

Potable Water Reuse through Advanced Membrane Technology

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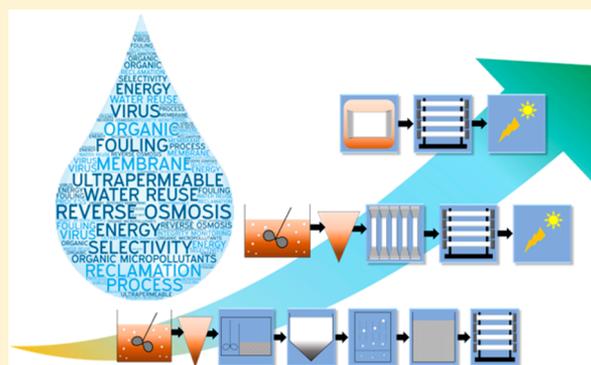
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ABSTRACT: Recycling water from municipal wastewater offers a reliable and sustainable solution to cities and regions facing shortage of water supply. Places including California and Singapore have developed advanced water reuse programs as an integral part of their water management strategy. Membrane technology, particularly reverse osmosis, has been playing a key role in producing high quality recycled water. This feature paper highlights the current status and future perspectives of advanced membrane processes to meet potable water reuse. Recent advances in membrane materials and process configurations are presented and opportunities and challenges are identified in the context of water reuse.



INTRODUCTION

Potable water reuse has become an important indispensable component of the water infrastructure in many cities and regions around the world to address water scarcity.^{1–4} Membrane technology, particularly reverse osmosis (RO), has played a key role in producing highly purified recycled water. Compared to alternative technologies such as activated carbon adsorption and soil aquifer treatment, RO provides better assurance for safe potable applications thanks to its ability to simultaneously remove a broad range of contaminants including total dissolved solids, pathogens (viruses and bacteria) and low molecular chemical contaminants.^{1,2}

As a notable example, water supply of Southern California traditionally relied heavily (about two-thirds) on imported water, whose availability has shrunk significantly over the last four decades due to more upstream demand, stringent environmental regulations and multiyear droughts. Severe overdraft of groundwater since 1940s has caused declining groundwater levels and seawater intrusion that contaminated freshwater aquifers.⁵ In the 1970s, Orange County Water District in Southern California started its Water Factory 21 (WF21), which employed RO-based advanced treatment processes to produce high quality recycled water for direct injection to the drinking water aquifers.⁶ Since 2008, a

new Groundwater Replenishment System (GWRS) has replaced WF21 to produce 70 million gallons per day (MGD) of highly purified water using RO technology.⁷ This world's largest advanced wastewater reclamation system for potable reuse has expanded its production to 100 MGD in 2015, with an ultimate capacity of 130 MGD to be completed by 2023.

Advancement in membrane technology in recent years has increased the number of water reuse projects worldwide. In California alone, several additional major projects have been implemented or planned (Figure 1a), including the 40-MGD Edward C. Little Water Recycling Facility,⁸ a potential 150-MGD Regional Recycled Water Program in Metropolitan Water District of Southern California,⁹ and a scheduled Groundwater Reliability Improvement Project (GRIP)¹⁰ to produce recycled water in 2018. Water reuse has gone far beyond any single region or country, stretching from the United States, Singapore in the Far East, South and Western Europe to Australia in the southern hemisphere.^{1–4} In Singapore, the five NEWater plants provide a total of 170 MGD, or 40% of the nation's water supply.¹¹ This number is scheduled to be increased to 55% by 2060. Australia commissioned the 20 MGD Beenyup plant,¹² the first RO plant in Australia for

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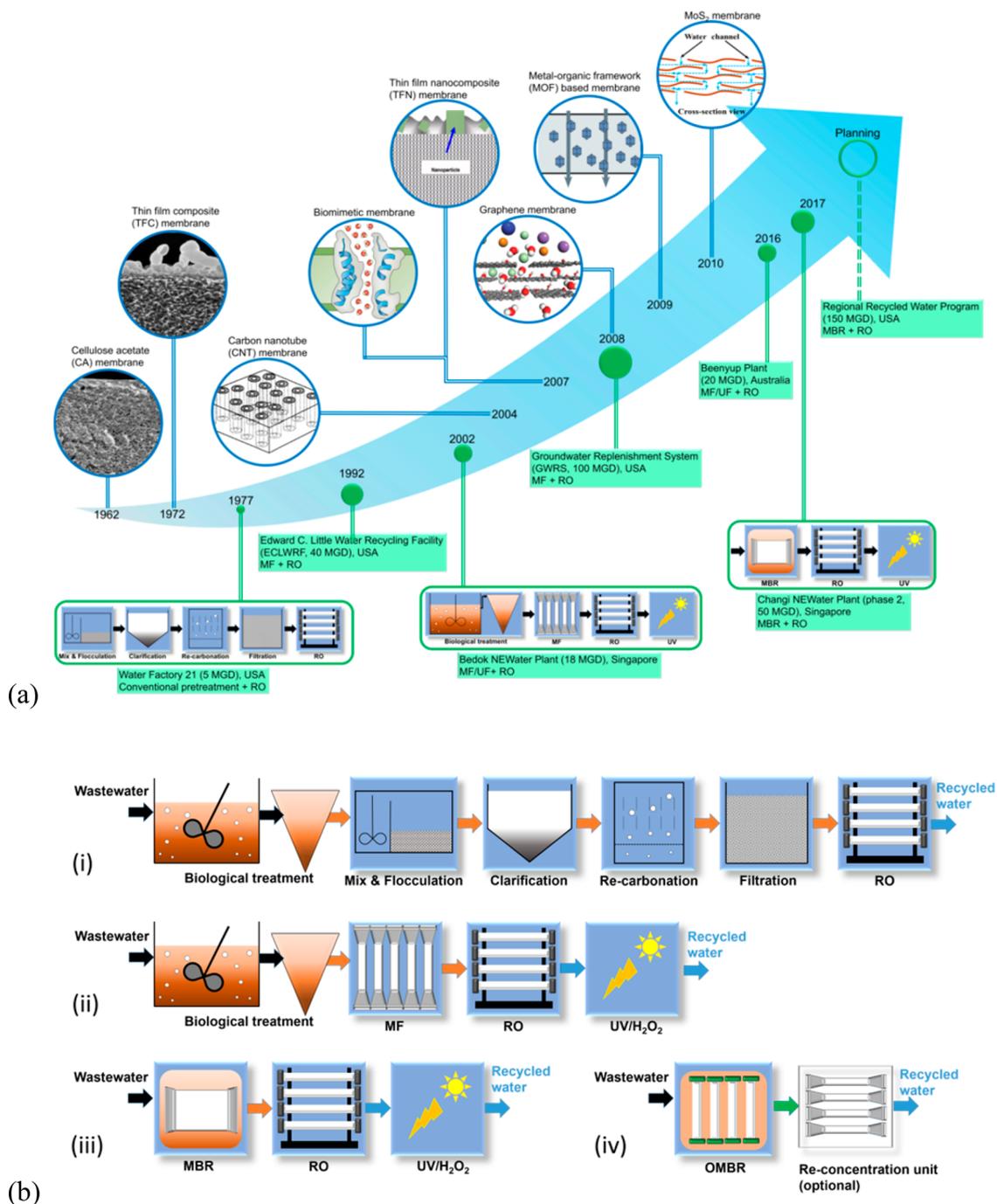


Figure 1. Developments of membrane-based wastewater reuse. (a) Historical development. The lower part of the figure shows notable examples of wastewater reuse plants together with their treatment schemes. The size of the sphere represents the relative size of a plant. The upper part of the figure presents the development of new desalting membranes. CA, TFC, and TFN membranes have already been commercialized, whereas most reported CNT, graphene, MOF, and MoS₂ membranes are synthesized at bench or miniature scales. The respective years of first appearance of aquaporin, graphene, MOF and MoS₂ membranes are based on the year of publication (refs 14–17). The images of the membranes in the upper part of the figure are reprinted with copyright permissions: CA and TFC membranes from ref 18, CNT membrane from ref 19, biomimetic membrane from ref 20, TFN membrane from ref 21, graphene membrane from ref 22, MOF membrane from ref 23, and MoS₂ membrane from ref 24. (b) Evolution of membrane-based water reuse: (i) conventional pretreatment of secondary effluent followed by RO; (ii) MF pretreatment of secondary effluent followed by RO, where an additional UV/H₂O₂ post-treatment may be used for the further removal of organic micropollutants; (iii) MBR-RO treatment, where an additional UV/H₂O₂ post-treatment may be used for the further removal of organic micropollutants; and (iv) OMBR with an optional draw solution reconcentration unit.

indirect potable reuse, in 2016. In parallel to the development of large scale reuse activities, plants of smaller sizes are also spreading all over the world (e.g., Belgium, South Africa, Namibia, the United Kingdom, and the United States).^{1–4,13}

With water scarcity becoming an increasingly serious threat globally,^{25,26} the thirst for water reuse is growing. This feature paper examines the evolution of membrane-based water reuse technology and highlights future opportunities and challenges in this field.

■ EVOLUTION OF MEMBRANE-BASED WATER REUSE

Reverse osmosis (RO) is a well-established technology that can be used in combination with other complementary processes (for pretreatment to remove particulate matter and posttreatment to ensure the destruction of any remaining micropollutants^{27–30}) to produce high quality recycled water (Figure 1a,b). WF21 in Southern California introduced the first RO plant in the world in 1977 to purify reclaimed water to meet drinking water standards.⁶ This 5 MGD RO plant was used to reduce the total dissolved solids of secondary effluent after pretreatment by conventional lime clarification, recarbonation, and multimedia filtration (Figure 1b(i)). In modern potable reuse plants, conventional pretreatment is often replaced by a single microfiltration (MF) process (Figure 1b(ii)), which is more compact and efficient for the removal of particulates. In addition, downstream low pressure-high intensity ultraviolet light with hydrogen peroxide (UV/H₂O₂) is typically used to ensure adequate destruction of small molecular weight micropollutants such as *N*-nitrosodimethylamine (NDMA) that cannot be completely removed by RO membranes.³¹ This advanced MF-RO-UV/H₂O₂ treatment scheme has been widely adopted in numerous potable reuse plants.^{7,11–13} A further significant improvement is the direct treatment of membrane bioreactor (MBR) effluent by RO (Figure 1b(iii)). In this new treatment scheme, the MBR achieves simultaneous roles of bioreactor, biomass separation, and RO pretreatment.^{32,33} The elimination of further RO pretreatment using the particulate-free MBR effluent translates into additional savings of space, energy, and cost, which prompts Changi NEWater Plant in Singapore to adopt the MBR-RO-UV/H₂O₂ scheme.¹¹

Despite these significant improvements over the last few decades, water reuse is still facing many challenges. A report on water reuse issued by the U.S. National Academies in 2012¹ highlighted the critical need to provide quality assurance with respect to pathogens and micropollutants. Issues of high energy consumption, membrane fouling, and concentrate disposal (particularly for inland RO water recycling plants) need to be further addressed through process innovation and novel membrane development (Table 1).

Table 1. Technical Challenges and Research Needs for Membrane-Based Water Reuse

challenges	research needs
high energy consumption	novel low-energy membrane processes and hybrid processes; removal; novel with high permeability and high selectivity
fouling	process design for better antifouling performance; antifouling membranes
micropollutants removal	novel membranes tailored for micropollutants removal novel process design (e.g., osmotic membrane bioreactor)
pathogens removal	membrane integrity assurance and real-time monitoring
concentrate disposal	novel hybrid process development

■ PROCESS INNOVATION

Alternative membrane processes such as forward osmosis (FO)^{34–37} have been explored for water reuse in recent years. FO itself is a low-energy process, since water transports through a dense semipermeable membrane spontaneously using a high osmotic pressure draw solution instead of hydraulic pressure.³⁸

One key challenge for FO is the energy-intensive reconcentration of draw solution for clean water production.³⁷ Applications that do not require draw solution reconcentration, such as osmotic dilution of seawater or brine with wastewater,^{39,40} are also gaining more attention. An osmotic membrane bioreactor (OMBR) patented in 2005⁴¹ is another innovative technique for the reclamation of wastewater, which combines activated sludge treatment and forward osmosis in a single unit process (Figure 1b(iv)).^{42,43} Compared to the MBR-RO scheme (Figure 1b(iii)), OMBR can be potentially more compact and less energy intensive for niche applications where draw solutions do not need to be reconcentrated. Other potential for osmotic membrane bioreactors include the high rejection of micropollutants⁴⁴ and the simultaneous recovery of water, minerals, and nutrients.^{45,46} Recent extension to anaerobic OMBRs further allow the recovery of energy in the form of biomethane.⁴⁷ Future studies shall further address the challenges of membrane fouling,⁴⁸ salinity accumulation in the bioreactor,⁴⁹ and membrane stability⁵⁰ in order to enable its full scale applications. Development of membrane-based hybrid processes also has the potential to address the issue of brine disposal (e.g., the hybridization of RO, FO, and/or membrane distillation with a crystallizer).⁵¹

■ MEMBRANE DEVELOPMENT

The state-of-the-art thin film composite (TFC) polyamide membranes are prepared by the interfacial polymerization reaction of amine monomers (typically *m*-phenylenediamine or MPD) and trimesoyl chloride (TMC).⁵² Commercial TFC polyamide membranes have a wide range of pH tolerance (pH 2–11), excellent mechanical stability (up to several MPa of applied pressure), high salt rejection (e.g., NaCl rejection of up to 99.8%) and yet a moderate water permeability (e.g., 1–8 L/m²-h-bar).^{53,54} Unlike seawater desalination whose energy consumption (~4 kWh/m³) is mainly dictated by the high osmotic pressure of seawater (~2.7 MPa), the energy consumption in RO-based water reuse (~1 kWh/m³ with approximately 0.555 kWh/m³ for RO¹³) is governed mostly by membrane resistance and fouling. Tripling membrane water permeability can potentially reduce the energy consumption for potable reuse by half.⁵⁵ Thus, developing low-pressure RO membranes with high permeability and good antifouling performance deserves to be a top research priority.

Nanocomposite Membranes. A new type of RO membranes, known as thin film nanocomposite (TFN) membranes, were developed by Hoek and co-workers in 2007.²¹ In this novel approach, zeolite nanoparticles of defined pore size are included into the polyamide rejection layer during an interfacial polymerization (Figure 2a). The inclusion of porous zeolite nanoparticles enhances the resulting membrane permeability while maintaining its salt rejection. The ease of fabricating TFN membranes at relatively cheap cost allows its commercial scale up.⁵⁶ In the meantime, many other materials, such as nanoparticles of silver, silica, or zinc oxide, have been extensively studied for the synthesis of TFN membranes,^{57,58} although the majority of the studies were performed at bench scale.

Nanostructured Polyamide Membranes. Another effective way to increase the membrane permeability is by reducing the thickness of the polyamide rejection layer. Gu and co-workers introduced a molecular layer-by-layer (mLBL) membrane.⁵⁹ In their approach, an ultrathin polyamide rejection layer was prepared by alternative soaking of a substrate in low-concentration MPD and TMC solutions for repeated cycles

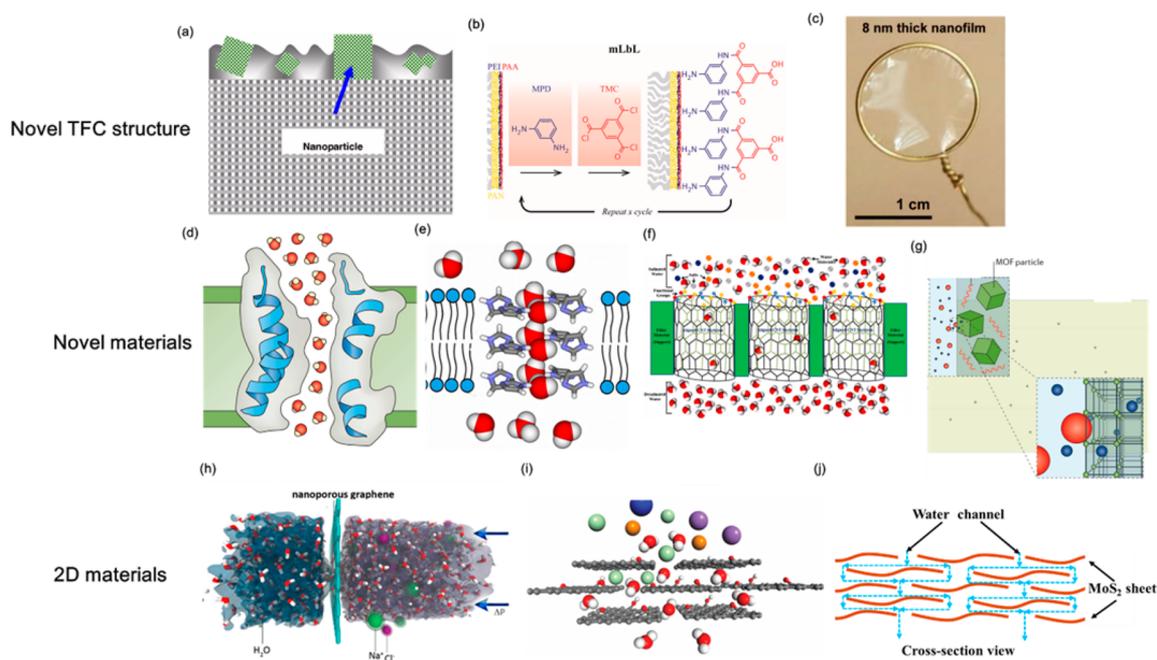


Figure 2. Novel RO membranes. Polyamide membranes include (a) a thin-film nanocomposite membrane with nanomaterials embedded into polyamide rejection layer,²¹ (b) a molecular layer-by-layer (mLBL) membrane fabricated by repeated cycles of interfacial polymerization of MPD with TMC,⁵⁹ and (c) a sub-10 nm-thick polyamide rejection layer fabricated by performing interfacial polymerization reaction on a sacrificial nanostrand interlayer.⁶⁰ Examples of emerging materials for high performance membranes include: (d) aquaporins,²⁰ (e) artificial water channel,⁶¹ (f) carbon nanotubes,⁶² (g) metal–organic frameworks,²³ (h) nanoporous graphene monolayers,⁶³ (i) graphene oxide frameworks,²² and (j) molybdenum disulfide (MoS_2) frameworks.²⁴ All figures are reprinted with copyright permissions from the respective references.

(Figure 2b). A mLBL membrane of 20–25 nm in thickness was prepared, which show 75% improvement in water permeability and similar NaCl rejection compared to a control TFC membrane prepared by conventional interfacial polymerization with rejection layer thickness of 110 nm. Livingston and co-workers⁶⁰ prepared an ultrathin polyamide membrane by performing interfacial polymerization reaction on a sacrificial layer of nanostrands. The presence of the nanostrand layer significantly reduced the diffusion of MPD monomers, which resulted in an ultrathin and smooth polyamide rejection layer of less than 10 nm in thickness (Figure 2c). Though the resultant membrane showed excellent permeability of more than two orders of magnitude higher than a commercial benchmark and similar selectivity, this method is unfortunately difficult to scale up. By electro spraying MPD and TMC monomer solutions into microdroplets for subsequent interfacial polymerization, Tang and co-workers demonstrated finely controlled growth of a polyamide rejection film at 1 nm/min.⁶⁴ This electro spray-assisted additive interfacial polymerization approach, a method that can be more easily scaled up, was able to prepare uniform ultrathin polyamide membranes of four to a few tens of nm in thickness.

Researchers are also paying increasing attention to the nanostructures of the polyamide rejection layer. A recent work demonstrated the feasibility to simultaneously increase the permeability and rejection of a TFC polyamide membrane by promoting the formation of nanosized gas bubbles to tune the membrane surface roughness.⁶⁵ This nanofoaming strategy can be easily realized by adding nanobubble precursors to the monomers solutions, for example, dosing NaHCO_3 into the MPD solution. Zhang and coworkers⁶⁶ developed another simple method to tune the membrane surface roughness using additives such as poly(vinyl alcohol) and demonstrated an

order of magnitude improvement in membrane water permeability. Several other researchers also show the possibility of order of magnitude improvement in permeability by including an interfacial layer (e.g., tannic acid–Fe complex,⁶⁷ polydopamine^{68,69} or CNTs⁷⁰) between the polyamide and its polysulfone support.

Next Generation Desalting Materials and Membranes. In recent years, novel materials have emerged as potential candidates for preparation of high performance RO membranes.^{20,71} One type of promising material is aquaporins (Figure 2d), or water channel proteins, that are found in cellular membranes for delivering water across biological cells with permeabilities of 2–3 orders of magnitude higher than the best commercially available RO membranes and with nearly complete rejection of solutes including H^+ .^{72–74} Synthetic channels and porous materials have also been investigated for their use in synthesizing ultrapermeable membranes; some of the most notable examples include self-assembled artificial water channels,⁶¹ carbon nanotubes (CNTs),^{15,19,62} microporous metal–organic frameworks (MOFs),^{56,75–77} graphene,⁶³ graphene oxide,^{78–80} and MoS_2 .^{81,82} (Figure 2e–j). Their intrinsic ultrafast water transport rates can potentially half the energy consumption for water reuse.⁵⁵ Nevertheless, a recent review highlights the challenges of defects prevention (for achieving high rejection) and scaling up (for commercial scale production).⁷¹ Indeed, most of the reported membranes prepared by these novel desalting materials have NaCl rejections of only ~ or <90%, which are significantly below commercial benchmarks. A compromised approach is to incorporate these materials in a thin film nanocomposite structure, which can effectively maintain salt rejection at the expense of water permeability.⁷¹ For the case of aquaporins, their proteinaceous nature also makes them unstable to directly withstand the harsh wastewater environment.

In contrast, the thin film nanocomposite⁸³ approach provides a much better protection to the aquaporins.⁸⁴

Antifouling Membranes. Developing membranes that are resistant to fouling, particularly biofouling, is a priority research area in the context of membrane-based water reuse. Various strategies have been developed to enhance antifouling performance of membranes, which often involves surface coating, grafting, and immobilization of antiadhesion and/or biocidal agents.⁸⁵ These approaches are generally designed to modify a membrane's hydrophilicity, surface charge, and/or roughness, or to impart antimicrobial moieties. Some notable examples of antifouling enhancement include the preparation of antiadhesion surfaces using poly(vinyl alcohol) grafting,⁸⁶ polydopamine coating,⁸⁷ and zwitterionic grafting⁸⁸ and the immobilization of antibacterial silver/copper nanoparticles.^{89–91}

■ ORGANIC MICROPOLLUTANTS REMOVAL

The presence of micropollutants in wastewater is a significant issue for membrane-based water reuse. NDMA is a notorious disinfectant byproduct and a human carcinogen that is frequently detected in RO permeate.^{31,92} California has set a Public Health Goal of 3 ng/L and a notification level of 10 ng/L for this suspected carcinogen.⁹³ NDMA rejection by RO membranes is in the range from 20 to 80%.^{92,94,95} Post-treatment by advanced oxidation processes, such as UV treatment, is effective in destructing NDMA. Nevertheless, it generally requires a very high UV intensity (e.g., 1000 MJ/cm²), a dosage of an order of magnitude higher than that used for UV disinfection.⁹² Besides NDMA, other micropollutants of concern include endocrine disruptors and pharmaceutically active compounds.^{96–98}

Due to their historical roots in desalination, commercial thin film composite polyamide RO membranes have been highly optimized for salt rejection and water permeability, yet they are often not adequate for the removal of micropollutants, particularly small polar or hydrophobic organic compounds. In recent years, researchers have started to realize the need for designing membranes specifically for micropollutants removal. Tailoring membrane surface properties by surface coating/grafting show some promising results.^{99–102} For instance, a hydrophilic polydopamine coating can effectively half the passage of hydrophobic EDCs through a polyamide membrane.¹⁰¹ To reduce the adverse effect on water permeability, materials of high selectivity against micropollutants are needed.¹⁰³ In this regard, some of the novel desalting materials such as aquaporins and MOFs are of great interest due to their highly defined pore structure and high specificity for water. Recent studies on aquaporin-embedded polyamide membranes showed improved rejection rates to a wide range of micropollutants.^{104,105} Graphene oxide sheets that are capable of forming highly hydrophilic water channels have also demonstrated great potential for micropollutants removal.^{106–108}

■ PATHOGEN REMOVAL AND MEMBRANE INTEGRITY MONITORING

An important advantage of RO is the high removal of viruses and bacteria (disinfection), of which viruses are most important because of their smaller size, typically 20–300 nm.^{109,110} High pressure RO will remove viruses due to steric hindrance, since viruses are much larger than membrane pores. Despite of this, RO systems can fail due to broken seals, damaged glue lines and oxidized membranes (related to chemical cleaning),

undesired back pressure damage from permeate side, abrasion by particles in the feed, etc.^{111,112}

Given the central role of RO in potable water reuse, several highly sensitive sensors have been developed to monitor chemical and microbial contaminants on a real-time or near real-time basis for membrane integrity assurance. In addition to traditional surrogate parameters such as conductivity, total organic carbon, and sulfate which can be readily monitored online, several new surrogates specific to potable water reuse have been added in recent years. They include UV254 or fluorescence for monitoring organic micropollutants and multiple light scattering or measurement of adenosine triphosphate for monitoring microbial contaminants. Nevertheless, currently there are no techniques to directly determine RO integrity for more than 2 log virus removal. Therefore, in the context of membrane integrity against virus removal, there is a strong need for novel methods containing: (i) high log removal values (≥ 4 log removal), (ii) surrogate/indicator virus with same size as natural virus size, (iii) inexpensive and fast method, (iv) real time and online monitoring, and (v) meeting the criteria for drinking water application.

Online monitoring of micropollutants can be another fruitful opportunity. Of a particular note, Fujioka et al.,¹¹³ have successfully developed an analytical technique consisting of high-performance liquid chromatography followed by photochemical reaction and chemiluminescence detection (HPLC PR-CL) for online monitoring of NDMA and several other *N*-nitrosamines in secondary treated effluent and RO permeate. The detection limit of their technique (0.3–2.7 ng/L) is comparable to the regulated concentrations of these organic micropollutants in most potable water reuse guidelines or standards. The HPLC PR-CL developed by Fujioka et al., marks a significant milestone as this is the first time target organic micropollutants can be monitored in near real-time.

■ THE SWEET SPOTS

Figure 3 compares the different technologies based on three individual dimensions: the current scale of development (horizontal axis), the ease of scale up (vertical axis), and the potential impact to water reuse (size of the symbol). Three important quadrants are shown in the figure. The upper right quadrant represents technologies that are already commercialized or near to commercialization, whereas the lower left quadrant represents technologies that are far away from commercialization. Sweet spots exist in the upper left quadrant: even though the relevant technologies are still at the bench-scale development, it is relatively easy to scale up them. For example, high performance nanostructured polyamide membranes can be potentially produced with only minor changes to the existing fabrication lines of TFC RO membranes (e.g., with addition of NaHCO₃⁶⁵ or poly(vinyl alcohol)⁶⁶ in the amine monomer solution). At the same time, the resulting improvements in membrane water permeability and selectivity can dramatically reduce the energy consumption and improve the product water quality. Other areas of paramount significance to water reuse include integrity monitoring and membranes for enhanced micropollutant removal, which deserves greater attention in future development.

■ BRIDGING THE GAP

The challenge of implementing water reuse is not confined solely to the technical domain. Public acceptance is a complex

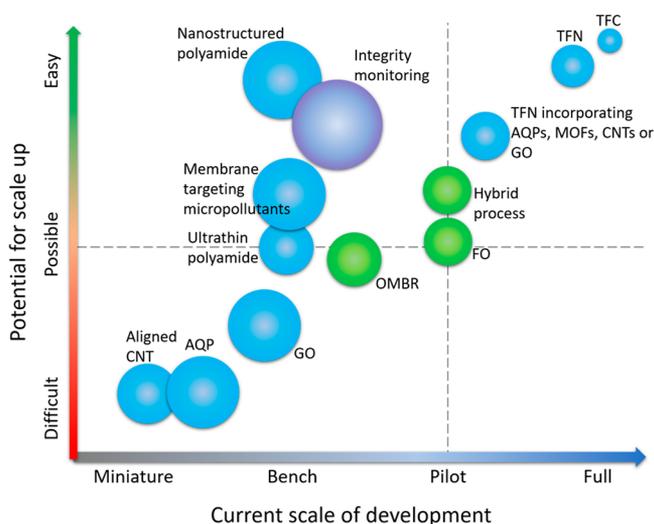


Figure 3. Outlook of water reuse technologies. The horizontal axis classifies different technologies into full (commercial) scale, pilot scale, bench scale, and miniature scale. The vertical axis classifies the technologies based on their potential for scale up, with considerations based on technical difficulties as well as potential cost of production (see references^{56,71}). The size of the sphere represents the potential impact of a particular technology. A larger sphere indicates a great potential to improve water reuse (e.g., enhanced reliability, reduced cost, and energy consumption, and/or improved water quality).

and thorny issue, one that has derailed a number of water reuse projects in the past.^{114,115} A particular high profile case is that of Toowoomba in Australia, where intense debate about a proposed indirect potable reuse scheme led to a referendum.¹¹⁶ As the result of the referendum, in which 60% of the participants opposed the scheme, it was abandoned.¹¹⁶ Toowoomba has been seen as the trigger point for the Queensland government in Australia to abandon the Western Corridor Recycled Water project, which was completed in 2009 but has never been used as intended. The fallout from Toowoomba underscores the need to fully understand the connection between public perception about water reuse and technological innovation.¹¹⁷

Public Acceptance. Since Toowoomba, significant efforts often by collaborations between social scientists and engineers, and practitioners in the water sector have been made to positively influence public perception about water reuse. Providing information about the treatment processes significantly increased public acceptance of water reuse.¹¹⁸ Experiential activities such as field visits, tasting opportunities, using reused water for public swimming pools, and water splash pads can also be helpful.¹¹ These efforts have resulted in better awareness by the public about the reliability and efficiency of membrane separation and other technologies used for water reclamation, and hence, a gradual shift in public acceptance and a growing number of successful water reuse schemes in recent years.¹¹⁴ As a notable example, strong public support to water reuse in Singapore can be attributed, at least in part, to a very concerted and systematic public engagement program that includes the attractive NEWater Visitor Centre at the Bedok plant.¹¹ The center has effectively become a tourist attraction, where the public can book a tour for free to learn about how Singapore copes with their water supply problem and be given a bottle of NEWater (reused water) as souvenir or for tasting. Efforts to garner public support to potable reuse has evolved

beyond simple marketing activities. Harris-Lovett et al.¹¹⁹ argued that establishing legitimacy for potable water reuse involves embedding RO, advanced oxidation, and other new technologies in the shared social belief system, moral standards, and cultural conventions through a set of strategies that go beyond traditional public relations and educational outreach.

Public Trust and Technical Reliability. A key component of the legitimacy framework proposed by Harris-Lovett et al.¹¹⁹ is reliable risk management procedures. A promising strategy is to make key innovations in potable water reuse namely membrane separation and other advanced technologies more understandable by relating to standards and procedures that have already gained legitimacy in other established sectors. Online monitoring is essential not only for establishing a safety record but also effective risk management. Indeed, while acknowledging the central role of technology innovation, Lee and Tan¹²⁰ accredited Singapore's success in supplying NEWater for potable use to an extensive data acquisition program to demonstrate the safety record of potable water reuse. Prior to the NEWater, the Singapore Public Utilities Board collected some 20 000 test results from different sampling locations in a demonstration plant, covering about 190 physical, chemical and microbiological parameters.¹²⁰ The results were benchmarked against the World Health Organization and United States Environmental Protection Agency drinking water standards to demonstrate the credibility of potable water reuse.

With a focus on public safety, real-time monitoring has been a crucial strategy for assurance and risk management of potable water reuse. Real-time monitoring offers an opportunity to engage with the public as well as quickly detect and rectify failure. Aside from routine monitoring of membrane separation performance through conductivity and total organic carbon, advanced real time monitoring methods for viruses and micropollutants can be highly valuable. Further development in online monitoring of RO performance along with other improvements in membranes and processes can be expected and will help to bridge the gap between technology innovation and public confidence in potable water reuse.

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Notes

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