejection layer plays a critical role on NF membrane separation performance. Various approaches such as interfacial polymerization (IP) [11], surface coating/grafting [12], layer-by-layer deposition [13], and so-gel process [14], can be used to fabricate the thin layer.

Generally, NF membranes possess an effective pore size around 0.5–2 nm with a corresponding molecular weight cut-off (MWCO) ranging from 100 to 1000 Da [6,15]. These features enable NF membranes to achieve effective removal of suspended solids, colloidal, bacteria, and organics, as well as partial removal of dissolved ions, while most of the components are listed as the target pollutants for drinking water treatment [16]. Compared with traditional drinking water treatment methods, NF enjoys several advantages including: 1) small footprint and ease of automation, 2) wide-spectrum removal of various water contaminants to ensure high product water quality, and 3) flexibility to adapt different feed water quality. On the other hand, the operational cost, membrane fouling, and long-term stability shall also be considered to comprehensively evaluate the capability of NF for drinking water production. In view of the great potential of NF-based drinking water treatment to address the critical challenge of water scarcity and pollution, it is important to timely and thoroughly review the progress of NF technique for water purification in terms of material development, membrane fabrication, and practical applications.

Although several reviews on NF membranes have been published [17–21], they either lack of strong emphasis on the specific application scenario of drinking water treatment, or have limited timelines to include latest research findings.

In this review, we comprehensively summarize the recent research progress on NF-based drinking water treatment. NF membrane materials and their fabrication approaches are first introduced to provide basic information on the technique development (Section 2). Subsequently, the practical applications of NF membranes for drinking water treatment under different scenarios are presented, namely, surface water treatment (Section 3), groundwater treatment (Section 4), water reuse (Section 5), brackish water treatment (Section 6), and point of use applications (Section 7). Based on the membrane performance for various applications, perspectives on the challenges and future development of NF membranes for drinking water production are also discussed.

2 NF membrane materials and fabrication

2.1 Polymeric NF membranes

The majority of existing NF membranes has a TFC structure consisting of a thin polyamide layer, a porous substrate layer (e.g., polysulfone or polyethersulfone), and a non-woven support [9]. Figure 2 shows the fabrication strategy of polyamide-based NF membranes, the morphology and structure of a NF270 membrane, as well as their separation performance and application potential. The

polyamide is generally fabricated by performing the IP reaction between an aqueous amine solution containing piperazine or *m*-phenylenediamine and an organic solution containing trimesoyl chloride (Fig. 2(a)). The formed polyamide layer (Figs. 2(b) and 2(c)) plays a critical role on membrane separation performance. Nevertheless, the separation performance of polyamide membranes are often limited by the trade-off between membrane permeability and selectivity (Fig. 2(d)) [22]. To break the limitation, various strategies were explored to improve membrane separation efficiency (Fig. 2(e)). For example, nanomaterials can be introduced into the polyamide layer via adding them in the aqueous/organic phase during IP reaction, forming thin-film nanocomposite (TFN) membranes [23]. These incorporated nanomaterials in TFN membranes not

only create vast nanochannels for enhanced water transport [24] but also introduce beneficial functions such as antimicrobial properties [25]. Alternatively, constructing an interlayer onto the support membrane before the IP process has been confirmed as an effective approach to regulate the properties of polyamide layer with enhanced membrane separation performance [26,27]. The incorporation of the interlayer could not only facilitate the formation of better-quality polyamide rejection layer (e.g., through fine-tuning reaction rate and the extent of the IP reaction [28,29]) but also optimize the water transport pathways in the dense polyamide rejection layer by acting as a high-permeability gutter layer [26]. These synergistical effects could simultaneously improve membrane water permeance (up to an order of magnitude) and rejection of solutes [29].

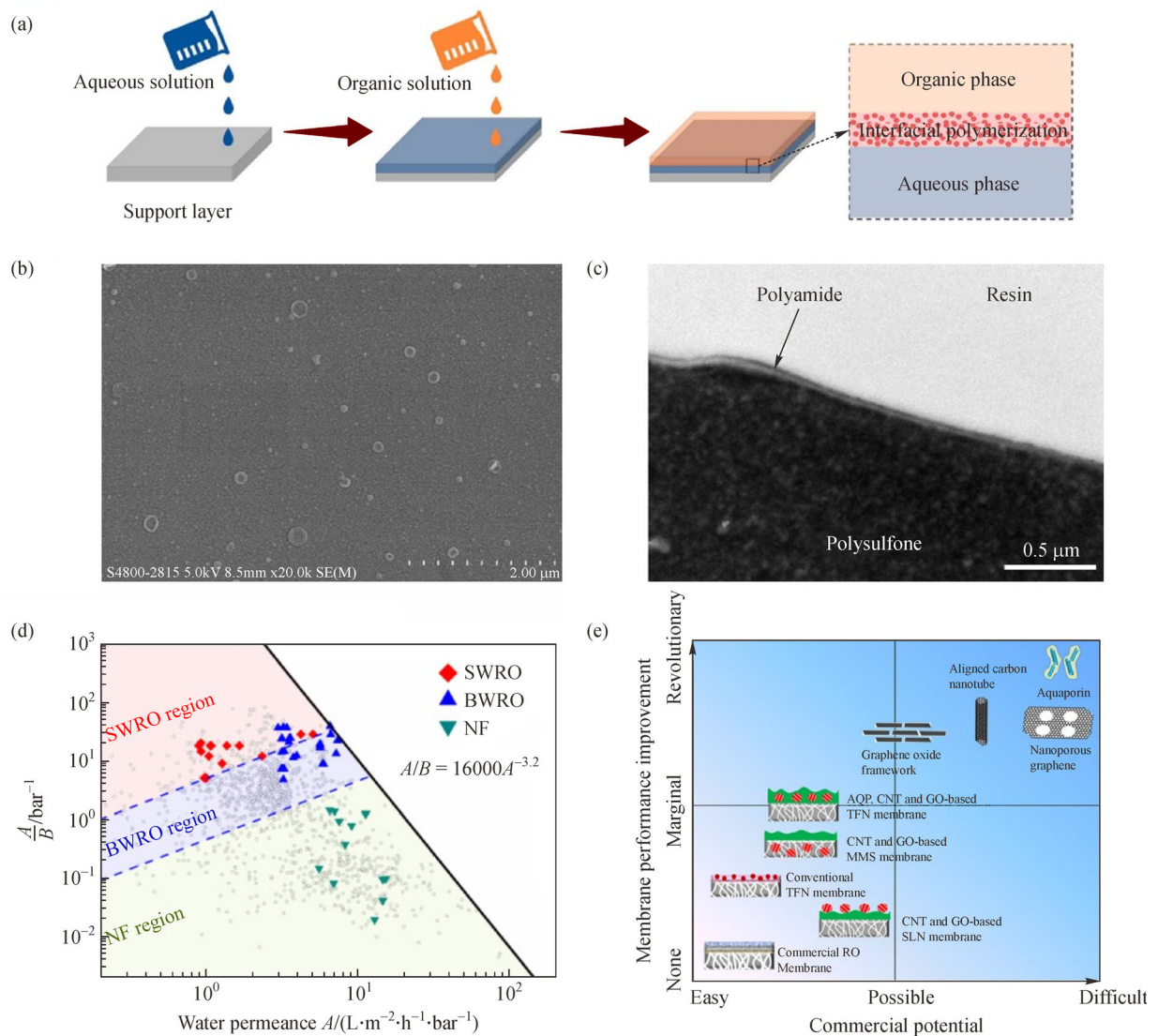


Fig. 2 (a) Schematic of interfacial polymerization process, (b) scanning electron microscope image of the surface for NF270 membrane, (c) transmission electron microscope image of the cross-section for NF270 membrane, (d) the upper bound of TFC polyamide membranes for desalination, and (e) comparison of membrane performance improvement and commercial potential of the TFC polyamide membranes. These figures are reprinted from ref. [22,30–33] with copyright permission.

As a result, this TFN structure with an interlayer (TFNi) is promising to break the longstanding permselectivity trade-off effects (also known as the upper bond [22]) owing to their largely improved permeability without compromising solutes retention.

In addition to polyamide, alternative polymers, such as polyurethanes, poly(bio-amides), polyanilines, polyesters, and polyimides, can also be used as membrane rejection layer materials through IP reaction (Table 1) [34]. Cellulose-based NF membranes are also commercially available, who have been used for water purification since their successful fabrication in the 1960s [35]. For example, cellulose acetate NF membrane was used to remove organic micropollutants from drinking water [36], and encountered low membrane fouling because of the hydrophilicity of the cellulose material. However, cellulose-based membranes (e.g., cellulose acetate) have relatively low tolerance to pH (2–8) and thermal (< 30 °C) change, which greatly restricts their applications [37]. In addition, sulfonated polyethersulfone is also used as the rejection layer of some commercially available NF membranes (e.g., NTR-7450, Nitto Denko, Japan) [38]. The existence of sulfonate group leads to a negatively charged membrane surface, which could benefit to the rejection of anions as a result of electrostatic repulsion [39].

Polyelectrolyte NF membranes fabricated by layer-by-layer assembly have also been investigated for water treatment [45]. The chemistry and structure of polyelectrolyte membranes can be tuned via controlling the polyelectrolyte types and number of deposited bilayers during layer-by-layer procedure. An extension of this technique is the electrospray-enabled layer-by-layer fabrication of NF membranes (also called three-dimensional printing) [46]. This technique allows the control of membrane structure via adjusting electrospray conditions, which could fabricate thinner and more uniform membranes with enhanced separation performance.

2.2 Non-polymeric NF membranes

Ceramic membranes with superior physical and chemical stabilities have also gained a growing interest. Similar to

polymeric membranes, ceramic NF membranes generally possess average pore size of 1–2 nm with effective MWCO ranging from 200 to 1000 Da [14]. As presented in Table 2, the nanoparticles of metal oxides such as SiO₂, TiO₂, and ZrO₂ are often used as the selective layer of ceramic NF membranes, which are usually fabricated through the sol-gel process [14]. In addition to single kind of metal oxide nanoparticles, the composite ceramic NF membranes using mixed metal oxide, such as ZrO₂/TiO₂, have also been commercially produced to improve membrane separation performance [47]. However, the water permeability of ceramic NF membrane is often lower than that of polymeric NF membranes with similar MWCO, which can be attributed to their lower porosity and higher thickness. To improve membrane porosity, additional sacrificial pore-foaming agents (e.g., cotton, starch, polymer beads, graphite, and Ni) could be introduced into the ceramic slurry [48]. In addition to sol-gel processes, other advanced fabrication methods, such as atomic layer deposition (ALD) and chemical vapor deposition (CVD) (Fig. 3), are also explored to prepare a more uniform and tight separation layer [49].

In addition to polymeric and ceramic membranes, emerging NF membranes fabricated by novel nanomaterials have attracted increasing attentions in recent years. For example, two-dimensional (2D) materials (such as MoS₂, graphene, graphene oxide, metal-organic frameworks, MXene, and covalent organic frameworks) exhibited high potential for NF membrane fabrication [51,52]. In general, 2D materials possess unique physicochemical properties (e.g., nanochannel size and thickness) which allow precise separation and thus enhance membrane selectivity. The 2D material-based NF membranes can be fabricated through physical stacking [53], chemical bonding [54], electro-assisted deposition [55], etc.

Currently, polyamide-based NF membranes are most commonly used for drinking water production owing to the matured fabrication technique, successful applications in separation industries, and reasonable cost-effectiveness [19]. Ceramic NF membranes also have great potential for drinking water treatment owing to their high chemical stability, which allows them to be used with oxidants (e.g., Cl) for disinfection and membrane cleaning [56]. However,

Table 1 Materials used for NF membrane preparation via IP

Material	Advantage	Drawback	Field of application	Ref.
Polyamide	Matured fabrication and high separation performance	Vulnerable to chlorination	Membranes mainly for NF and RO	[9]
Poly(bio-amide)	Biocompatibility and switchable properties	Relatively high MWCO values	NF applications with high permeability	[34]
Polyester	High resistance to chlorine	Susceptibility to hydrolytic degradation	NF applications with improved chlorine stability	[40,41]
Polyimide	High tolerance to organic solvent and thermal stability	Instability of amic acid groups in water	Organic solvent NF	[42]
Polyaniline	Electrochemical properties	Difficult to control nanostructures	Organic solvent NF and electrically conductive membranes	[43,44]

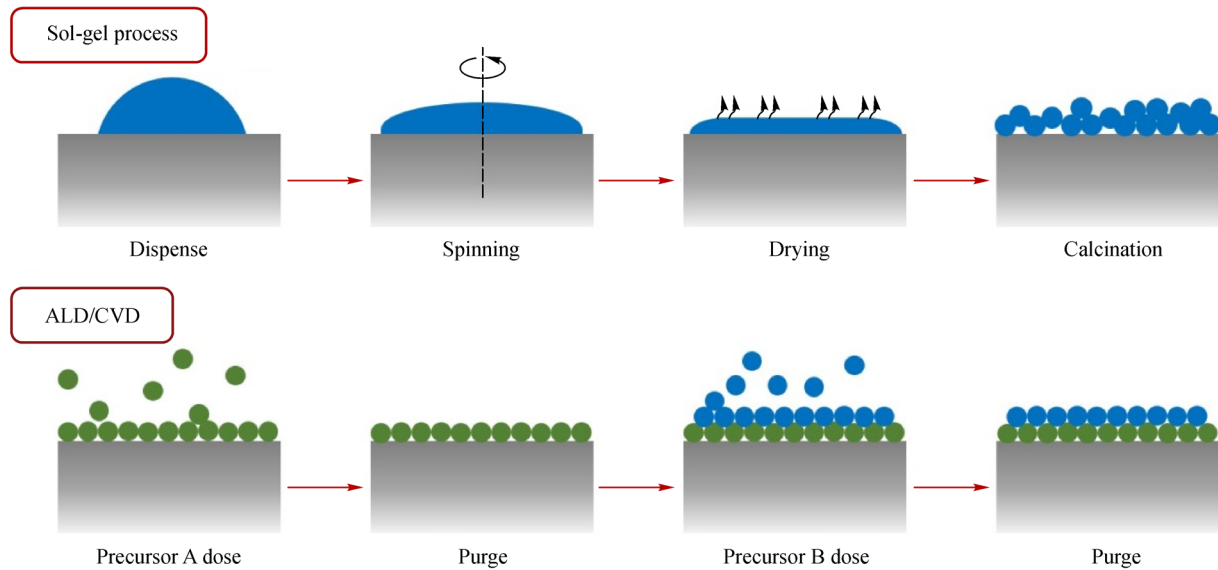


Fig. 3 Schematic illustration of sol-gel and ALD/CVD processes.

Table 2 Key features of various ceramic NF membranes and commercially available products ^{a)}

Material	MWCO/Da	Fabrication method	Product	Ref.
SiO ₂	600	Colloidal sol-gel method	Ceramic Inopor [®] membrane	[14]
TiO ₂	200–1000	Colloidal sol-gel method; electrophoretic deposition	Fine UF N001 of TAMI Industries and Ceramic Inopor [®] membrane	[14]
ZrO ₂	350–400	Colloidal sol-gel method	–	[14]
ZrO ₂ /TiO ₂	400–500	Colloidal sol-gel method	–	[14]

a) According to the literature, the price of commercial ceramic NF membrane is around 1000–2500 USD·m⁻² [50].

the application of ceramic membranes in drinking water treatment is largely limited due to the high cost (e.g., up to 10 times of the cost for polymeric membranes [50]). In recent decades, the development of novel materials (e.g., 2D materials) is also driving the fabrication of high-performance NF membranes using such materials [57,58]. Nevertheless, these emerging membranes using novel materials/fabrication methods are mainly investigated at laboratory-scale, their full-scale manufacturing is facing various challenges (e.g., low production rate, high cost, etc.). Future research needs to pay more attention to the scale-up of novel membranes and their performance for realistic applications.

3 NF for surface water treatment

Surface water, the water in a river, stream, creek, lake, and reservoir, is the major source of drinking water. In China, surface water accounted for 84.2% of the total water supply in 2019. In general, surface water possesses relatively high quality with a total dissolved solids less than 1000 mg·L⁻¹. However, the water quality is easily

affected by runoff, rainfall, point/non-point source pollution, etc. Consequently, the water quality may seasonally fluctuate [59] with potential contamination from various contaminants such as synthetic organic compounds [60]. Moreover, surface water is likely to be accidentally polluted by chemical discharge and algal blooms [61].

Conventional treatment process (coagulation-sedimentation-filtration-disinfection) is mostly used for the purification of surface water. This conventional process is designed for the removal of particles and pathogens, and may not effectively remove some emerging pollutants [62]. More importantly, in the unit process of disinfection, disinfection byproducts (DBPs) are formed because of the reaction between the disinfectants and precursors in surface water, such as natural organic matter (NOM) and bromide. Because NF membrane could reject most of organic matters, it becomes an alternative to address above challenges in addition to existing treatment system.

3.1 NF system for surface water treatment

In 1999, the world's first large-scale water treatment plant using NF, i.e., Méry-sur-Oise plant, was built in France.

The plant had a production capacity of $140000 \text{ m}^3 \cdot \text{d}^{-1}$ and could effectively remove the NOM and pesticides in the feed river water [63]. In China, although NF has been used for saline water treatment for many years (total dissolved solids is the major concern), it has not been specifically used for the control of DBPs and other pollutants in drinking water until recent years. For example, a $300000 \text{ m}^3 \cdot \text{d}^{-1}$ NF-based water plant is under construction in Zhangjiagang City of China, who will use NF to treat the Yangtze River water to improve the quality of drinking water.

A typical NF system for surface water purification consists of pretreatment process, membrane process, and posttreatment process (Fig. 4). Because of the relatively high NOM content in surface water, the priority of pretreatment is to reduce the risks of membrane fouling for NF unit. The commonly used pretreatment processes include coagulation, flocculation, sedimentation, sand filtration, microfiltration, and ultrafiltration [64–66]. These pretreatments could largely remove particles, organic matters, and bacteria, thereby reducing membrane fouling in NF units. After filtering through the NF membrane, most of the pollutants are removed. However, alkalinity is also partially removed during pretreatment, resulting in high corrosion potential. Therefore, stabilizing chemicals such as sodium carbonate and lime should be added to adjust the water alkalinity [64]. A more flexible approach for water stabilization is to blend the effluent of the pretreatment units (i.e., the feed of NF unit) with the NF permeate (Fig. 4), while this approach was practiced by Kaotan Water Treatment Plant ($300000 \text{ m}^3 \cdot \text{d}^{-1}$, Taiwan, China) [67]. Before entering to the distribution system, the treated water needs chlorination to prevent the regrowth of bacteria in the pipe network.

3.2 Control of DBPs formation

The NF membrane could effectively remove microorganisms, divalent ions, heavy metal ions, NOMs, and many trace organic matters [7]. Here, we mainly discuss the major issues of controlling DBPs in the surface water treatment system. The removal of organic contaminants, pathogens, and dissolved ions will be discussed in Section 5 NF for water reuse and Section 6 NF for brackish water treatment.

Disinfection is a vital step to eliminate the concerns of pathogens in drinking water. However, the process has

high tendency to generate toxic DBPs associated with healthy concerns [68]. Most DBPs are formed by the reaction between chlorine/chloramines (disinfectants) and the precursors such as NOM [69]. Typical DBPs include trihalomethanes, haloacetic acids, and *N*-nitrosodimethylamine. NF has a high rejection of NOM (50%–95%) [70], which could largely suppress the formation of DBPs in subsequent disinfection. For example, commercial NF membranes (ESNA, TS80, and NF270) could achieve a removal rate of 72%–97% and 57%–83% for *N*-nitrosodimethylamine and trihalomethanes, respectively [71]. For the world's first large-scale NF water treatment plants (i.e., Méry-sur-Oise plant), the trihalomethanes reduced by 50% in their distribution system after introducing NF process [72]. In addition to chlorine, the presence of bromine can also increase the generation of DBPs and shift the speciation to brominated species. However, NF shows low rejection for bromide (< 30%) [73]. The low removal of bromide coupled to the high removal of NOM could shift the trihalomethanes and haloacetic acids to more brominated species during the chlorination of NF permeate [74]. Because NF process has a high removal of pathogen (physical disinfection), the dosage of chlorine could be reduced in the following disinfection unit [75], which could also reduce the formation of DBPs and the odor of chlorine in tap water. In short, NF process can effectively remove the DPBs precursors and decrease the chlorine dosage, thereby effectively reducing the DBPs formation.

4 NF for groundwater treatment

Groundwater is an important natural resource serving for agricultural irrigation and drinking water supply in most countries over the globe [76]. Many groundwaters contain high concentration of hardness ions such as Ca and Mg due to ambient geochemical conditions, and these ions have to be removed prior to use [77]. Meanwhile, with the intensified chemical use and continuous discharge of industrial waste, groundwater quality has been greatly hampered and resulted in numerous contaminated sites [78]. Emerging contaminants such as per- and polyfluoroalkyl substances (PFASs) and As appeared in the groundwater and posed great health concern to humans [79,80]. NF technology, as a technology for multi-contaminants removal, are thus required to purify groundwater for drinking purpose.

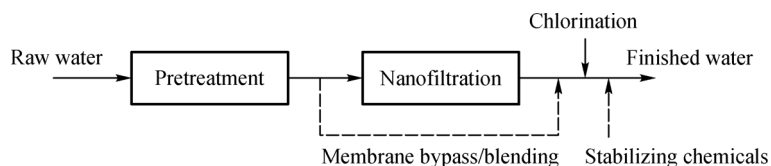


Fig. 4 A typical NF system for surface water purification.

4.1 Hardness removal

NF membranes have been used for hardness removal in groundwater treatment for decades [81]. The emerging use of NF membranes originates from two reasons: 1) traditional high-pressure RO membranes resulted in high energy cost; 2) the permeate quality was often too good and required post remineralization [82]. NF membranes possessed moderate rejections with higher fluxes compared to high pressure RO membranes, which make NF advantageous for hardness removal in groundwater treatment. NF membranes can typically achieve >98% removal of Ca and Mg, through size exclusion mechanism [83,84]. As Ca is essentially beneficial for human health, recent research started focusing on enlarging the pore size of NF through incorporation of a high molecular weight bipiperidine monomer during the IP to form a loose, nanoporous selective layer structure, which selectively rejected sulfate while partially passing through Ca ion [85]. The fabricated NF membrane not only maximized water permeability but also reduced membrane scaling due to less CaSO_4 accumulation on membrane surface. It provides important insights for future groundwater treatment adopting NF processes with strong emphasis on tailoring the selectivity and permeability of NF with enhanced water recovery.

4.2 As and PFASs removal

Figure 5 illustrates the rejection mechanisms of As and PFASs under different water chemistry by NF membranes. As has now become a typical toxic element in groundwater systems due to geochemical occurrence and industrial pollution, and posed elevated health risks such as skin and lung cancer [86,87]. Common forms of As in groundwater are trivalent arsenite, i.e., As(III), and pentavalent arsenate, i.e., As(V), with concentration ranges from <0.05 ppb up

to 5000 ppb [88]. The As(III) and As(V) rejections by NF varied depending on different charge states. The rejection of uncharged As(III) by NF membranes is mainly governed by size exclusion, while it often suffered lower rejections of <90% [88,89]. In comparison, NF membranes could achieve high rejection of 96%–99% for negatively charged As(V) thanks to the combined effects of electrostatic repulsion and size exclusion (Fig. 5(a)) [79,90]. Peroxidation of As(III) to As(V) is a viable method and can be integrated with NF process to boost the overall arsenic removal efficiency [91]. Therefore, it is important to have adequate knowledge of As speciation and ambient aqueous chemistry condition for the efficient removal of As during groundwater purification with NF technology. Moreover, tailoring the pore size of NF membranes with enhanced effect of size exclusion might be another way for removing uncharged As(III) form [92].

PFASs are a class of emerging contaminants that are persistent in groundwater and resistant to natural degradation process [95–97]. PFASs have been shown to be bio-accumulative and toxic at trace concentrations, thereby causing great health concerns [98,99]. NF membranes are likely to achieve relatively high rejection of >95% of PFASs, which allows them to purify groundwater with fluctuating PFASs concentrations and ensure stable permeate quality [100–102]. The removal of PFASs by NF membranes is mainly governed by size exclusion in addition to electrostatic repulsion between anionic PFASs and negatively charged membrane surface. For example, NF270 membrane has an average pore size of 0.88 nm [31], which is significantly smaller than the molecule size of 1.09 nm for PFOS, resulting in a high PFOS rejection of >95% by the membrane [94]. The rejection could be further enhanced with increasing the pH of feed solution as a result of stronger electrostatic repulsion at high pH [94]. Nevertheless, further research on the efficacy and mechanism of PFASs removal by NF under different fouling

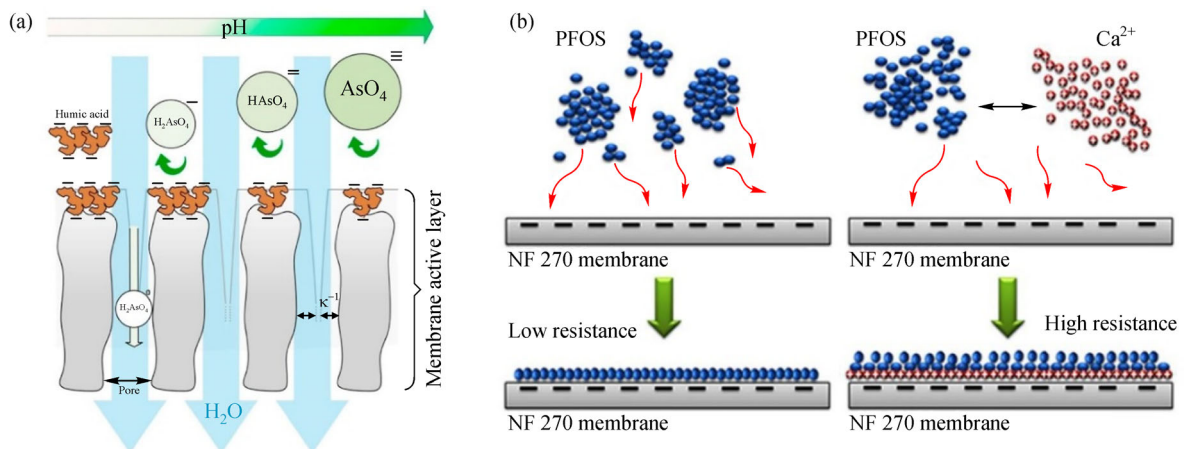


Fig. 5 (a) Schematic diagram presenting the removal of As(V) at different pH and fouling condition by a NF membrane. Reprinted with permission from ref. [93], copyright 2021, Elsevier BV. (b) Illustration of perfluorooctane sulfonate (PFOS) removal at the presence of calcium. Reprinted with permission from ref. [94], copyright 2013, Elsevier BV.

conditions and water matrices is still needed (Fig. 5(b)). In addition, it is also worthwhile to explore the control of emerging PFASs (e.g., hexafluoropropylene oxide dimer acid, also known as GenX) using NF membranes [103].

The use of NF in groundwater purification is an all-in-one approach to concurrently remove hardness, organic matters, and micropollutants. Previous studies generally focused on removing one or two targeted species, while groundwater is usually complex and consists of varied substances. Tailoring the separation properties of NF membranes for complex types of groundwater is important in future studies to balance the trade-off between permeability and selectivity [104]. Developing novel NF membranes with environmentally friendly materials and manufacturing processes is also needed for sustainable development.

5 NF for water reuse

In addition to conventional water source (e.g., surface water and groundwater), wastewater is also considered as a supplementary source to produce potable/non-potable water through membrane-based water reuse [105]. In fact, some countries/regions with limited water source (e.g., Singapore, California, and Australia) have integrate reused water into their drinking water supply system for decades [106,107]. For example, Singapore has recognized the reused water (known as NEWater) as one of their national taps, contributing up to 40% water supply of the country [108]. Advanced membrane separation process such as RO/NF is a vital unit in water reuse system as they can effectively remove the majority of toxic and harmful pollutants from the feed. Although RO is more competitive to produce highly purified water, NF is also considered as a qualified barrier for the removal of water pollutants in water reuse [109]. For instance, NF is often proposed to generate reused water through the treatment of textile wastewater containing large amount of dyes and salts [110,111].

5.1 Removal of organic contaminants

The effluent of wastewater treatment plant is mainly used as the feed water for membrane-base water reuse. Compared with surface water and groundwater, the effluent tends to have more complicated water matrices even after appropriate treatment, which poses great challenges to the separation performance of NF membranes [112]. One of the challenges is to achieve effective removal of organic contaminants, a major component presented in the effluents of wastewater treatment plant associated with healthy concerns and reused water safety [113–115]. NF membrane rejection of organic contaminants is governed by several mechanisms, including size

exclusion, electrostatic interactions, hydrophobic interactions, and polar effects [116,117]. The organic contaminants presented with hydrophilic nature and large molecular weight (e.g., higher than the MWCO of membranes) are more easily removed by NF membranes thanks to the strong size exclusion effect. For example, NF90 membrane has a MWCO of 200 Da, which allow it to achieve > 95% rejection of hydrophilic antibiotics with molecular weight ranging from 250 to 361 Da [118]. Meanwhile, NF membrane surface is often negatively charged at environmental pH (e.g., pH 6–9) which is beneficial for the removal of contaminants with negative charge (e.g., dyes and PFASs) as a result of electrostatic repulsion [110,119]. In contrast, positively charged organic contaminants may suffer low membrane rejection due to the Donnan effect [120].

On the other hand, organic contaminants with strong hydrophobicity and/or high polarity are often poorly rejected by NF membranes, e.g., < 50% for endocrine disrupting compounds (EDCs) by NF90 [121] and NF270 membranes [31]. The hydrophobic interactions between EDCs and membrane facilitated the partition of these compounds into membrane material (e.g., polyamide) following by the diffusion through the membrane [122,123], resulting in the high permeance of EDCs (thus low rejection). Suppressing the hydrophobic interactions between compounds and membrane is an effective way to reduce the partition behavior and improve membrane rejection of hydrophobic contaminants (Fig. 6). For example, a hydrophilic polydopamine coating on a commercial NF90 membrane could significantly enhance its rejection of hydrophobic EDCs [121]. Similar phenomena were also observed in the cases of using tannic acid-iron based NF membrane (Fig. 6(a)) [118] and hydrophilic ceramic NF membrane [124] to remove hydrophobic contaminants. In addition, accelerating water transport through the membrane (i.e., enhancing membrane water permeance) is also useful to enhance membrane rejection of organic contaminants because of the dilution effects in the permeate side (Fig. 6(b)). Various strategies such as addition of porous nanofillers [125], creation of selective nanochannels [24], and introduction of interlayer [126] can prompt membrane water permeance and thus increase the rejection of contaminants.

5.2 Pathogen removal

Pathogens (e.g., bacteria and virus) are of critical concerns in wastewater treatment chain and are listed as the removal target with highest priority in membrane-based water reuse, especially after the global outbreak of COVID-19 [127]. Generally, the average size of pathogens (e.g., 20–300 nm for virus [128]) are significantly larger than the effective pore size of NF membranes (≤ 2 nm), therefore they should be completely removed by the membrane. However, there are several studies that reported incomplete

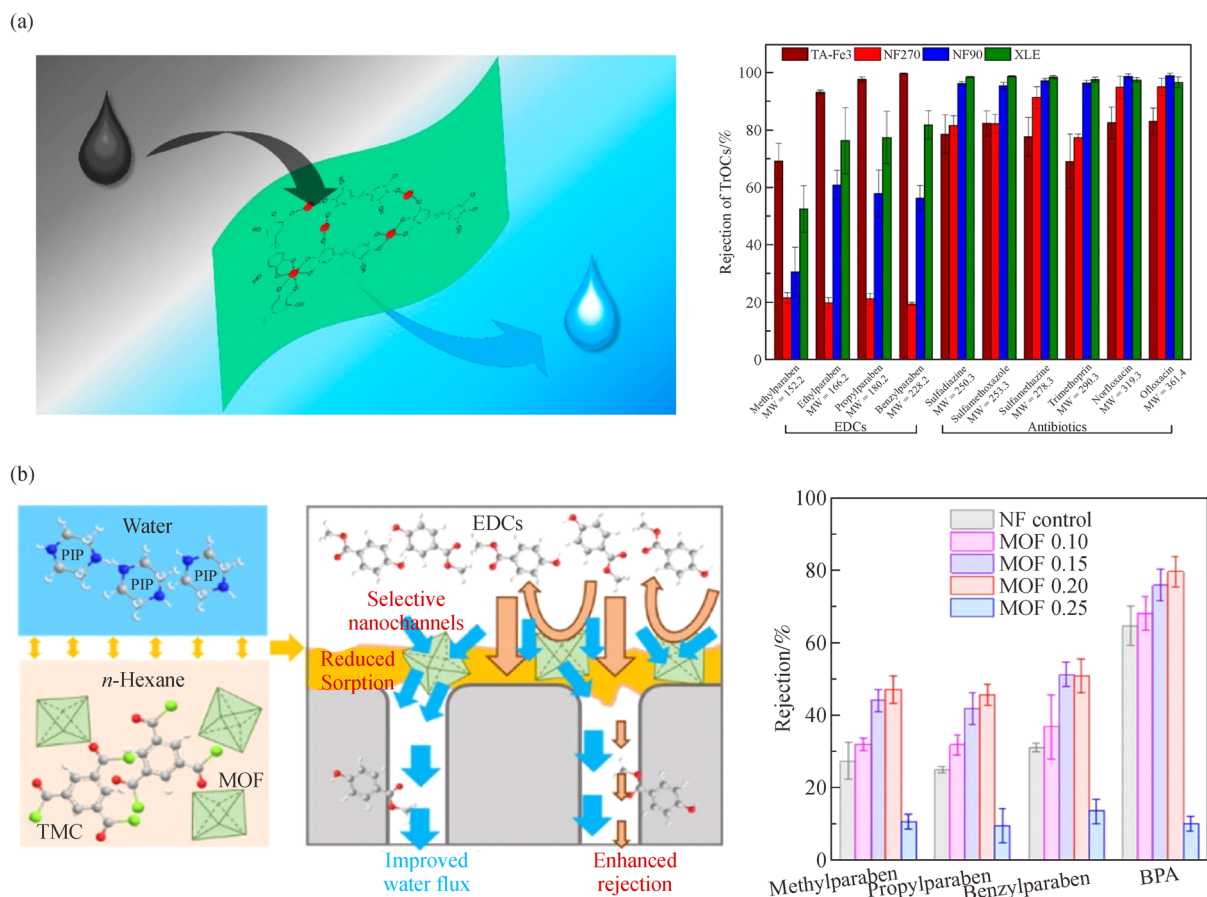


Fig. 6 (a) Removal of trace organic contaminants by a tannic acid-iron complexes-based NF membrane. Reprinted with permission from ref. [118], copyright 2019, American Chemical Society. (b) Incorporation of hydrophilic metal organic frameworks into a polyamide active layer for improved rejection of hydrophobic EDCs. Reprinted with permission from ref. [125], copyright 2019, American Chemical Society.

removal of virus and bacteria by RO membranes [129–131]. For example, Mi et al. reported a rejection of > 99.9995% (i.e., > 5-log removal) for bacteriophage MS2 (~25 nm) by spiral-wound RO elements [129]. A possible explanation for the incomplete rejection is that the virus may pass through the imperfections in the elements (e.g., insufficient sealing at the glue lines). In addition, a recent study reported that intrinsic nanosized defects are likely to be formed during the fabrication of polyamide NF membranes by IP [132]. The defects may serve as hot spots for virus transport, partially resulting in the incomplete rejection. To keep the safety of reused water away from pathogens, highly sensitive methods are strongly required to monitor the integrity of membrane and elements. Polishing strategies (e.g., *in situ* healing) may also be considered to improve membrane integrity toward satisfactory virus removal.

6 NF for brackish water treatment

Brackish water is one of the salinity water resources for

potable use with the salinity between fresh water and seawater. Commonly, the total dissolved solids of brackish water are in the range of 1–5 g·L⁻¹ with highly different compositions, such as Ca²⁺, Mg²⁺, SO₄²⁻, F⁻, NO₃⁻, and NOM. NF is an alternative separation technology with great application potential in brackish water desalination. According to the main separation mechanisms of NF, e.g., Donnan (charge) exclusion and size (steric) exclusion [133], NF can be used for the removal of hardness [81], F⁻ [134], and NO₃⁻ [135] from brackish water to improve the produced water quality or to reduce membrane scaling in the following desalination stage. Similarly, partial salts in brackish water could also be removed by NF as a result of the moderate rejection of NF membrane for monovalent salts and high rejection for divalent salts. Brackish water desalination performance of commercial TFC NF membranes were evaluated by Hilal et al. [136]. The results showed a reduced NaCl rejection from 95% to 41% with increasing the salinity of feed solution from to 25 g·L⁻¹. Gekas et al. used NF membrane to treat brackish groundwater with a high hardness [137], and the NF membrane gave a relatively high water flux of 15–47 L·m⁻²·h⁻¹

together with a hardness removal of 70%–76% and salt rejection of 44%–46%. Other candidates for brackish water treatment, such as RO and electro dialysis were also studied and compared with NF [138]. The results showed that NF permitted to reduce the concentrations of divalent ions, but the total dissolved solids of the produced water was still higher than World Health Organization standard. In addition to commercial membranes, novel NF membranes were also explored to improve the efficiency of brackish water treatment. For example, electrically assisted NF membranes were designed to improve membrane desalination performance (e.g., enhanced NaCl rejection from 54% to 82% via increasing applied voltage from 0 to 2.5 V) through finely tuned surface charge [139]. TFN membranes were also investigated to treat real brackish water [140].

NF process also has great flexibility to integrate with other separation technique to enhance the overall treatment efficiency (Fig. 7). For example, NF process was designed to couple with coagulation process to reduce membrane fouling propensity and improve desalination performance (Fig. 7(a)) [141–143]. Colloidal/organic fouling and scaling during NF would be successfully mitigated by the coagulation process. The integrated desalination system showed excellent sulfate removal and high rejection of divalent cations. Ion exchange could also be

integrated with NF process for brackish water desalination (Fig. 7(b)). The results indicate the significant reduction on energy consumption by the hybrid ion exchange-NF process compared with RO treatment [144]. Moreover, the hybrid system could potentially treat hard brackish water via separately removing Ca^{2+} and SO_4^{2-} through ion exchange and NF process, respectively. Meanwhile, the concentrate solution containing NaCl could be used to regenerate the ion exchange resins. Capacitive deionization (CDI) process has also been coupled with NF process to treat brackish water (Fig. 7(c)) [145]. The CDI-NF hybrid system could produce high-quality drinking water with a significantly reduced energy consumption compared to RO. Forward osmosis (FO) could also be combined NF to treat brackish water (Fig. 7(d)) [146]. Briefly, FO process used divalent salts solution (e.g., Na_2SO_4 or MgSO_4) as draw solution to treat brackish water, following by the NF re-concentration of diluted draw solution and production of clean water.

7 NF for point of use applications

Conventional drinking water supply system consists of collection system, centralized treatment plant, and distribution system. After the treatment in plant, water may

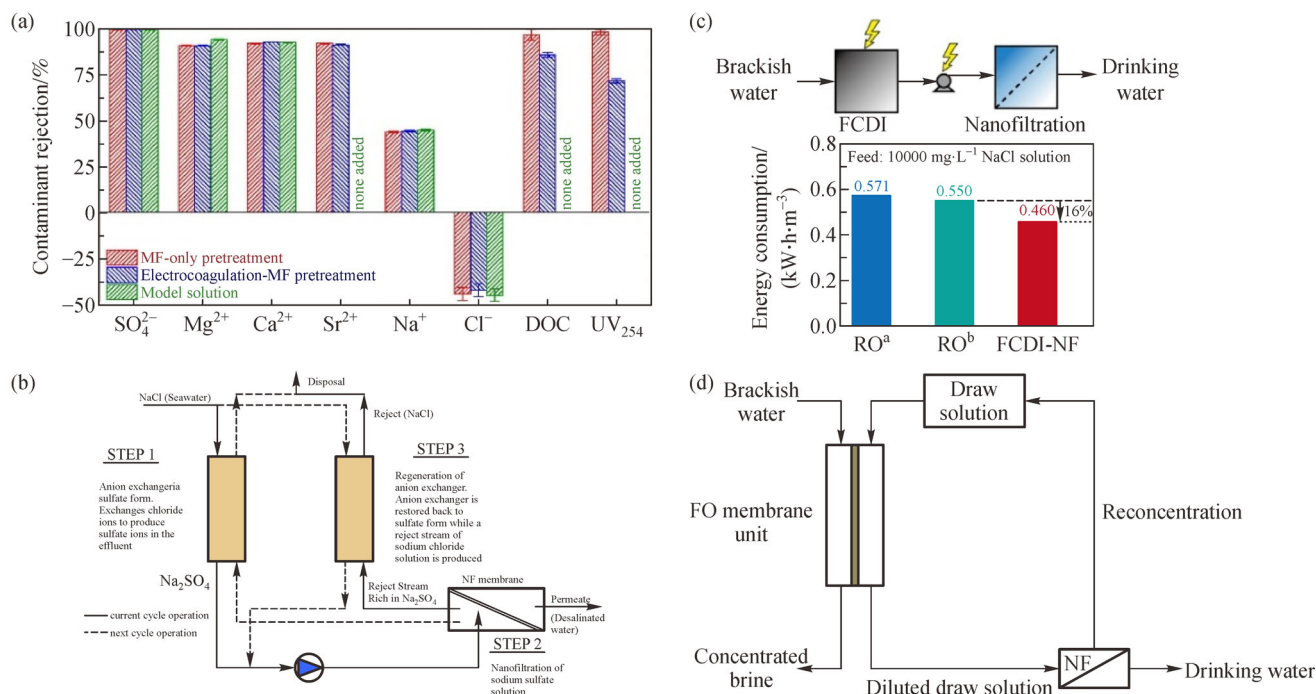


Fig. 7 (a) Removal of various ions and compounds from reservoir water using an integrated electrocoagulation-microfiltration-NF system. Reprinted with permission from ref. [143], copyright 2017, John Wiley & Sons. (b) Illustration of an ion exchange-NF system for desalination. Reprinted with permission from ref. [144], copyright 2008, Elsevier BV. (c) An energy saving CDI-NF hybrid system for brackish water treatment. Reprinted with permission from ref. [145], copyright 2017, Elsevier BV. (d) An FO-NF system for brackish water treatment. Reprinted with permission from ref. [146], copyright 2012, John Wiley & Sons.

also be polluted by pipe deterioration, pipe leakage, and microbial contamination in water distribution system [147]. Moreover, the occurrence of DBPs in the drinking water distribution system also poses great challenges to the safety of end users [148,149]. The mismatch between the increasing concerns on drinking water safety and increasing demand on high-quality drinking water is calling for effective strategies to produce reliable and safe water for end users.

One promising strategy is point of use drinking water treatment, which employs water purification systems at the end of the drinking water system (e.g., water tap, Fig. 8) [150]. As a decentralized water purification device, the point-of-use purifier should meet the criteria of low maintenance, ease of automation, and chemical free. NF cannot only well meet these requirements, but also shows good efficiencies for pollutant removal [151,152]. For example, a recent study reported the application of NF for treating disinfected drinking water [153], which should be directly send to the end users via distribution system. The results showed that the water quality was significantly improved via reducing dissolved organic carbon, total dissolved solids, and DBPs in the permeate water. Furthermore, NF-based point of use water purification system has great flexibility to integrate with other treatment methods to ensure the quality of product water. For instance, a NF system combined with granular-filtration/granular activated carbon sorption could substantially improve the quality of tap waters [154]. Point of use NF filter also benefits to the drinking water supply in vehicles. For example, NF-based locomotive drinking water purification could be realized at low pressure of 0.3–0.5 MPa with varying feed water quality and environmental conditions. The removal efficiency of excessive MgSO_4 by the point of use system could reach 97%–98% [155]. In addition, NF-based point of use drinking water treatment systems are beneficial for rural areas and

developing countries/regions thanks to its small footprint and easy maintenance [150]. The technology showed promising results for removing hardness, fluoride, dissolved organic carbon, and viruses, allowing people to obtain reliable drinking water.

8 Outlook and perspectives

This review comprehensively summarized the progress of NF-based technology for drinking water production under different application scenarios. With the development of novel membrane materials (e.g., carbon-based materials [156], 2D materials [58,157], metal organic frameworks [57], and covalent organic frameworks [158]) and improvement on membrane fabrication technique, NF membranes can achieve enhanced drinking water production and quality together with reduced energy consumption. Although NF enjoys significant advantages on membrane-based drinking water treatment, there are still several challenges including fouling, chemical stability, integrity, and selectivity, which need to be more effectively addressed in the future.

8.1 Fouling control

Fouling is a major obstacle for membrane-based drinking water treatment [159–161]. Specifically, severe fouling could not only increase membrane hydraulic resistance (i.e., increase operational pressure) but also decreased solutes rejection [162–164]. Surface coating, such as polydopamine [165,166] and polyvinyl alcohol [167] can effectively enhance membrane surface hydrophilicity, hence increasing membrane antifouling properties toward hydrophobic foulants, such as bovine serum albumin. Nevertheless, coating an additional layer onto a membrane could adversely result in decrease membrane permeance

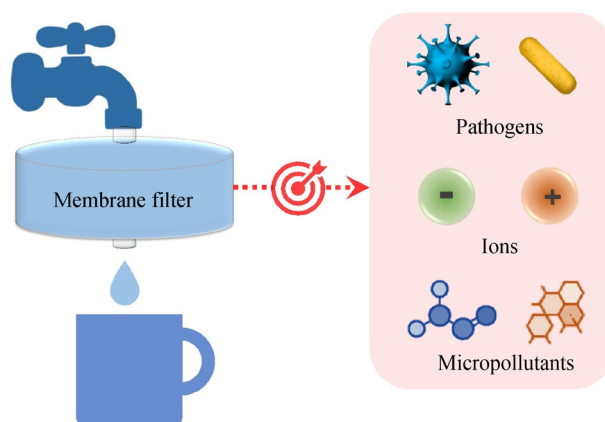


Fig. 8 Schematic illustration of a membrane-based tap water filter for point of use drinking water production. The filter shall able to remove various contaminants such as pathogens, dissolved ions, and micropollutants from drinking water source and thus safeguard product water quality.

due to the increased hydraulic resistance according to the resistance-in-series model [168]. Another effective strategy for membrane fouling control is to tune membrane surface roughness. Traditional wisdom believes a membrane with a smoother surface is less prone to fouling [169,170]. In contrast, a recent highlight some compelling evidence that a membrane with a rougher surface may less be fouled [171]. Such controversy may be related to the characteristic length of roughness and calls for more future studies to resolve the relationship membrane fouling behavior and its surface roughness. In addition to organic and inorganic fouling, microbial fouling (i.e., biofouling) issues can be potentially mitigated by incorporating antimicrobial agents in membranes (e.g., silver nanoparticles [172] and graphene oxide [173]) and engineering the biofilm formed on the membrane surface [174].

8.2 Chlorine stability

Another important dilemma of the TFC NF membrane is its vulnerability to chlorine, which is often incorporated in the drinking water treatment process for disinfection purpose [175]. In principle, polyamide could be chemically degraded when encountering chlorine species, resulting in reduced membrane separation performance [176,177]. Many efforts have been undertaken to design membranes with enhanced anti-chlorine properties, such as synthesizing novel monomer [40] and membrane surface modification [167,178]. For instance, polyester chemistry shows improved chlorine resistance compared to polyamide, which has been extensively studied and utilized as a membrane rejection layer [40,41]. Nevertheless, to fully unleash the potential of the polyester-based membrane, the chemical and mechanical stability of the polyester chemistry needs further systematic investigation, such as the pH tolerance and long-term filtration stability under cross-flow conditions.

8.3 Integrity

Membrane integrity is also a critical aspect influencing membrane separation performance, especially for the rejection of pathogens. However, the integrity of the membrane module is not the primary focus for manufacturers because NF module is primarily designed for total dissolved solids removal. To better remove pathogen in NF, the integrity of NF could be improved by optimizing manufacturing process, and sensitive methods should be established for integrity monitoring. As discussed in Section 5.2, NF membranes may contain intrinsically nanosized defects [132] which would partially contribute to the unsatisfactory rejection of virus (i.e., ~25 nm in size, > 5-log removal) [129]. Accordingly, many strategies have been proposed to minimize defect regions in the rejection layer, such as regulating IP reaction by the addition of surfactant in aqueous amine solution [179], the

incorporation of an inhibitor to control monomer's diffusion [180], and the inclusion of an additional interlayer between the substrate and polyamide rejection layer [26]. Nevertheless, future studies need to systematically address the formation mechanisms and healing strategies to minimize the effects of membrane integrity loss and thus improve membrane separation efficiency.

8.4 Selectivity

Based on the above discussion, a key priority for NF-based drinking water treatment is to effectively remove organic micropollutants and heavy metals with critical concerns, while the removal of salt ions may not be over emphasized in most cases. The high rejection of salt ions (e.g., Ca^{2+} , Mg^{2+} , and SO_4^{2-}) by a NF membrane may not only lead to the severe concentration polarization and further scaling issue, but also compromise the health values of produced drinking water. Instead, a NF membrane with a high selectivity against the targeted pollutants and a relatively low removal of salt is preferred. The selectivity of NF membrane can be tailored through suppressing the passage of targeted solutes and/or accelerating water transport across the membrane. For example, membrane surface properties (e.g., charge [181], pore size/distribution [15], and hydrophilicity [121]) can be tuned through surface modification to reduce the partition of pollutants into membrane thereby enhancing their rejections. Introduction of additional components (e.g., nanofillers and interlayer) into the membrane matrix (e.g., TFN and TFNi membranes) could often facilitate water transport across the membrane because of the creation of additional water channels and/or reduced membrane thickness [24,26]. Nevertheless, a systematic understanding on the selectivity of NF membranes is still lack in the existing literature. Following research efforts are needed to build a concise and effective framework to guide the optimization of NF membranes targeting for highly selective removal of pollutants in drinking water treatment.

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