Potable water reuse through advanced membrane technology

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Abstract

Recycling water from municipal wastewater offers a reliable and sustainable solution to cities and regions facing shortage in conventional 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 historical development, current status and future perspectives of advanced membrane processes to meet both indirect and direct potable 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. 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 multi-year droughts. Severe overdraft of groundwater since 1940s caused declining groundwater levels and seawater intrusion that contaminated freshwater aquifers.[1] In the 1970s, Orange County Water District in Southern California started its Water Factory 21 (WF21), which employed advanced treatment processes to produce high quality recycled water for direct injection to the drinking water aquifers.[2] Since 2008, a new Groundwater Replenishment System (GWRS) has replaced WF21 to produce 70 MGD of highly purified water using reverse osmosis (RO) technology.[3] 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.

Membrane technology, particularly RO, has played a key role in producing highly purified recycled water for potable reuse. 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 broader range of contaminants including total dissolved solids, pathogenic agents, and organic micropollutants [ref.]. Advancement in membrane technology in recent years has increased the number of water reuse projects worldwide (Figure 1). In California alone, several additional major projects have been implemented or planned, including the 40-MGD Edward C. Little Water Recycling Facility,[4] a potential 150-MGD Regional Recycled Water Program in Metropolitan Water District of Southern California,[5] and a scheduled Groundwater Reliability Improvement Project (GRIP) [6] 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. In Singapore, the five NEWater plants provide a total of 170 MGD, or 40% of the nation's water supply.[7] This number is scheduled to be increased to 55% by 2060. Other notable examples include: the 20 MGD Beenyup plant commissioned in Perth, Western Australia in 2016, which is the first RO plant in Australia for indirect potable reuse; … ~~In Belgium (Europe) the Intermunicipal Water Company of Veurne-Ambacht (IWVA) treats secondary wastewater effluent at the Torreele facility for indirect potable reuse via groundwater recharge in the dune water catchment of St. André.[8] This 2 MGD has been in operation since 2002 and has brackish water reverse osmosis at its core for high quality water production. In Australia, after a successful full-scale trial over 5 years, in 2016, the Beenyup Advanced Water Recycling plant was officially opened in Perth as the first indirect potable reuse scheme in the country. In conjunction with aquifer storage and recovery, the Beenyup plant can produce 20 MGD of recycled water, which is enough to supply up to 100,000 households in Perth.~~

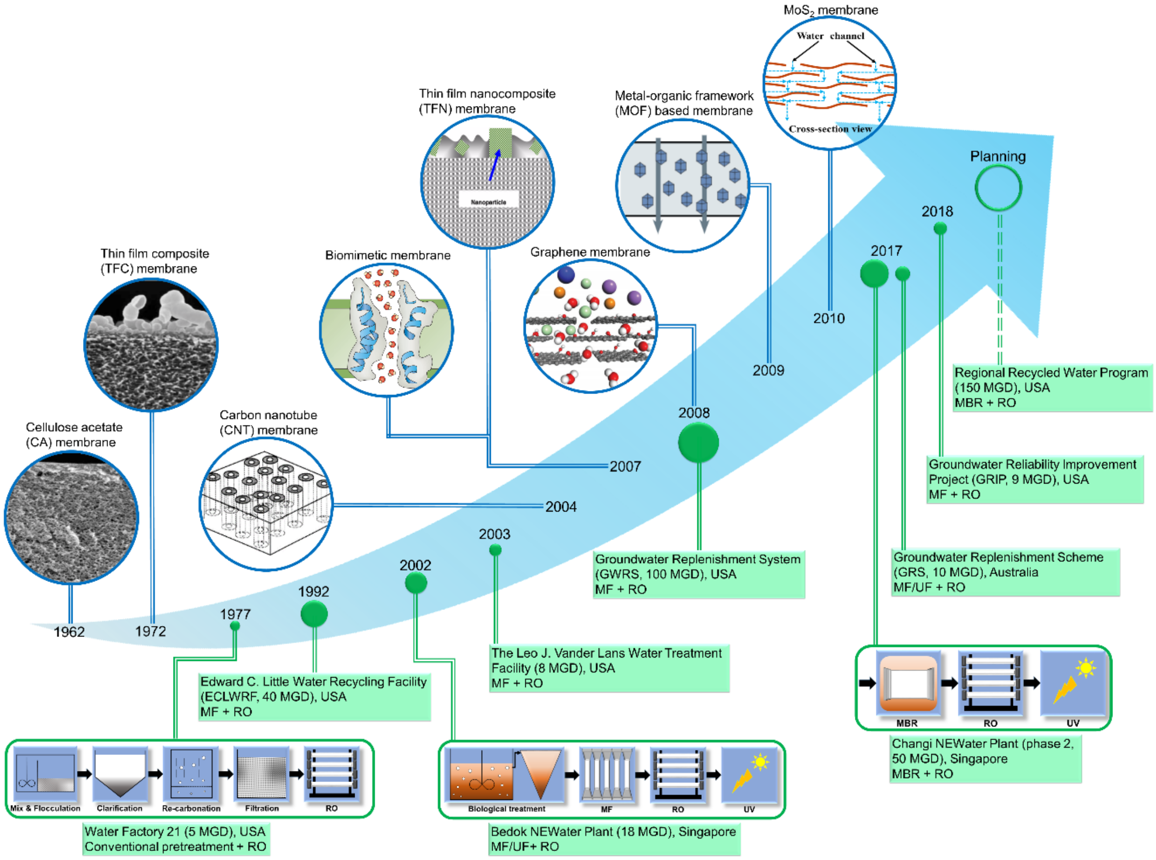

Figure 1. Historical developments of membrane-based wastewater reuse. The lower part of the figure shows notable examples of wastewater reuse plants together with their treatment schemes. The size of the sphere represents the relative size of a plant. The upper part of the figure presents the development of new desalting membranes. The respective years of first appearance of aquaporin, graphene, MOF and MoS$_2$ membranes are based on Refs [9-12]. The images of the membranes in the upper part of the figure are reprinted with copyright permissions: CA and TFC membranes from Ref. [13], CNT membrane from Ref. [14], biomimetic membrane from Ref. [15], TFN membrane from Ref. [16], graphene membrane from Ref. [17], MOF membrane from Ref. [18] and MoS$_2$ membrane from Ref. [19].

With water scarcity becoming an increasingly serious threat globally,[20] 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 [21-24] and remineralization in the case of potable reuse) to produce high quality recycled water (Figure 1 and Figure 2). WF21 in Southern California introduced the first RO plant in the world in 1977 to purify reclaimed water to meet drinking water standards.[2] 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 2a). In modern potable reuse plants, conventional pretreatment is often replaced by a single microfiltration (MF) process (Figure 2b), which is more compact and efficient for the removal of particulates. In addition, downstream low pressure-high intensity ultraviolet light with hydrogen peroxide (UV/H_2O_2) is typically used to ensure adequate destruction of small molecular weight micropollutants such as N-nitrosodimethylamine (NDMA), N-nitrosodiethylamine (NDEA), and 1,4=Dioxane that cannot be completely removed by RO membranes.[25] This advanced MF-RO-UV/H_2O_2 treatment scheme has been widely adopted in many potable reuse plants, such as the grand-scale GWRS in Southern California [3], the Bedok and Kranji NEWater plants in Singapore [7], the Beenyup plant in Australia (ref?), and the Toreele Reuse plant in Belgium (ref?). A further significant improvement is the direct treatment of membrane bioreactor (MBR) effluent by RO (Figure 2c). In this new treatment scheme, the MBR achieves simultaneous roles of bioreactor, biomass separation, and RO pretreatment.[26, 27] 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/H_2O_2 scheme.[7]

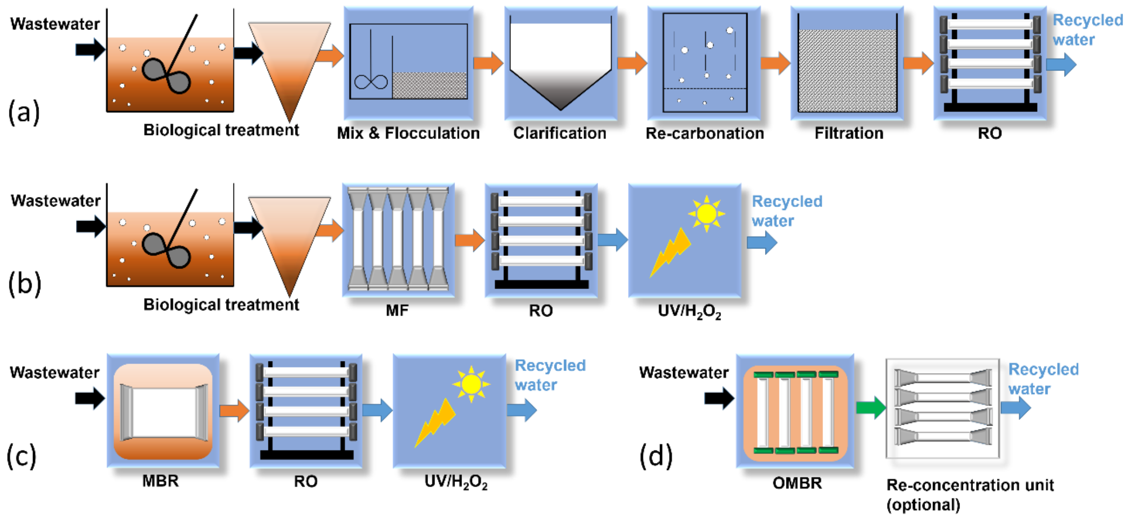

Figure 2. Evolution of membrane-based water reuse: (a) conventional pre-treatment of secondary effluent followed by RO; (b) MF pre-treatment of secondary effluent followed by RO, where an additional UV/H_2O_2 post-treatment may be used for the further removal of organic micropollutants; (c) MBR-RO treatment, where an additional UV/H_2O_2 post-treatment may be used for the further removal of organic micropollutants; and (d) OMBR with an optional draw solution re-concentration unit. Some OMBR applications (e.g., using fertilizer-based draw solution) do not require the re-concentration unit.

Alternative membrane processes such as forward osmosis (FO) [28-31] have also been explored for water reuse. FO-based processes, in which water transports through a dense semi-permeable membrane using a high osmotic pressure draw solution, are interesting due to their better ability to deal with difficult-to-treat waste streams (e.g., with high organic loading).[32] One key challenge for FO is the energy-intensive re-concentration of draw solution for clean water production.[31] To overcome this issue, Shon and co-workers [33] developed a fertilizer-drawn FO process, in which the FO permeate water can be reused for fertigation. Other applications that do not require draw solution re-concentration, such as osmotic dilution of seawater or brine with wastewater [34, 35], are also gaining more attention. Nevertheless, economic benefits of water reuse through osmotic dilution are yet to be proven.[29]

An osmotic membrane bioreactor (OMBR) patented in 2005 [36] is an innovative MBR technique for the reclamation of wastewater, which combines activated sludge treatment and forward osmosis in a single unit process (Figure 2d).[37, 38] Compared to the MBR-RO scheme, OMBR can be potentially more compact and less energy intensive for niche applications where draw solutions do not need to be re-concentrated (e.g., by using fertilizer draw solutions). Other potential for osmotic membrane bioreactors include the high rejection of micropollutants [39] and the simultaneous recovery of water, mineral, and nutrient [40, 41]. Recent extension to anaerobic OMBRs further allow the recovery of energy in the form of biomethane.[42] Nevertheless, challenges of membrane fouling [43], salinity accumulation in the bioreactor [44], and membrane stability [45] need to be further addressed to enable its full scale applications.

▪ TOWARDS BETTER PERFORMANCE MEMBRANES

Membranes play a critical role in RO-based water reuse. The quest for high-permeability and high-selectivity RO membranes is summarized in Figure 1. The first-generation RO membranes are of cellulose acetate with an asymmetrical structure. With the development of more permeable and more selective thin film composite (TFC) polyamide membranes in the 1970s, existing commercial RO membranes are largely dominated by the latter.[46] A typical TFC RO membrane consists of a polyamide rejection layer of several tens to hundreds of nanometers in thickness, which is formed by an interfacial polymerization reaction of an amine monomer (typically m-phenylenediamine or MPD) and an acyl chloride monomer (typically trimesoyl chloride) on an ultrafiltration support substrate.[46] 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 99.7%) and yet a moderate water permeability (e.g., 1-8 L/($m^2h^1bar^1$)).[47, 48] Unlike seawater desalination whose energy consumption (~ 4 kWh/m^3) is mainly dictated by the high osmotic pressure of seawater (~ 2.7 MPa), the energy consumption in RO-based water reuse (~ 1 kWh/m^3 with approximately 0.555 kWh/m^3 for RO[8]) is governed mostly by membrane resistance and fouling. Tripling membrane water permeability can potentially reduce the energy consumption for potable reuse by half.[49] 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 coworkers in 2007.[16] In this novel approach, zeolite nanoparticles of defined pore size are included into the polyamide rejection layer during an interfacial polymerization (Figure 3a). 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.[50] 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 [51, 52], although the majority of the studies were performed at bench scale.

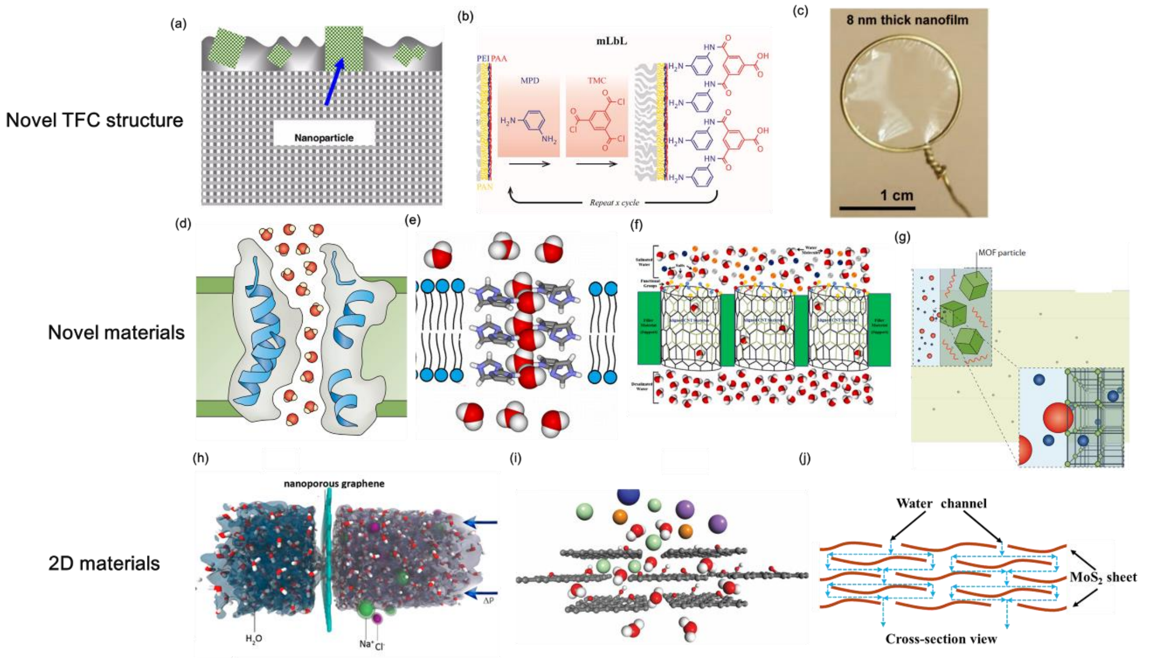

Figure 3. Novel RO membranes. Polyamide membranes include (a) a thin-film nanocomposite membrane with nanomaterials embedded into polyamide rejection layer,[16] (b) a molecular layer-by-layer (mLBL) membrane fabricated by repeated cycles of interfacial polymerization of MPD with TMC,[53] and (c) a sub-10-nm-thick polyamide rejection layer fabricated by performing interfacial polymerization reaction on a sacrificial nanostrand interlayer.[54] Examples of emerging materials for high performance membranes include: (d) aquaporins,[15] (e) artificial water channel,[55] (f) carbon nanotubes,[56] (g) metal-organic frameworks,[18] (h) nanoporous graphene monolayers,[57] (i) graphene oxide frameworks,[17] and molybdenum disulfide (MoS_2) frameworks.[19] All figures are reprinted with copyright permissions from the respective references.

Ultrathin 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.[53] 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 3b). 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-workers [54] prepared an ultrathin polyamide membrane by performing interfacial polymerization reaction on a sacrificial layer of nanostrands. The presence of nanostrand layer significantly reduced the diffusion of MPD monomers, which resulted in an ultra-thin and smooth polyamide rejection layer of less than 10 nm in thickness (Figure 3c). Though the resultant membrane had excellent water 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 electrospraying 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.[58] This electrospray-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.

Next generation desalting materials and membranes. In recent years, novel materials have emerged as potential candidates for preparation of high performance RO membranes.[15, 59] One type of promising material is aquaporins (Figure 3d), 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^+.[60-62] Synthetic channels and porous materials have also been investigated for their use in synthesizing ultra-permeable membranes; some of the most notable examples include self-assembled artificial water channels [55], carbon nanotubes (CNTs) [10, 14, 56], microporous metal-organic frameworks (MOFs) [50, 63, 64], and graphene [57], graphene oxide [65-67], and MoS_2 [68, 69] (Figure 3e-j). Their intrinsic ultra-fast water transport rates can potentially half the energy consumption for water reuse.[49] Nevertheless, a recent review highlights the challenges of defects prevention (for achieving high rejection) and scaling up (for commercial scale production).[59] 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 compromise approach is to incorporate these materials in a thin film nanocomposite structure, which can effectively maintain salt rejection at the expense of water permeability.[59]

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 anti-adhesion and/or biocidal agents.[70] 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 anti-fouling enhancement include polyvinyl alcohol grafting [71], polydopamine coating [72], zwitterionic grafting [73] and silver/copper nanoparticles immobilization [74-76].

▪ DEALING WITH ORGANIC MICROPOLLUTANTS

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.[25, 77] California has set a Public Health Goal of 3 ng/L and a notification level of 10 ng/L for this suspected carcinogen.[78] NDMA rejection by

RO membranes is in the range from 20 to 80%.[77, 79, 80] 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/cm^2), a dosage of an order of magnitude higher than that used for UV disinfection.[77] Besides NDMA, other micropollutants of concern include endocrine disruptors and pharmaceutically active compounds.[81-83]. Recent issues in the Netherlands with discharge of perfluorpolymers and pyrazole in surface water bodies challenged Dutch drinking water facilities. While perflourpolymer rejection by RO membranes is very high (>95%)[84], the pyrazole rejection is low (approximately 35%)[85] depending on the type of RO membrane resulting in the need of a post UV/peroxide treatment.

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 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.[86-89] For instance, a hydrophilic polydopamine coating can effectively half the passage of hydrophobic EDCs through a polyamide membrane.[88] To reduce the adverse effect on water permeability, materials of high selectivity to micropollutants are needed.[90] 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.[91, 92] Graphene oxide sheets that are capable of forming highly hydrophilic water channels have also demonstrated great potential for micropollutants removal.[93-95]

▪ BRIDGING THE GAP

The challenge of implementing water reuse is not confined solely to the technical domain. Public acceptance is a complex and thorny issue, one that has derailed a number of water reuse projects in the past.[96, 97] 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.[98] As the result of the referendum, in which 60% of the participants opposed the scheme, it was abandoned.[98] 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.

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. 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. For examples, the City of San Diego reported an increase in public support of potable water reuse from 26% in 2004 to 73% in 2012 after sustained investment in research and public engagement activities.[96]

The legitimacy of potable reuse. Recent socio-psychological studies have also added considerable depth to our understanding of the complex interactions amongst factors that can influence public acceptance of water reuse. Through an experiment with 1000 Australian correspondents, Dolnicar et al. [100] conclusively observed that providing information about the treatment processes significantly increased public acceptance of water reuse. Proactively working with the media is also an important component of public engagement activities. Ormerod and Silvia analysed 158 newspaper articles about potable water reuse in the Orange County Water District from 2000 to 2016 and did not identify any negative coverage. While some of these articles were positive, the majority was neutral and uncommitted about potable water reuse. These results echo previous findings from Hurlimann and Dolnicar [101] who observe concluded that the media coverage of potable water reuse can often be characterized by lack of inclusion of views, a low level of support statements with scientific evidence, a low level of impartiality, and a high level of hedging language. The implicit uncertainty about the reported information in newspaper coverage highlights the need for better engagement between key stakeholders of potable water reuse and the.

Improving public knowledge alone may not be sufficient to change public perception. In the context of water reuse, there is a pre-cognitive and irrational perception that prevents many people from separating the final product (clean water) and its contaminated source (human excreta).[96] This is despite the fact that no traces of the original contamination exists and that no incident on human health due to water reuse has ever been reported albeit the many intentionally and unintentionally (unplanned) potable water reuse schemes that have been in operation, in some cases, for several decades. One effective strategy to overcome the challenge of irrational public perception provide experiential activities such as field visits, tasting opportunities, using reused water for public swimming pools and water splash pads. 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.[7] The centre 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.

Effort to garner public support to potable reuse has evolved beyond simple marketing activities. Harris-Lovett et al., have recently proposed a framework based on societal legitimacy for engaging the public on issues of potable water reuse. On the same vein, Binz et al., point out that technological innovation is incongruous with established social rules. Thus, given the perceived unprecedented nature of potable water reuse, it is often confronted with strong skepticism and a lack of societal legitimacy. Harris-Lovett et al., 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 Herris-Lovett et al., is reliable risk management procedures. A promising strategy is to make key innovation 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 Tan 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 Utility Board collected some 20,000 test results from different sampling locations in a demonstration plant, covering about 190 physical, chemical and microbiological parameters. The results were benchmarked again the WHO and USEPA 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. 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 multi-able light scattering or measurement of adenosine triphosphate for monitoring microbial contaminants.

Of a particular note, Fujioka et al., 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 N-nitrosodimethylamine (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. Further development in online monitoring of RO performance 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

The authors declare no completing financial interest.

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