An Overview of Osmosis Study in Living Cells and its Implication in Forwarding Osmosis for Water Treatment Application

Ahmad Sofyan Sulaeman 1, Permono Adi Putro 1, Siti Nikmatin 2*  

1 Department of Physics, Faculty of Science, Universitas Mandiri, Subang 41211 Indonesia; ahmadsofyansulaeman@gmail.com (A.S.S); permonoadi29@gmail.com (P.A.P);  
2 Department of Physics, Faculty of Mathematics and Natural Sciences, IPB University, Bogor 166680 Indonesia; snikmatin@apps.ipb.ac.id (S.N); * Correspondence: snikmatin@apps.ipb.ac.id (S.N); Scopus Author ID 56515330600  

Received: 5.01.2022; Accepted: 5.02.2022; Published: 3.04.2022

Abstract: Osmosis is a fundamental water flow process through a selectively semi-permeable membrane. In this study, we overview the osmosis principle in the living cell and the implication of forwarding osmosis (FO). Various draw solutes (DS) and selective membranes are introduced in the FO process to identify their characters. By recognizing the character of the most important part of the FO process, it is hoped to increase knowledge and find the latest development strategies in wastewater treatment and desalination.

Keywords: osmosis; forward osmosis; water treatment; desalination.

Abbreviations: CP= Concentration Polarization; DS= Draw Solutes; FO = Forward Osmosis; ICP= Internal Concentration Polymerization; IP= Interfacial Polymerization; LbL= Layer-by-Layer; LCST= Lower Critical Solution Temperature; NF= Nanofiltration; P-SDS= Polarity-Switchable Draw Solutes; RO= Reverse Osmosis; SPS = Switchable Polarity Solvents; TFC= Thin-Film Composite

© 2022 by the authors. This article is an open-access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https://creativecommons.org/licenses/by/4.0/).

1. Introduction

Osmosis is a universal phenomenon defined as the flux of solvent as water through a selectively semi-permeable membrane [1,2]. Osmosis usually means the establishment of an osmotic gradient such as force and pressure [3]. Its requirement is only to permit water molecules to diffuse through the membrane and is relatively impermeable for solute molecules [1]. It was found in the case of water movements across the cell membranes [4], resulting in some tonicities conditions such as hypotonic, isotonic, and hypertonic. Therefore, it is engineered to be applied for wastewater filtration and seawater desalination to ensure the availability of clean water [5].

Osmosis is known as a physical phenomenon that is defined as a natural process in which water molecules spontaneously move from a solution of low solute concentration to a solution of high solute concentration across a semi-permeable membrane [6,7]. The semi-permeable membrane rejects the solutes and only allows water molecules to pass through. The osmosis process continues until a state of osmotic equilibrium is reached where the chemical potentials across the membrane become equal [8]. Then, the flow of water molecules can be...
stopped or reversed by applying external pressure on the solution of higher concentration (feed solution) [9].

FO is one of the modified processes of osmosis due to an osmotic pressure gradient [6,8–10]. The movement process of water is drawing a water-permeable but solute-impermeable membrane from the feed solution into the more concentrated draw solution [2,9–12]. Finally, it doesn’t need more pressure with hydraulic assistance [6,13].

To date, to our knowledge, no earlier report about osmosis and its implication on the FO process for water purification and desalination. We describe the concept of tonicity in the osmosis process and its relationship with the FO process. Several modifications in the FO process, such as DS and membranes development also discussed in this overview. The main objective of this study is to help researchers and academia find the latest development strategies for water treatment and desalination.

2. A Brief History of Osmosis

Osmosis is a very old topic, as it was first observed in the 18th century by Jean-Antoine Nollet. In his report, he observed swelling and emptying of pockets driven by the presence of various dissolved components in water, more precisely in different sugars in plants and sperm in slugs. Based on his observation, he introduced a new term, namely “osmose” which means a push, then served as a pioneer due to this phenomenon. However, the mechanism of driving osmotic flow is still unclear. Thus, it needs to be developed extensively [3]. Furthermore, it was rationalized more than one century later by van’t Hoff, who showed that the osmotic pressure took the form of a perfect gas equation of state. Etymologically, osmosis was known as a push, and indeed, osmosis is usually associated with the notion of force and pressure (as shown in Figure 1). It is influenced by an osmotic pressure which is typically expressed across a semi-permeable membrane. The membrane only allows the solvent to pass while retaining solutes [3].

Since biologists discovered the structure and composition of the mammalian cell membrane, they have been cleared to understand how substances enter and exit the cell’s interior. The selectively permeable membrane of the cell perhaps allows the movement of several solutes and prevents others [4]. In this case, the concepts of osmolarity play an important role in realizing the tonicity. Tonicity is the ability of an extracellular solution to make water move into or out of a cell by osmosis. It was divided into three terms, hypotonic, isotonic, and hypertonic. The three only differed in the solute and water conditions, where hypertonic has a greater number of solutes than water, but the opposite occurs in hypotonic conditions. Thus the water can move out or into the cells. However, in isotonic conditions, the solute and water are in a balanced condition, thus it resulting in no transport in cells [14,15].

This condition made early researchers study the mechanism of osmosis through natural materials. In the 1960s, special attention was given to osmosis through synthetic materials [6]. Generally, osmosis is a physical phenomenon that explains water flows through a semi-permeable membrane from a solution with a low concentration of dissolved solutes to a high concentration of dissolved solutes [16]. So, scientists have extensively studied it in various science and engineering disciplines.
Figure 1. Osmosis process through the semi-permeable membrane.

The applications of the osmosis concept are also obviously considerable and span very diverse fields. For example, membrane science, especially in water purification and desalination, is a great field that promises to be applied in the osmosis concept, specifically in FO application. The next section discusses FO definition and the system’s main components, such as DS and membranes development.

3. Forward Osmosis

Forward osmosis (FO) is the movement of water through a semi-permeable membrane due to an osmotic pressure gradient [6,9,17]; the water is drawn across a water-permeable but solute-impermeable membrane from the feed solution into the more concentrated draw solution [2,9,11]. This phenomenon is similar to the hypotonic concept due to the water concentration in the feed side (s) is higher than in the draw side (d), thus feed solution and draw solution generate an osmotic pressure gradient and trigger some driving forces such as solute concentrations ($C_s, C_d$), water activities ($a_s, a_d$) and water chemical potentials ($\mu_s, \mu_d$) as shown at Figure 2 [10,12]. All driving forces are formed by solute-solvent interaction because the presence of solute disturbs the structure of the solvent, which is water [18].

Figure 2. (a) Schematic representation of the driving force involved in FO in an ideal system where only water is transported across the membrane (i.e., 100% solute rejection by the membrane); (b) Solute-solvent interaction effect on solute concentration, water activity and water chemical potential [21] Reprinted from Journal of Food Engineering, Vol 155, Malak Hamdan, Adel O. Sharif, Ghazi Derwish, Sami Al-Aibi, Ali Altaee, Draw solution for Forward Osmosis process: Osmotic pressure of binary and ternary aqueous solutions of magnesium chloride, sodium chloride, sucrose and maltose, Copyright (2018), with permission from Elsevier.
In pure liquid water, the molecules are heavily hydrogen-bonded in an ordered structure. The presence of ions from the solute content disturbs the structures by creating strong electric fields; the water dipoles are orderly, arranged, and strongly bound, thus affecting the freedom of water molecules and their hydrogen bond system [19,20]. In addition, the non-ionic solutes, different structures may be produced depending on the nature of the solute molecules, polar or non-polar.

In the polar case, dipole-dipole and dipole-induced dipole interactions are manifested, producing structures affecting the free movement of water molecules; for the solute molecules can hydrogen-bond to water molecules, this will greatly disturb the structure of liquid water, which influences strongly the hydrogen-bonded of water structure. Consequently, the solution will decrease its chemical potential. In comparison, the molecules of non-polar solutes are perhaps enclosed in micelle-type structures and disturb the hydrogen-bonded structure of liquid water. Moreover, a second solute that can influence the solute-solvent interaction, the osmotic pressure of the resulting solution may be greatly affected. Depending on the nature of the second solute, the solute-solute and solute-solvent interactions may lead to different osmotic behaviors [20–24]. This system purposes in resulting the right FO processes, which is \( C_d \) and \( \mu_d \) should higher than \( C_s \) and \( \mu_s \), whereas \( a_d \) must be higher than \( a_s \), respectively [18].

FO has become an innovative dewatering technology [25]. It doesn’t just need a low hydraulic pressure for water separation processing [26–29]; it aims to reduce energy consumption in electrical pumping, lower fouling propensity, and higher fouling reversibility [30–37]. These may prolong the membrane’s service life and reduce operational costs [38]. There are two crucial components that have been the center of attention in FO study: draw solution and membrane development.

**3.1. Draw solution**

The draw solute plays an important role as its task is to produce the actual driving force for water transport through the membrane in FO [39–41].
A good draw solution must-have requirements such as no damage to the FO membrane, low viscosity, low toxicity, high diffusivity, low costs, and chemically stable [10,38,39]. In the various requirements, viscosity and diffusivity are more important factors due to their direct influence on internal concentration polarization (ICP) and reducing the diffusivity following water flux [10]. Thus it will enhance the hypotonicity in the feed side. Draw solutions are categorized into responsive and non-responsive, as shown in Figure 3. All DS categories aim to form a hypertonic condition on the draw side and enhance hypotonic on the feed side.

3.1.1. Responsive DS.

Responsive DS are those in which the water affinity significantly changes in response to different stimulants such as temperature, pH, light, and electric or magnetic fields [38]. It is important to realize that, although the recovery of the draw can be achieved easier with a responsive draw, it will still require significant amounts of energy (related to the osmotic pressure of the draw). This means that responsive DS does not require less energy to be recovered, but cheap or waste energy can be used (e.g., residual heat). The various responsive DS are nanoparticles, hydrogels, metathesis precipitable salts, soluble gases or volatile liquids, NH$_3$-CO$_2$ DS, polarity-switchable DS, and thermally responsive molecules.

3.1.1.1. Nanoparticles.

Nanoparticles (NPs) that are widely used as DS are paramagnetic NPs [42]. Magnetic NPs such as Fe$_3$O$_4$ with surface decoration would generate the osmotic pressure needed to extract water through the FO membrane. Fe$_3$O$_4$ capped by a series of hydrophilic species was studied as magnetically responsive DS, and the magnetic NPs can be rapidly separated from the draw solution [43–46]. In addition, magnetic NPs are highly attractive for it easy to regenerate with magnet-assisted and very low back diffusion [38,47]. However, the FO water flux is less impressive even when the feed solution is deionized water [45,46,48] or diluted brackish water (2 g/l MgSO$_4$) [44]. Kim *et al.* [49] found that although smaller NPs are preferred in the FO process, NPs less than 11 nm were not easy to be separated even under a strong magnetic field, whereas even bigger NPs (>20 nm) were difficult to recover from the magnetic separation column. In addition, NPs with uniform dimensions are desirable for efficient regeneration via a membrane process. However, achieving this in large quantity is still a challenge. Another issue is the agglomeration of NPs under a magnetic field during regeneration, which is difficult to be completely resolved even with intense ultrasonication. One possible solution is to introduce hydrophobic association between particles to aid the regeneration process, and hence a weaker magnetic field can be used during regeneration to prevent agglomeration [38]. The hydrophobic side of NPs doesn’t bind water; thus, it makes easier regeneration process in separating water and drawing solute recovery. Therefore, the FO process with NPs DS is required to find the uniform dimension.

3.1.1.2. Hydrogels.

Hydrogels are cross-linked hydrophilic polymers that can absorb an amount of water within the network [50,51]. Hydrogels’ first direct test was successfully examined as polyelectrolyte with immersing and swelling performance for seawater desalination [52,53]. However, it is insufficient to achieve a high salt rejection and slightly water recovery under high hydraulic pressure; in more latest studies, they are effectively served as semi-solid DS in
the FO process [38]. Moreover, the water can be released in the regeneration process because hydrogels trigger a response to shrinking under various external stimuli. Unlike DS in general, which dissolves or disperses in water, the water flux in FO with hydrogels is driven by the chemical potential water activity. In contrast, the osmotic pressure is only a convenient parameter for characterizing liquid solutions. Consequently, it doesn’t fit in the concept of osmotic pressure as always [54,55].

Hydrogels as DS firstly synthesized from poly(sodium acrylate) (P-SA) [56]. P-SA generated the highest water flux and formed strong interaction with water molecules (e.g., hydration and ionization) [57]. These results are caused by reducing the water chemical potential and increasing the potential chemical gradient across the membrane but only recovering a small portion of water from hydrogels.

Another alternative, such as thermo-responsive hydrogels from poly(N-isopropylacrylamide) (P-NIPAm), was also achieved for water recovery. Nevertheless, it is poor in drawing ability while swelling, even when SA copolymerizes NIPAm to fix it. It still failed to produce a significant improvement because of electrostatic repulsion and ions hydration [58]. To enhance the deswelling of hydrogels, poly(SA-NIPAm) random copolymers were identified by three investigations as extrinsic parameters of hydrogels [59,60], incorporation of different additives [61–63], and construction of a double-layered structure [64]. However, their problems in the FO process are that most of the water was released in vapor form due to little or rapid macroscopic shrinkage in releasing the absorbed water.

Based on several studies, some major problems of hydrogels in the FO process are the imbalance of swelling-deswelling rates while heat-induced evaporation using light irradiation [38]. Various ways to resolve these obstacles are modifying the structure of the hydrogel. Cai et al. [65] prepared semi-interpenetrating networks (semi-IPNs). Semi-IPNs are a system in which one linear polymer is tied with another cross-linked polymer [66,67]. In this result, the sustainability of water flux was demonstrated for the first time even though the value was still very low.

Furthermore, other semi-IPNs hydrogels based on thermo-responsive poly-ionic liquids based P-NIPAm resulted in more water flux than quadrupled [68]. However, several studies about the reversibility performance of hydrogels are still possible to find in low water flux. Hydrogels are more suitable for low salinity water treatment unless the discovery of modified materials results in some novelty to resolve this problem.

### 3.1.1.3. Metathesis precipitable salts.

Metathesis precipitable salts is a draw solute reaction in which two compounds exchange ions, typically with precipitation of an insoluble product for the regeneration process [69]. In the early 1970s, this method was examined to precipitate Al₂(SO₄)₃ using Ca(OH)₂ [70]. It helps the regeneration process enhances the water filtration, so the DS will be precipitated by salts precipitable agent and cannot be dispersed in the clean water [69]. For example, Alnaizy et al. explained that several inorganic salts, such as CuSO₄ and MgSO₄, are judged as smart DS; and their regeneration is a metathesis reaction [69,71]. They added the Ba(OH)₂ into the diluted draw solution to generate BaSO₄ and Cu(OH)₂ or Mg(OH)₂ precipitation chemically. These DS can be used effectively for brackish water solutions; BaSO₄ is simple to separate from precipitation mixtures with CuSO₄ or MgSO₄ DS using H₂SO₄ to recover.
Furthermore, fresh water can be obtained via microfiltration. Besides, Al$_2$(SO$_4$)$_3$ as draw solute for FO was also promoted by Liu et al. [72]. They played a charged equilibrium between positive and negative charges for destabilization in the regeneration process using SiO$_2$ coated magnetic Fe$_3$O$_4$ NPs. The utilized external magnetic field as DS removal and H$_2$SO$_4$ with insoluble CaSO$_4$ as the by-product. However, the resulted water is not required to consume; the Al$^{3+}$ and Cu$^{2+}$ availability in the final product.

### 3.1.1.4. Soluble gases or volatile liquids.

Soluble gases or volatile liquids are another way that can be utilized to optimize the regeneration process in the FO system. Basically, it used liquid-gas phase separation as Batcheleter reported that several gases such as SO$_2$ and NH$_3$ are quite soluble in water, and the osmotic pressure can be achieved [73]. In the regeneration process, air-stripping or evaporation can be used to remove these gases under controlled temperature and pressure. McGinnis [74] promoted his dual-stage draw solution system using acidic or basic gases based on SO$_2$ or CO$_2$. Both gases were used to extract water from the first draw solution, but an obstacle with these gases is ionizing after dissolution and resisting the regeneration process [74]. Later, Sato et al. [75] reported a water-inert low boiling gas or high volatile liquid, dimethyl ether, as the draw solute in FO desalination. The freshwater with 24 bar osmotic pressure can be obtained after drawing solute evaporation at room temperature. Unfortunately, this one needs a large operation and matches hydraulic pressure in the draw and feed side. Therefore, the energy consumption does not occur in regeneration but the draw solution preparation. Therefore, the future requirements for FO membranes to have a better water-organic liquid selectivity in order to explore the potential of these types of DS.

#### 3.1.1.5. NH$_3$–CO$_2$ DS.

NH$_3$–CO$_2$ DS is the thermolytic system studied as DS and has shown good promise in pilot-scale operations; it was first discovered in 1964 [76]. Thermolytic salts are made from the reaction between NH$_3$ and CO$_2$, thus, water can produce a very high osmotic pressure in the FO process. In this process, NH$_3$ and CO$_2$ role as original gases formed by decomposing DS; water as freshwater is separated from the regeneration process via evaporation. DS can be used in the FO process if it forms water-soluble salts (NH$_4$)$_2$CO$_3$ and H$_2$NCOONH$_4$ as long as the ratio of NH$_3$:CO$_2$ is more than one [77]. However, they can form CaCO$_3$ because of the Ca$^{2+}$ ion content in the feed side; and it has resulted in the water back diffusion from the draw side to the feed side, damaging the membrane and disrupting the evaporation process [78]. Even worse, the regeneration process via evaporation requires greater energy consumption, and the DS obtained is more unstable than before [79]. This problem can be solved by modifying the feed side containing Ca$^{2+}$ ions pre-treatment, such as chemical softening, media filtration, activated carbon, and cartridge filtration [77]. However, all of these methods have high costs. Despite all these shortcomings, it is still one of the most studied DS and has shown very good promise for water treatment and desalination [80,81].

#### 3.1.1.6. Polarity-switchable draw solute (P-SDS).

P-SDS was introduced by Stone et al. [82] as DS, which can change their affinity with water. This type of DS is in the form of a hydrophobic tertiary amine, which can turn into
ionizable hydrophilic after reacting with CO₂. This reaction produces a high osmotic pressure, which facilitates water flow from the feed side to the draw side (Figure 4). P-SDS returns to the neutral condition due to the addition of NaOH causing CO₂ release, but its solubility to water is relatively high [82,83]. As a result, P-SDS will swell and damage the selectivity of the FO membrane; thus, it has poor compatibility [84].

\[
\text{NR}_3^+ + \text{CO}_2 + \text{H}_2\text{O} \quad \text{HNR}_3^+ + \text{HCO}_3^- 
\]

**Figure 4.** Hydrophobic tertiary amine as P-SDS form to hydrophilic P-SDS.

Thereafter, Cai et al. [85] investigated the potential of poly[2-(dimethylamino) ethyl methacrylate] (PDMAEMA) to solve the problem of the regeneration process, which resulted in poor P-SDS compatibility with FO membranes. The PDMAEMA-based P-SDS regeneration process is carried out by depositing the solute at 60 °C to separate P-SDS from CO₂. As a result, the separation process is more effective and achieves a water recovery of 95%. However, these findings are not comprehensive enough to explain concentration polarization to optimize molecular weight and architecture [38].

**3.1.1.7. Thermally responsive molecules (TRM).**

Thermally responsive molecules (TRM) are DS which have strong temperature-dependent solubility [86]. TRM in the FO process is a homogeneous single-phase solution at temperatures below the lower critical solution temperature (LCST). However, at high temperatures than LCST, the DS undergoes a phase separation. TRM can produce fresh water from the seawater in the FO process via LCST. Polypropylene-based TRM with 425 g/mol molecular weight can treat seawater in a bench-scale FO system with an osmotic pressure of up to 40 bar [87]. However, the performance in the regeneration process is poor due to the polydispersity in molecular weight, which causes DS to become dense and difficult to recover. The draw solution simultaneously results in a DS-rich and water-rich phase that can be further polished for freshwater production. Another studies [88,89], N-acrylate poly(ethylene imine) derivatives and glycol ethers as possible LCST-type DS. These DS have a very stable phase separation behavior than LCST due to the uniform molecular weight. Unfortunately, they have low osmotic pressure and can hardly draw water from NaCl feed solution as artificial seawater.

Furthermore, Cai et al. [90] introduce LCST-type ionic liquids as desalination DS with high osmotic pressure. In addition, the enthalpy of the phase separation of these responsive ionic liquid DS are substantially lower than the decomposition enthalpy of NH₄HCO₃ ammonium salts; then, which makes them more attractive. These LCST-type ionic liquids are also more favorable than a liquid-gas or gas-liquid transition for regeneration, for it is non-volatile and has excellent thermal and chemical stability [91]. Ionic liquid DS with a more careful molecular structure design is better to develop with hydrophobicity and hydrophilicity equilibrium. The role of hydrophobic molecules is to facilitate easier regeneration, whereas hydrophilic molecules are designed in warmer climates [92,93].
3.1.2. Non-responsive DS.

Non-responsive DS is the solution that has no changes in their water affinity even if some stimulants such as temperature, pH, and the electromagnetic field of exposure to light were given [38]. Recently, it has been divided into several types, such as inorganic salts, polymers, and organic molecules.

3.1.2.1. Inorganic salts DS.

In the 1970s, inorganic salts were introduced as DS for the extraction of water from seawater. Initial studies on inorganic salts based on DS have been carried out and resulted in a protocol containing a complete list of inorganic salts. Several inorganic salts were tested for their potential utilization by comparing important parameters such as water flux, draw solute reverse diffusion, draw solute loss and recharge costs [94–96].

Various inorganic salts, namely NaCl, KCl, MgCl₂, MgSO₄, Na₂SO₄, (NH)₂SO₄, Ca(NO₃)₂, and KHCO₃ [97,98]. They utilized the concentration polarization process for water extraction because it was ideal and helpful [99] in understanding the relevant issues associated with FO, such as concentration polarization and mass transport [100–105]. In terms of performance, KCl and MgSO₄ showed a higher water flux and retention than others [97]. However, their regeneration in the FO process is lacking; thus, these need mature technologies (e.g., thermal distillation, membrane distillation, or nanofiltration) to use in the regeneration process.

3.1.2.2. Polymers and organic molecules.

Polymers and organic molecules were also investigated as non-responsive DS. Mostly, polyelectrolyte is used as DS, for it has a high viscosity to produce a high-water flux and reduce reverse diffusion [106]. One type of polyelectrolyte is linear poly(sodium acrylate) (PSA) and utilized as DS for seawater desalination. It performs the reverse diffusion reduction into feed solution, and it has a higher molecular weight than inorganic salts. However, the higher viscosity of polyelectrolyte aggravated obstacles such as concentration polarization, circulation difficulties, and regeneration process in the FO system [107]. In order to solve that problem, poly(aspartic acid) modified sodium acrylate (SA) with thermally responsive as NIPAm showed a good regeneration process, but it has a poor osmotic pressure; for the absence of counter-ions which decreases the charge density in the DS [108–111]. Thus, there is another attempt to overcome the previous obstacle, namely by modifying the monomer structure and composition of the polyelectrolyte to study its molecular architecture. These efforts have been carried out ranging from linear chains to hyperbranched or dendritic chain structures. The results show that dendritic polyelectrolyte produced higher water flux in FO and improved draw solute regeneration, for it has a lower viscosity and higher radius of gyration than the linear polyelectrolyte counterpart at similar osmotic pressures [112,113].

Meanwhile, organic salts or electrolytes also have a good property of low viscosity, generating higher water flux. A variety of possible DS-based organic salts can be used in FO process, namely ethylenediamine tetrapropionic [114], hexavalent phosphazene [115], ethylenediaminetetraacetic [116], hydroacid complexes [117–119], zwitterions [120], and ionic liquids [121]. However, their regeneration is similar to polyelectrolyte to replace monovalent NaCl through nanofiltration and membrane distillation. It aims to lower reverse diffusion and a comparable water flux.
3.2. Membrane characteristics for FO.

An ideal FO membrane must give a high-water flux, a high rejection towards both feed and DS, and a reasonable mechanical strength. FO membranes consist of a selective layer and support structure [122]. The selective layer is tasked to reject particles and solutes from feed and draw sides, whereas the support structure provides mechanical stability. But, a challenging issue of the FO membrane is reverse permeation of the draw solution, as it affects membrane fouling [123], osmotic pressure loss, and water flux [124]. Therefore, the main objectives that govern FO membrane design such as: (a) Having at least a water permeability of $> 1 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$; (b) Lowering the structural parameter of the support as much as possible; (c) Increasing the selective layer’s rejection of the draw solute to maintain the osmotic pressure difference [125]. A selective layer and support structure have some general explanations that are necessary to focus.

3.2.1. Selective layer.

In fact, the selective layer in determining the membrane’s separation properties. FO membrane needs a highly rejecting selective layer to preserve this high driving force. Otherwise, the feed and DS will penetrate across the membrane and decrease the osmotic pressure gradient [126]. Consequently, these phenomena will affect the mass transfer resistance. The selective layer can be obtained by various methods, such as interfacial polymerization (IP), layer-by-layer assembly (LbL), surface blending, cross-link, and grafting, as can be seen in Figure 4.

![Figure 4. Various selective layer membranes preparation for FO.](https://biointerfaceresearch.com/)

3.2.1.1. Interfacial polymerization (IP).

IP is a process of dissolving two highly reactive monomers in two immiscible liquids [127]. The reaction takes place at the liquid-liquid interface following a thin film formation, namely a thin-film composite (TFC) [128–130]. TFC has the potential to be modified and tailored as for FO membrane [131]; for they have a thin polyamide selective layer with high
salt rejection and high-water permeability [132,133]. TFC is commonly synthesized from two monomers like m-phenylenediamine as nucleophile reactant (multifunctional amine) and trimesoyl chloride as electrophile reactants (multifunctional acid chlorides) [134]. In TFC formation, the IP process consists of three different kinetic stages such as primary film formation, slow polymerization, and diffusion-limited growth [135,136].

The characteristics that should be obtained in TFC are molecular weight, surface roughness, and layer thickness. All of these can be determined by choice of the monomer, diffusivity, concentration, reactivity, and solubility [134]. Nevertheless, TFC also needs some support surface properties such as pore size, porosity, roughness, and hydrophilicity, for these have several significant roles in producing a stable and suitable interface [137]. In addition, non-uniform structures become an obstacle to TFC; thus, charge or polymer density parameters are difficult to characterize [138,139].

The problem of non-uniform can be solved with support’s pore size. Singh et al. successfully synthesized polyamide with polysulfone as support pore’s size to reduce the polyamide penetration; thus, it resulted in a greater salt rejection efficiency with an average pore size (0.07 μm and 0.15 μm) [140]. The pore size can control the thickness of the TFC to produce low water flow; thus, the water flow and salt rejection were in unbalanced conditions [10].

The optimal IP reaction on the support surface can be achieved when the membrane's pore size is between 1 and 100 nm. However, it is experimentally hard to fulfill; conditional parameters such as concentration of monomers, humidity, reaction time between monomers, drying time, and researchers' experience or skills also affect the characteristics of the polyamide layer [141,142]. As evidence, several IPs with the same support can show a different selective layer characteristics, and IP processing is very sensitive [143]. Consequently, it can lead to delamination and lower reproducibility [144].

Furthermore, another effort to resolve the reproducibility problem is to notice the drying step between two monomers used for IP [10]. It can be fabricated easily using a rubber roller for flat sheets, followed by a drying process with an air purge. The hollow fibers will result in more frequent residual drops in this drying condition. Unfortunately, these residual drops influence a proper liquid-liquid interface following defects in the polyamide layer. In addition, the pressure differences from hydrodynamic flow along the hollow fiber membrane may also result in a different degree of penetration of the IP layer, especially in the porous support along with the fiber. These main drawbacks mean that IP-based layer development should be carried out. Several alternative solutions can be recommended to solve this problem, including utilizing cellulosic membranes [145], carbon nanotubes incorporation [146,147], and aquaporin [148–150].

3.2.1.2. Layer-by-layer assembly (LbL).

LbL is a new technique for FO membrane fabrication [151–154]. LbL is fabricated by opposite polyelectrolyte, which is alternately deposited on top of a charged support to form a thin selective film [10,155,156]. LbL also has several advantages of LbL: flexible, simple, sustainable, and tunable preparation and possible to fabricate using various types of polyelectrolyte and unsophisticated instruments [157]. In addition, this method is more sustainable with polar solvents than water. It creates an ultra-thin defect-free layer to increase membrane permeability, great thermal stability, and high solvent resistance [157,158].
Thereby, all of these advantages solved some problems related to the cost and sustainability of FO membranes fabrication.

Poly (allylamine hydrochloride) (PAH) is an ordinary polymer that forms a membrane bilayer and can be used as ultra-thin layers-based FO membranes [151]. The bilayer membrane has a higher water-flux in 28 L m\(^{-2}\) h\(^{-1}\), but it is a low rejection of very small DS like NaCl [159,160]. Then, MgCl\(_2\) is used as DS to replace NaCl because its radius is bigger than NaCl. PAH can be increased by adding glutaraldehyde as a cross-linker so that it performs better MgCl\(_2\) rejection and lowers reverse salt flux, but the water flux is low [152].

### 3.2.1.3. Other preparation methods.

The continuity of membranes preparation still experienced several modifications, such as asymmetric membranes by surface cross-linking, blending, and grafting have been studied. In the surface cross-linking method, polyamide-imide can be fabricated by phase inversion using poly(ethylene imine) as a cross-linker to form asymmetric membrane [161,162], whereas asymmetric membranes by surface blending commonly practice improving separation characteristics by adding some NPs such as TiO\(_2\) [163,164], carbon nanotubes [165], zeolite [166], and graphene oxide (GO) [167,168], or magnetic nanoparticles [169,170]. These NPs improve water flux, rejection, hydrophilicity, mechanical stability, and antifouling behavior [171,172].

Unlike cross-linking and blending methods, surface grafting chemical bonding was prepared using inorganic NPs or organic functional groups via plasma discharge, UV irradiation, and ozone [173]. In this case, surface grafting generates more integration into the polymer structure. This method proposes of enhancing the antifouling FO membrane-based polyamide selective layer with amine-terminated sulfonated poly(arylene ether sulfone) [174]. The same antifouling behavior was observed after surface-grafting of a zwitterionic monomer onto the selective layer of the FO membrane [175,176]. Furthermore, GO nanosheets also have a role in grafted polyamide selective layer and successfully resulted in a high salt rejection with a smooth surface, sufficient hydrophilicity, and low biofouling tendency [177].

### 3.3. Further application of FO for wastewater treatment & desalination.

The application of FO for wastewater treatment and desalination is an effort to resolve the scarcity of water [178]. Several elucidations about FO development aim to obtain a high purity of water production to consumption. The biggest impact in wastewater treatment and desalination is important to clarify based on specific information. Therefore, specific work can be developed as soon as possible.

#### 3.3.1. Wastewater treatment.

FO has been utilized in municipal wastewater (sewage) treatment systems [179]. FO system has shown over time in various research studies to work effectively to reject different pollutants and recover nutrients from various types of wastewater [31]. Chen et al. [180] found the anaerobic digestion feed replacement using concentrated sewage to produce a high-quality final effluent. However, problems still need to be addressed to make the FO process practical for deployment in this area by combining FO with reverse osmosis (RO) in a hybrid process. The wastewater used in the FO produces high-quality water, and it is used to dilute seawater before the RO step. In another study [181], an FO-membrane distillation hybrid process was
used to remove tetracycline from wastewater, with a rejection and water recovery are 99.9% and 15–22%, respectively. In addition, FO can be used in wastewater treatment for nutrient and energy recovery [182]. For example, biogas production and recovery of nutrients such as phosphate, ammonia, and potassium [182].

3.3.2. desalination.

In 1975, one of the first studies describing the use of FO in water desalination was published [42]. In 1975, one of the first studies describing the use of FO in water desalination was published [94]. This paper confirmed that desalination of Atlantic Ocean seawater was possible using an FO membrane with a glucose solution as the draw solution. Application of an FO–nanofiltration (FO-NF) hybrid process in brackish water desalination instead of a stand-alone RO unit resulted in less fouling in the NF and a high-water recovery (>90%) due to the addition of the FO step [128,183]. McCutcheon et al. [81] investigated the possibility of FO for seawater desalination with 0.05 to 2 M NaCl as feed solution and 1.1 to 1.4 M carbon dioxide as draw solution. They found that salt rejections achieve 95% to 99%.

In another study, a flat-sheet cellulose tri-acetate FO membrane was used in water desalination, producing a high-water flux and high salt rejection (over 95%) with NH₄HCO₃ as a draw solution [80]. FO displayed a high rejection towards NaCl and little fouling [80,81,94,183]. To obtain a reasonable water flux, the draw concentration should be higher than seawater to generate a sufficient osmotic pressure difference. In addition, the final product is not clean water but a diluted draw solution, which needs an additional step to recover both water and draw solute. FO is only viable for desalination if the concentrated draw solution can be directly used (negating the need for a secondary treatment step) or if the concentrated draw solution can be recovered with lower-quality (or residual) energy.

4. Conclusions

Osmosis, as a fundamental process of water movements in living cells, has been implicated in FO for water treatment and desalination. The tonicity phenomenon such as hypotonic and hypertonic showed a successful and failure in the FO process. At the same time, the isotonic condition is found in the FO process due to the water retention process during the regeneration of DS. Some benefits also resulted in this process from the components classification and their development, such as DS and selective membranes. KCl and MgSO₄ are still better draw solutes than others; they achieved the highest water flux and highest retention rate during regeneration, respectively. However, the better membrane still has to improve its compatibility with the DS to fix the water-flux problem. Thus, recognition of membrane characteristics and their preparation methods, such as IP, LbL, and other assembly, needs to be studied comprehensively to show great potential in wastewater treatment and desalination applications. Based on our overview, researchers and academia can utilize this study to find the latest development strategies in water treatment and desalination.

Funding

This review received no external funding.

Acknowledgment

This review was conducted without financial support.

https://biointerfaceresearch.com/
Conflicts of Interest

The authors declare no conflict of interest.

References

1. Wood-Robinson, C. Osmosis in the balance. J. Biol. Educ. 2001, 35, 98–99, https://doi.org/10.1080/00219266.2000.9655750.
2. Bajraktari, N.; Hélix-Nielsen, C.; Madsen, H.T. Pressure retarded osmosis from hypersaline sources — A review. Desalination 2017, 413, 65–85, https://doi.org/10.1016/j.desal.2017.02.017.
3. Marbach, S.; Boquet, L. Osmosis, from molecular insights to large-scale applications. Chem. Soc. Rev. 2019, 48, 3102–3144, https://doi.org/10.1039/C8CS00420J.
4. Goodhead, L.K.; MacMillan, F.M. Measuring osmosis and hemolysis of red blood cells. Adv. Physiol. Educ. 2017, 41, 298–305, https://doi.org/10.1152/advan.00083.2016.
5. Volpin, F.; Fons, E.; Chekli, L.; Kim, J.E.; Jang, A.; Shon, H.K. Hybrid forward osmosis-reverse osmosis for wastewater reuse and seawater desalination: Understanding the optimal feed solution to minimise fouling. Process Saf. Environ. Prot. 2018, 117, 523–532, https://doi.org/10.1016/j.pse.2018.05.006.
6. Cath, T.; Childress, A.; Elimelech, M. Forward osmosis: Principles, applications, and recent developments. J. Membr. Sci. 2006, 281, 70–87, https://doi.org/10.1016/j.memsci.2006.05.048.
7. Qasim, M.; Badreldaman, M.; Darwish, N.N.; Darwish, N.A.; Hilal, N. Reverse osmosis desalination: A state-of-the-art review. Desalination 2019, 459, 59–104, https://doi.org/10.1016/j.desal.2019.02.008.
8. Jain, H.; Garg, M.C. Fabrication of polymeric nanocomposite forward osmosis membranes for water desalination—A review. Environ. Technol. Innov. 2021, 23, 101561, doi:10.1016/j.eti.2021.101561.
9. Li, L.; Shi, W.; Yu, S. Research on forward osmosis membrane technology still needs improvement in water recovery and wastewater treatment. Water (Switzerland) 2020, 12, 1–27, https://doi.org/10.3390/w12010107.
10. Mohammadifakhri, M.; Grooth, J. de; Roesink, H.D.W.; Kemperman, A.J.B. Forward osmosis: A critical review. Processes 2020, 8, 1–29, https://doi.org/10.3390/PR8040404.
11. Korenak, J.; Basu, S.; Balakrishnan, M.; Hélix-Nielsen, C.; Petrinic, I. Forward osmosis in wastewater treatment processes. Acta Chim. Slov. 2017, 64, 83–94, https://doi.org/10.17344/acsi.2016.2852.
12. Patel, A.; Mungray, A.K.; Mungray, A. A novel concept of Vertical Up-Flow Forward Osmosis reactor: Design, performance and evaluation. Chemosphere 2021, 281, 130741, doi:10.1016/j.chemosphere.2021.130741.
13. Milczarek, R.R.; Sedef, I. Aroma profiling of forward-osmosis watermelon juice concentrate and comparison to fresh fruit and thermal concentrate. Lwt 2021, 151, 112147, doi:10.1016/j.lwt.2021.112147.
14. Talaat, K.M. Forward osmosis process for dialysis fluid regeneration. Artif. Organs 2009, 33, 1133–1135, https://doi.org/10.1111/j.1525-1594.2009.00816.x.
15. Artemi, A.; Chen, G.Q.; Kentish, S.E.; Lee, J. The relevance of critical flux concept in the concentration of skim milk using forward osmosis and reverse osmosis. J. Membr. Sci. 2020, 611, 118357, https://doi.org/10.1016/j.memsci.2020.118357.
16. Orłowski, J.P.; Abulleil, M.M.; Phillips, J.M. The hemodynamic and cardiovascular effects of near-drowning in hypotonic, isotonic, or hypertonic solutions. Ann. Emerg. Med. 1989, 18, 1044–1049, https://doi.org/10.1016/S0196-0644(89)80927-8.
17. Argyropoulos, C.; Rondon-Berrios, H.; Raj, D.S.; Malhotra, D.; Agaba, E.I.; Rohrscheib, M.; Khitan, Z.; Murata, G.H.; Shapiro, J.J.; Tzamaloukas, A.H. Hypertonicity: Pathophysiologic concept and experimental studies. Curesus 2016, 8, 1–11, https://doi.org/10.7759/curesus.596.
18. Kucera, J. Reverse Osmosis, Industrial Applications and Processes 2010; ISBN 9780470882634.
19. Molitor, H.R.; Schaeffer, A.K. Schnoor, J.L. Sustainably cultivating and harvesting microalgae through sedimentation and forward osmosis using wastes. ACS Omega 2021, 6, 17362–17371, https://doi.org/10.1021/acsomega.1c01474.
20. Hafiz, M.A.; Hawari, A.H.; Das, P.; Khan, S.; Altaee, A. Comparison of dual stage ultrafiltration and hybrid ultrafiltration-forward osmosis process for harvesting microalgae (Tetraselmis sp.) biomass. Chem. Eng. Process. - Process Intensif. 2020, 157, 108112, https://doi.org/10.1016/j.cep.2020.108112.
21. Hamdan, M.; Sharif, A.O.; Derwish, G.; Al-Aibi, S.; Altaee, A. Draw solutions for Forward Osmosis process: Osmotic pressure of binary and ternary aqueous solutions of magnesium chloride, sodium chloride, sucrose and maltose. J. Food Eng. 2015, 155, 10–15, https://doi.org/10.1016/j.jfoodeng.2015.01.010.
22. Finney, J.L. The water molecule and its interactions: The interaction between theory, modelling, and experiment. J. Mol. Liq. 2001, 90, 303–312, https://doi.org/10.1016/S0167-7322(01)00134-9.
23. Hribar, B.; Southall, N.T.; Vlachy, V.; Dill, K.A. How Ions Affect the Structure of Water. J. Am. Chem. Soc. 2002, 124, 12302–12311.
24. Gallo, P.; Corradini, D.; Rovere, M. Do ions affect the structure of water? the case of potassium halides. J. Mol. Liq. 2014, 189, 52–56, https://doi.org/10.1016/j.molliq.2013.05.023.
25. Yousef, M.A.; Datta, R.; Rodgers, V.G.J. Understanding nonidealities of the osmotic pressure of 
https://biointerfacereasearch.com/
concentrated bovine serum albumin. *J. Colloid Interface Sci.* 1998, 207, 273–282, https://doi.org/10.1006/jcis.1998.5789.

26. Soper, A.K.; Weckström, K. Ion solvation and water structure in potassium halide aqueous solutions. *Biophys. Chem.* 2006, 124, 180–191, https://doi.org/10.1016/j.bpc.2006.04.009.

27. Collins, K.D.; Neilson, G.W.; Enderby, J.E. Ions in water: Characterizing the forces that control chemical processes and biological structure. *Biophys. Chem.* 2007, 128, 95–104, https://doi.org/10.1016/j.bpc.2007.03.009.

28. Cao, D.Q.; Sun, X.Z.; Zhang, W.Y.; Ji, Y.T.; Yang, X.X.; Hao, X. Di News on alginate recovery by forward osmosis: Reverse solute diffusion is useful. *Chemosphere* 2021, 285, 131483, doi:10.1016/j.chemosphere.2021.131483.

29. Ray, H.; Perreault, F.; Boyer, T.H. Ammonia Recovery from Hydrolyzed Human Urine by Forward Osmosis with Acidified Draw Solution. *Environ. Sci. Technol.* 2020, 54, 11556–11565, https://doi.org/10.1021/acs.est.0c02751.

30. Arias-Paíć, M.S.; Korak, J.A. Forward Osmosis for Ion Exchange Waste Brine Management. *Environ. Sci. Technol. Lett.* 2020, 7, 111–117, https://doi.org/10.1021/acs.estlett.9b00733.

31. Francis, L.; Ogubiyi, O.; Saththasivam, J.; Lawler, J.; Liu, Z. A comprehensive review of forward osmosis and niche applications. *Environ. Sci. Water Res. Technol.* 2020, 6, 1986–2015, https://doi.org/10.1039/d0ew00181c.

32. Tavares, H.M.; Cardozo, N.S.M.; Tessaro, I.C. Concentration of grape juice: Combined forward osmosis/evaporation versus conventional evaporation. *Innov. Food Sci. Emerg. Technol.* 2021, 75, 102905, doi:10.1016/j.ifset.2021.102905.

33. Mi, B.; Elimelech, M. Organic fouling of forward osmosis membranes: Fouling reversibility and cleaning without chemical reagents. *J. Membr. Sci.* 2010, 348, 337–345, https://doi.org/10.1016/j.memsci.2009.11.021.

34. Kalafatakis, S.; Zarebska, A.; Lange, L.; Hélix-Nielsen, C.; Skiadas, I. V.; Gavala, H.N. Biofouling mitigation approaches during water recovery from fermented broth via forward osmosis. *Membranes (Basel).* 2020, 10, 1–18, https://doi.org/10.3390/membranes10110307.

35. Xie, M.; Lee, J.; Nghiem, L.D.; Elimelech, M. Role of pressure in organic fouling in forward osmosis and reverse osmosis. *J. Membr. Sci.* 2015, 493, 748–754, https://doi.org/10.1016/j.memsci.2015.07.033.

36. Saha, S.; Mondal, S. Performance of a forward osmosis mass exchanger based on detailed mass transfer boundary layer analysis. *Desalination 2020*, 496, 114708, https://doi.org/10.1016/j.desal.2020.114708.

37. Trishitman, D.; Negi, P.S.; Rastogi, N.K. Concentration of beetroot juice colorant (betalains) by forward osmosis and its comparison with thermal processing. *Lwt* 2021, 145, 111522, doi:10.1016/j.lwt.2021.111522.

38. Cai, Y.; Hu, X.M. A critical review on draw solutes development for forward osmosis. *Desalination 2016*, 391, 16–29, https://doi.org/10.1016/j.desal.2016.03.021.

39. Ge, Q.; Ling, M.; Chung, T.S. Draw solutions for forward osmosis processes: Developments, challenges, and prospects for the future. *J. Membr. Sci.* 2013, 442, 225–237, https://doi.org/10.1016/j.memsci.2013.03.046.

40. Giagnorio, M.; Casasso, A.; Tiraferri, A. Environmental sustainability of forward osmosis: The role of draw solute and its management. *Environ. Int.* 2021, 152, 106498, https://doi.org/10.1016/j.envint.2021.106498.

41. Sanahuja-Embuna, V.; Frauholz, J.; Oruc, T.; Trzaskus, K.; Hélix-Nielsen, C. Transport mechanisms behind enhanced solute rejection in forward osmosis compared to reverse osmosis mode. *J. Membr. Sci.* 2021, 636, https://doi.org/10.1016/j.memsci.2021.119561.

42. Mounir, M.; Gadallah, H.; Ali, H.M.; Souaya, E.R.; Azab, A.A. Ferric hydroacid & diamine complex as draw solute for forward osmosis (FO) desalination processes. *Chem. Eng. Technol.* 2021, 5, 100316, doi:10.1016/j.let.2021.100316.

43. Ling, M.M.; Chung, T.S. Desalination process using super hydrophilic nanoparticles via forward osmosis integrated with ultrafiltration regeneration. *Desalination 2011*, 278, 194–202, https://doi.org/10.1016/j.desal.2011.05.019.

44. Bai, H.; Liu, Z.; Sun, D.D. Highly water soluble and recovered dextran coated Fe3O4 magnetic nanoparticles for brackish water desalination. *Sep. Purif. Technol.* 2011, 81, 392–399, https://doi.org/10.1016/j.seppur.2011.08.007.

45. Ling, M.M.; Wang, K.Y.; Chung, T.S. Highly water-soluble magnetic nanoparticles as novel draw solutes in forward osmosis for water reuse. *Ind. Eng. Chem. Res.* 2010, 49, 5869–5876.

46. Dey, P.; Izake, E.L. Magnetic nanoparticles boosting the osmotic efficiency of a polymeric FO draw agent: Effect of polymer conformation. *Desalination 2015*, 373, 79–85, https://doi.org/10.1016/j.desal.2015.07.010.

47. Lubick, N. Tiny filters fix big water problems. *Environ. Sci. Technol.* 2007, 41, 4495–4496.

48. Na, Y.; Yang, S.; Lee, S. Evaluation of citrate-coated magnetic nanoparticles as draw solute for forward osmosis. *Desalination 2014*, 347, 34–42, https://doi.org/10.1016/j.desal.2014.04.032.

49. Kim, Y.C.; Han, S.; Hong, S. A feasibility study of magnetic separation of magnetic nanoparticle for forward osmosis. *Water Sci. Technol.* 2011, 64, 469–476, https://doi.org/10.2166/wst.2011.566.
50. Putro, P.A.; Sulaeman, A.S.; Erizal Synthesis and characterization of superabsorbent Hydrogel Carboxymethylcellulose-g-Poly (Acrylic Acid)/Natrium Alginate cross-linked by gamma-ray irradiation technique. J. Phys. Conf. Ser. 2019, 1171, 012011–1–012011–10, https://doi.org/10.1088/1742-6596/1171/1/012011.

51. Zhang, X.; Li, Y.; Ma, Z.; He, D.; Li, H. Modulating degradation of sodium alginate/bioglass hydrogel for improving tissue infiltration and promoting wound healing. Bioact. Mater. 2021, 6, 3692–3704, https://doi.org/10.1016/j.bioactmat.2021.03.038.

52. Höpfnner, J.; Klein, C.; Wilhelm, M. A novel approach for the desalination of seawater by means of reusable poly (acrylic acid) hydrogels and mechanical force. Macromol. Rapid Commun. 2010, 31, 1337–1342, https://doi.org/10.1002/marc.201000058.

53. Ali, W.; Gebert, B.; Hennecke, T.; Graf, K.; Ulbricht, M.; Gutmann, J.S. Design of thermally responsive polymeric hydrogels for brackish water desalination: Effect of architecture on swelling, deswelling, and salt rejection. ACS Appl. Mater. Interfaces 2015, 7, 15696–15706, https://doi.org/10.1021/acsami.5b03878.

54. Wang, H.; Wei, J.; Simon, G.P. Response to osmotic pressure versus swelling pressure: Comment on “bifunctional polymer hydrogel layers as forward osmosis draw agents for continuous production of fresh water using solar energy.” Environ. Sci. Technol. 2014, 48, 4214–4215, https://doi.org/10.1021/es5011016.

55. Zhao, S. Osmotic Pressure versus Swelling Pressure: Comment on “Bifunctional Polymer Hydrogel Layers As Forward Osmosis Draw Agents for Continuous Production of Fresh Water Using Solar Energy.” Environ. Sci. Technol. 2014, 48, 4212–4213, https://doi.org/10.1021/es5011016.

56. Li, D.; Zhang, X.; Yao, J.; Simon, G.P.; Wang, H. Stimuli-responsive polymer hydrogels as a new class of draw agent for forward osmosis desalination. Chem. Commun. 2011, 47, 1710–1712, https://doi.org/10.1039/c0cc04701e.

57. Luo, H.; Wang, Q.; Tao, T.; Zhang, T.C.; Zhou, A. Performance of strong ionic hydrogels based on 2-acrylamido-2-methylpropane sulfonate as draw agents for forward osmosis. J. Environ. Eng. 2014, 140, 04014044–1–04014044–8, https://doi.org/10.1061/(asce)ee.1943-7870.0000875.

58. Hirose, Y.; Amiya, T.; Hirokawa, Y.; Tanaka, T. Phase transition of submicron gel beads. Macromolecules 1987, 20, 1342–1344, https://doi.org/10.1021/ma00172a029.

59. Li, D.; Zhang, X.; Simon, G.P.; Wang, H. Forward osmosis desalination using polymeric hydrogels as a draw agent: Influence of draw agent, feed solution and membrane on process performance. Water Res. 2013, 47, 209–215, https://doi.org/10.1016/j.watres.2012.09.049.

60. Razmjou, A.; Simon, G.P.; Wang, H. Effect of particle size on the performance of forward osmosis desalination by stimuli-responsive polymer hydrogels as a draw agent. Chem. Eng. J. 2013, 215–216, 913–920, https://doi.org/10.1016/j.cej.2012.11.088.

61. Li, D.; Zhang, X.; Yao, J.; Zeng, Y.; Simon, G.P.; Wang, H. Composite polymer hydrogels as draw agents in forward osmosis and solar dewatering. Soft Matter 2011, 7, 10048–10056, https://doi.org/10.1039/c1sm06043k.

62. Zeng, Y.; Qiu, L.; Wang, K.; Yao, J.; Li, D.; Simon, G.P.; Wang, R.; Wang, H. Significantly enhanced water flux in forward osmosis desalination with polymer-graphene composite hydrogels as a draw agent. RSC Adv. 2013, 3, 887–894, https://doi.org/10.1039/c2ra22173j.

63. Razmjou, A.; Barati, M.R.; Simon, G.P.; Suzuki, K.; Wang, H. Fast deswelling of nanocomposite polymer hydrogels via magnetic field-induced heating for emerging FO desalination. Environ. Sci. Technol. 2013, 47, 6297–6305, https://doi.org/10.1021/es4005152.

64. Razmjou, A.; Liu, Q.; Simon, G.P.; Wang, H. Bifunctional polymer hydrogel layers as forward osmosis draw agents for continuous production of fresh water using solar energy. Environ. Sci. Technol. 2013, 47, 13160–13166, https://doi.org/10.1021/es403266y.

65. Cai, Y.; Sheng, V.; Loo, S.L.; Krantz, W.B.; Wang, R.; Fane, A.G.; Hu, X. Towards temperature driven forward osmosis desalination using Semi-IPN hydrogels as reversible draw agents. Water Res. 2013, 47, 3773–3781, https://doi.org/10.1016/j.watres.2013.04.034.

66. Dragan, E.S. Design and applications of interpenetrating polymer network hydrogels. A review. Chem. Eng. J. 2014, 243, 572–590, https://doi.org/10.1016/j.cej.2014.01.065.

67. Kim, N.U.; Park, B.J.; Park, M.S.; Park, J.T.; Kim, J.H. Semi-interpenetrating polymer network membranes based on a self-crosslinkable comb copolymer for CO2 capture. Chem. Eng. J. 2019, 360, 1468–1476, https://doi.org/10.1016/j.cej.2018.10.152.

68. Cai, Y.; Wang, R.; Krantz, W.B.; Fane, A.G.; Hu, X. “M” Exploration of using thermally responsive polyionic liquid hydrogels as draw agents in forward osmosis. RSC Adv. 2015, 5, 97143–97150, https://doi.org/10.1039/c5ra0018e.

69. Alnaizy, R.; Aidan, A.; Qasim, M. Draw solute recovery by metathesis precipitation in forward osmosis desalination. Desalin. Water Treat. 2013, 51, 5516–5525, https://doi.org/10.1080/19443994.2013.770238.

70. Frank, B.S. Desalination of Sea Water, US Patent 1972.

71. Alnaizy, R.; Aidan, A.; Qasim, M. Copper sulfate as draw solute in forward osmosis desalination. J. Environ. Chem. Eng. 2013, 1, 424–430, https://doi.org/10.1016/j.jece.2013.06.005.

72. Liu, Z.; Bai, H.; Lee, J.; Sun, D.D.A low-energy forward osmosis process to produce drinking water. Energy Environ. Sci. 2011, 4, 2582–2585, https://doi.org/10.1039/c1ee01186c.
Yong, J.S.; Phillip, W.A.; Elimelech, M. Coupled reverse draw solute permeation and water flux in forward osmosis with neutral draw solutes. *J. Memb. Sci.* **2012**, *392–393*, 9–17, https://doi.org/10.1016/j.memsci.2011.11.020.

Kim, B.; Lee, S.; Hong, S. A novel analysis of reverse draw and feed solute fluxes in forward osmosis membrane process. *Desalination* **2014**, *352*, 128–135, https://doi.org/10.1016/j.desal.2014.08.012.

Zhao, P.; Yue, Q.; Gao, B.; Kong, J.; Rong, H.; Liu, P.; Shon, H.K.; Li, Q. Influence of different ion types and membrane orientations on the forward osmosis performance. *Desalination* **2014**, *344*, 123–128, https://doi.org/10.1016/j.desal.2014.03.018.

Yasukawa, M.; Tanaka, Y.; Takahashi, T.; Shibuya, M.; Mishima, S.; Matsuyama, H. Effect of molecular weight of draw solute on water permeation in forward osmosis process. *Ind. Eng. Chem. Res.* **2015**, *54*, 8239–8246, https://doi.org/10.1021/acs.iecr.5b01960.

Ge, Q.; Su, J.; Amy, G.L.; Chung, T.S. Exploration of polyelectrolytes as draw solutes in forward osmosis processes. *Water Res.* **2012**, *46*, 1318–1326, https://doi.org/10.1016/j.watres.2011.12.043.

Tian, E.; Hu, C.; Qin, Y.; Ren, Y.; Wang, X.; Wang, X.; Xiao, P.; Yang, X. A study of poly (sodium 4-styrenesulfonate) as draw solute in forward osmosis. *Desalination* **2015**, *360*, 130–137, https://doi.org/10.1016/j.desal.2015.01.001.

Sarp, S.; Lee, S.; Park, K.; Park, M.; Kim, J.H.; Cho, J. Using macromolecules as osmotically active compounds in osmosis followed by filtration (OF) system. *Desalin. Water Treat.* **2012**, *43*, 131–137, https://doi.org/10.1080/19443994.2012.672163.

Jun, B.M.; Nguyen, T.P.N.; Ahn, S.H.; Kim, I.C.; Kwon, Y.N. The application of polyethyleneimine draw solution in a combined forward osmosis/nanofiltration system. *J. Appl. Polym. Sci.* **2015**, *132*, 42198–42198–9, https://doi.org/10.1002/app.42198.

Zhao, P.; Gao, B.; Yue, Q.; Kong, J.; Shon, H.K.; Liu, P.; Gao, Y. Explore the forward osmosis performance using hydrolyzed polyacrylamide as draw solute for dye wastewater reclamation in the long-term process. *Chem. Eng. J.* **2015**, *273*, 316–324, https://doi.org/10.1016/j.cej.2015.03.093.

Kim, J.; Chung, J.S.; Kang, H.; Yu, Y.A.; Choi, W.J.; Kim, H.J.; Lee, J.C. Thermo-responsive copolymers with ionic group as novel draw solutes for forward osmosis processes. *Macromol. Res.* **2014**, *22*, 963–970, https://doi.org/10.1007/s13233-014-2142-6.

Zhao, D.; Chen, S.; Wang, P.; Zhao, Q.; Lu, X. A dendrimer-based forward osmosis draw solute for seawater desalination. *Ind. Eng. Chem. Res.* **2014**, *53*, 16170–16175, https://doi.org/10.1021/ie5031997.

Hobson, L.J.; Feast, W.J. Poly(amideamine) hyperbranched systems: Synthesis, structure and characterization. *Polymer (Guildf).* **1999**, *40*, 1279–1297, https://doi.org/10.1016/S0032-3861(98)00268-7.

Qiao, Z.; Wang, Z.; Zhang, C.; Yuan, S.; Zhu, Y.; Wang, J. PVAm–PnP/PS composite membrane with high performance for CO2/N2 separation. *AICHE J.* **2012**, *58*, 215–228, https://doi.org/10.1002/aic.2012.

Stone, M.L.; Wilson, A.D.; Harrup, M.K.; Stewart, F.F. An initial study of hexavalent phosphazene salts as draw solutes in forward osmosis. *Desalination* **2013**, *312*, 130–136, https://doi.org/10.1016/j.desal.2012.09.030.

Hau, N.T.; Chen, S.S.; Nguyen, N.C.; Huang, K.Z.; Ngo, H.H.; Guo, W. Exploration of EDTA sodium salt as novel draw solution in forward osmosis process for dewatering of high nutrient sludge. *J. Memb. Sci.* **2014**, *455*, 305–311, https://doi.org/10.1016/j.memsci.2013.12.068.

Ge, Q.; Chung, T.S. Hydroacid complexes: A new class of draw solutes to promote forward osmosis (FO) processes. *Chem. Commun.* **2013**, *49*, 8471–8473, https://doi.org/10.1039/c3cc43951h.

Ge, Q.; Chung, T.S. Oxalic acid complexes: Promising draw solutes for forward osmosis (FO) in protein enrichment. *Chem. Commun.* **2015**, *51*, 4854–4857, https://doi.org/10.1039/c5cc00168d.

Ge, Q.; Fu, F.; Chung, T.S. Ferric and cobaltous hydroacid complexes for forward osmosis (FO) processes. *Water Res.* **2014**, *58*, 230–238, https://doi.org/10.1016/j.watres.2014.03.024.

Lutchmiah, K.; Lauber, L.; Roest, K.; Harmsen, D.J.H.; Post, J.W.; Rietveld, L.C.; van Lier, J.B.; Cornelissen, E.R. Zwitterions as alternative draw solutions in forward osmosis for application in wastewater reclamation. *J. Memb. Sci.* **2014**, *460*, 82–90, https://doi.org/10.1016/j.memsci.2014.02.032.

Yen, S.K.; Mehnas Haja N., F.; Su, M.; Wang, K.Y.; Chung, T.S. Study of draw solutes using 2-methylimidazole-based compounds in forward osmosis. *J. Memb. Sci.* **2010**, *364*, 242–252, https://doi.org/10.1016/j.memsci.2010.08.021.

Li, R.; Drakeveld, S.; De Carfort, J.L.N.; Hussain, S.; Bollmann, U.E.; Bester, K. Laboratory and pilot evaluation of aquaporin-based forward osmosis membranes for rejection of micropollutants. *Water Res.* **2021**, *194*, doi:10.1016/j.watres.2021.116924.

Kastl, A.; Bar-Zeev, E.; Spinnler, M.; Sattelmayer, T. Impact of pulsating flows on particle deposition in forward osmosis with spacers. *J. Memb. Sci.* **2021**, *635*, 119444, doi:10.1016/j.memsci.2021.119444.

Kim, Y.; Elimelech, M.; Shon, H.K.; Hong, S. Combined organic and colloidal fouling in forward osmosis: Fouling reversibility and the role of applied pressure. *J. Memb. Sci.* **2014**, *460*, 206–212, https://doi.org/10.1016/j.memsci.2014.02.038.

Shaffer, D.L.; Werber, J.R.; Jaramillo, H.; Lin, S.; Elimelech, M. Forward osmosis: Where are we now? *Desalination* **2015**, *356*, 271–284, https://doi.org/10.1016/j.desal.2014.10.031.
126. Manickam, S.S.; McCutcheon, J.R. Understanding mass transfer through asymmetric membranes during forward osmosis: A historical perspective and critical review on measuring structural parameter with semi-empirical models and characterization approaches. Desalination 2017, 421, 110–126, https://doi.org/10.1016/j.desal.2016.12.016.

127. Cadotte, J.E.; Petersen, R.J.; Larson, R.E.; Erickson, E.E. A new thin-film composite seawater reverse osmosis membrane. Desalination 1980, 32, 25–31, https://doi.org/10.1016/S0011-9164(00)86003-8.

128. Binger, Z.M.; Achilli, A. Forward osmosis and pressure retarded osmosis process modeling for integration with seawater reverse osmosis desalination. Desalination 2020, 491, 114583, https://doi.org/10.1016/j.desal.2020.114583.

129. Zhang, X.; Liu, Y. Integrated forward osmosis-adsorption process for strontium-containing water treatment: Pre-concentration and solidification. J. Hazard. Mater. 2021, 414, 125518, doi:10.1016/j.jhazmat.2021.125518.

130. Kallem, P.; Gaur, R.; Pandey, R.P.; Hasan, S.W.; Choi, H.; Banat, F. Thin film composite forward osmosis membranes based on thermally treated PAN hydrophilized PVDF electrospun nanofiber substrates for improved performance. J. Environ. Chem. Eng. 2021, 9, 106240, doi:10.1016/j.jece.2021.106240.

131. Phillip, W.A.; Schifflman, J.D.; Elimelech, M. High performance thin-film composite forward osmosis membranes. Environ. Sci. Technol. 2010, 44, 3812–3818.

132. Mohammad, A.W.; Teow, Y.H.; Ang, W.L.; Chung, Y.T.; Oatley-Radcliffe, D.L.; Hilal, N. Nanofiltration membranes review: Recent advances and future prospects. Desalination 2015, 356, 226–254, https://doi.org/10.1016/j.desal.2014.10.043.

133. Zhang, Z.; Hu, J.; Liu, S.; Hao, X.; Li, L.; Zou, G.; Hou, H.; Ji, X. Channel regulation of TFC membrane with hydrophobic carbon dots in forward osmosis. Chinese Chem. Lett. 2021, 32, 2882–2886, doi:10.1016/j.cclet.2021.03.028.

134. Raaajmakers, M.J.T.; Benes, N.E. Current trends in interfacial polymerization chemistry. Prog. Polym. Sci. 2016, 63, 86–142, https://doi.org/10.1016/j.progpolymsci.2016.06.004.

135. Alsvik, I.L.; Hägg, M.B. Pressure retarded osmosis and forward osmosis membranes: Materials and methods. Polymers (Basel). 2013, 5, 303–327, https://doi.org/10.3390/polym5010303.

136. Zhang, S.; Liu, S.; Ding, H.; Zhu, H.; Zhao, D.; Zhang, M.; Fu, J. An innovative strategy for improving the performance of forward osmosis membrane: stripe-like Turching structure constructed by introducing hydrophilic polyvinylpyrrolidone. J. Mater. Sci. 2020, 55, 16482–16496, https://doi.org/10.1007/s10853-020-05151-y.

137. Ghosh, A.K.; Hoek, E.M.V. Impacts of support membrane structure and chemistry on polyamide-polysulfone interfacial composite membranes. J. Membr. Sci. 2009, 336, 140–148, https://doi.org/10.1016/j.memsci.2009.03.024.

138. Freger, V. Nanoscale heterogeneity of polyamide membranes formed by interfacial polymerization. Langmuir 2003, 19, 4791–4797, https://doi.org/10.1021/la020920q.

139. Freger, V.; Srebnik, S. Mathematical model of charge and density distributions in interfacial polymerization of thin films. J. Appl. Polym. Sci. 2003, 88, 1162–1169, https://doi.org/10.1002/app.11716.

140. Singh, P.S.; Joshi, S.V.; Trivedi, J.J.; Devmurari, C.V.; Rao, A.P.; Ghosh, P.K. Probing the structural variations of thin film composite RO membranes obtained by coating polyamide over polysulfone membranes of different pore dimensions. J. Membr. Sci. 2006, 278, 19–25, https://doi.org/10.1016/j.memsci.2005.10.039.

141. Wang, W.; Li, G. One-step fabrication of high selective hollow fiber nanofiltration membrane module. Fibers Polym. 2010, 11, 1041–1048, https://doi.org/10.1007/s12221-010-1041-5.

142. Ahmad, A.L.; Ooi, B.S.; Choudhury, J.P. Effect of hydrophilization additive and reaction time on separation properties of polyamide nanofiltration membrane. Sep. Sci. Technol. 2004, 39, 1815–1831, https://doi.org/10.1081/SS-120030770.

143. Foglia, F.; Karan, S.; Nania, M.; Jiang, Z.; Porter, A.E.; Barker, R.; Livingston, A.G.; Cabral, J.T. Neutron reflectivity and performance of polyamide nanofilms for water desalination. Adv. Funct. Mater. 2017, 27, 18–20, https://doi.org/10.1002/adfm.201701738.

144. Zhang, Q.; Zhang, Z.; Dai, L.; Wang, H.; Li, S.; Zhang, S. Novel insights into the interplay between support and active layer in the thin film composite polyamide membranes. J. Membr. Sci. 2017, 537, 372–383, https://doi.org/10.1016/j.memsci.2017.05.033.

145. Ma, D.; Peh, S.B.; Han, G.; Chen, S.B. Thin-film nanocomposite (TFN) membranes incorporated with super-hydrophobic metal-organic framework (MOF) UiO-66: Toward enhancement of water flux and salt rejection. ACS Appl. Mater. Interfaces 2017, 9, 7523–7534, https://doi.org/10.1021/acsami.6b14223.

146. Goh, K.; Setiawan, L.; Wei, L.; Jiang, W.; Wang, R.; Chen, Y. Fabrication of novel functionalized multi-walled carbon nanotube immobilized hollow fiber membranes for enhanced performance in forward osmosis process. J. Membr. Sci. 2013, 446, 244–254, https://doi.org/10.1016/j.memsci.2013.06.022.

147. Aminia, M.; Jahanshahi, M.; Rahimpour, A. Synthesis of novel thin film nanocomposite (TFN) forward osmosis membranes using functionalized multi-walled carbon nanotubes. J. Membr. Sci. 2013, 435, 233–241, https://doi.org/10.1016/j.memsci.2013.01.041.

148. Wang, H.; Chung, T.S.; Tong, Y.W.; Jeyaseelan, K.; Armugam, A.; Chen, Z.; Hong, M.; Meier, W. Highly
permeable and selective pore-spanning biomimetic membrane embedded with aquaporin Z. Small 2012, 8, 1185–1190, https://doi.org/10.1002/smll.201102120.

149. Xia, L.; Andersen, M.F.; Helix-Nielsen, C.; McCutcheon, J.R. Novel commercial aquaporin flat-sheet membrane for forward osmosis. Ind. Eng. Chem. Res. 2017, 56, 11919–11925, https://doi.org/10.1021/acs.iecr.7b02368.

150. Xie, W.; He, F.; Wang, B.; Chung, T.S.; Jeyaseelan, K.; Armagam, A.; Tong, Y.W. An aquaporin-based vesicle-embedded polymeric membrane for low water energy filtration. J. Mater. Chem. A 2013, 1, 7592–7600, https://doi.org/10.1039/c3ta01731k.

151. Qi, S.; Liu, X.; Li, Y.; Huo, F.; Tang, C.Y. Synthesis and characterization of silica gel-polyacrylonitrile mixed matrix forward osmosis membranes based on layer-by-layer assembly. Sep. Purif. Technol. 2014, 124, 207–216, https://doi.org/10.1016/j.seppur.2014.01.029.

152. Qiu, C.; Qi, S.; Tang, C.Y. Synthesis of high flux forward osmosis membranes by chemically cross-linked layer-by-layer polyelectrolytes. J. Memb. Sci. 2011, 381, 74–80, https://doi.org/10.1016/j.memsci.2011.07.013.

153. Duong, P.H.H.; Zuo, J.; Chung, T.S. Highly cross-linked layer-by-layer polyelectrolyte FO membranes: Understanding effects of salt concentration and deposition time on FO performance. J. Memb. Sci. 2013, 427, 411–421, https://doi.org/10.1016/j.memsci.2012.10.014.

154. Liu, C.; Fang, W.; Chou, S.; Shi, L.; Fane, A.G.; Wang, R. Fabrication of layer-by-layer assembled FO hollow fiber membranes and their performances using low concentration draw solutions. Desalination 2013, 308, 147–153, https://doi.org/10.1016/j.desal.2012.07.027.

155. Mohammadifakhr, M.; de Grooth, J.; Trzaskus, K.; Roesink, H.D.W.; Kemperman, A.B. Single-step synthesis of a polyelectrolyte complex hollow-fiber membrane for forward osmosis. Sep. Purif. Technol. 2021, 264, 118430, https://doi.org/10.1016/j.seppur.2021.118430.

156. Reurink, D.M.; De Vos, W.M.; Roesink, H.D.W.; De Grooth, J. Polyelectrolyte Multilayers for Forward Osmosis, Combining the Right Multilayer and Draw Solution. Ind. Eng. Chem. Res. 2021, 60, 7331–7341, https://doi.org/10.1021/acs.iecr.1c00205.

157. Paquin, F.; Rivnay, J.; Salleo, A.; Stingelin, N.; Silva, C. Multi-phase semicrystalline microstructures drive exciton dissociation in neat plastic semiconductors. J. Mater. Chem. C 2015, 3, 10715–10722, https://doi.org/10.1039/b000000x.

158. Bertrand, P.; Jonas, A.; Laschewsky, A.; Legras, R. Ultrathin polymer coatings by complexation of polyelectrolytes at interfaces: Suitable materials, structure and properties. Macromol. Rapid Commun. 2000, 21, 319–348, https://doi.org/10.1002/(SICI)1521-3927(20000401)21:7&lt;319::AID-MARC319&gt;3.0.CO;2-J.

159. Deng, H.Y.; Xu, Y.Y.; Zhu, B.K.; Wei, X.Z.; Liu, F.; Cui, Z.Y. Polyelectrolyte membranes prepared by dynamic self-assembly of poly (4-styrenesulfonic acid-co-maleic acid) sodium salt (PSSMA) for nanofiltration (I). J. Memb. Sci. 2008, 323, 125–133, https://doi.org/10.1016/j.memsci.2008.06.028.

160. Ouyang, L.; Malaisamy, R.; Bruening, M.L. Multilayer polyelectrolyte films as nanofiltration membranes for separating monovalent and divalent cations. J. Memb. Sci. 2008, 310, 76–84, https://doi.org/10.1016/j.memsci.2007.10.031.

161. Qiu, C.; Setiawan, L.; Wang, R.; Tang, C.Y.; Fane, A.G. High performance flat sheet forward osmosis membrane with an NF-like selective layer on a woven fabric embedded substrate. Desalination 2012, 287, 266–270, https://doi.org/10.1016/j.desal.2011.06.047.

162. Geng, X.; Li, S.; Zhan, X.; Li, J.; Lei, J.; Wang, L. Fabrication of carbon nanotubes-modified poly(ethyleneimine)/sodium lignosulfonate membranes for improved selectivity performance and antifouling capability in forward osmosis process. J. Mater. Sci. 2021, 56, 15499–15511, https://doi.org/10.1007/s10853-021-06261-x.

163. Emadzadeh, D.; Lau, W.J.; Matsuura, T.; Ismail, A.F.; Rahbari-Sisakht, M. Synthesis and characterization of thin film nanocomposite forward osmosis membrane with hydrophilic nanocomposite support to reduce internal concentration polarization. J. Memb. Sci. 2014, 449, 74–85, https://doi.org/10.1016/j.memsci.2013.08.014.

164. Emadzadeh, D.; Lau, W.J.; Matsuura, T.; Rahbari-Sisakht, M.; Ismail, A.F. A novel thin film composite forward osmosis membrane prepared from PSf-TiO2 nanocomposite substrate for water desalination. Chem. Eng. J. 2014, 237, 70–80, https://doi.org/10.1016/j.cej.2013.09.081.

165. Wang, Y.; Ou, R.; Ge, Q.; Wang, H.; Xu, T. Preparation of polyethersulfone/carbon nanotube substrate for high-performance forward osmosis membrane. Desalination 2013, 330, 70–78, https://doi.org/10.1016/j.desal.2013.09.028.

166. Ma, N.; Wei, J.; Qi, S.; Zhao, Y.; Gao, Y.; Tang, C.Y. Nanocomposite substrates for controlling internal concentration polarization in forward osmosis membranes. J. Memb. Sci. 2013, 441, 54–62, https://doi.org/10.1016/j.memsci.2013.04.004.

167. Wang, Y.; Ou, R.; Wang, H.; Xu, T. Graphene oxide modified graphitic carbon nitride as a modifier for thin film composite forward osmosis membrane. J. Memb. Sci. 2015, 475, 281–289, https://doi.org/10.1016/j.memsci.2014.10.028.

168. Romaniak, G.; Dybowsk, K.; Jeziora, A.; Kula, P.; Kazmierczak, T. Synthesis and characterization of semi-permeable graphene/graphene oxide membranes for water desalination. J. Mater. Sci. 2020, 55, 9775–
9786, https://doi.org/10.1007/s10853-020-04648-w.

169. Shakeri, A.; Salehi, H.; Khankeshipour, N.; Nakhjiri, M.T.; Ghorbani, F. Magnetic nanoparticle-crosslinked ferrohydrogel as a novel class of forward osmosis draw agent. *J. Nanoparticle Res.* 2018, 20, https://doi.org/10.1007/s11051-018-4437-6.

170. Liu, M.J.; Li, P.; Meng, Q.W.; Ge, Q. Membranes constructed by metal–ligand complexation for efficient phosphorus removal and fouling resistance in forward osmosis. *Adv. Compos. Hybrid Mater.* 2021, https://doi.org/10.1007/s42114-021-00254-9.

171. Sun, W.; Shi, J.; Chen, C.; Li, N.; Xu, Z.; Li, J.; Lv, H.; Qian, X.; Zhao, L. A review on organic-inorganic hybrid nanocomposite membranes: A versatile tool to overcome the barriers of forward osmosis. *RSC Adv.* 2018, 8, 10040–10056, https://doi.org/10.1039/c7ra12835e.

172. Kim, J.; Van Der Bruggen, B. The use of nanoparticles in polymeric and ceramic membrane structures: Review of manufacturing procedures and performance improvement for water treatment. *Environ. Pollut.* 2010, 158, 2335–2349, https://doi.org/10.1016/j.envpol.2010.03.024.

173. Kato, K.; Uchida, E.; Kang, E.T.; Uyama, Y.; Ikada, Y. Polymer surface with graft chains. *Prog. Polym. Sci.* 2003, 28, 209–259, https://doi.org/10.1016/S0079-6700(02)00032-1.

174. Zhang, X.; Tian, J.; Gao, S.; Zhang, Z.; Cui, F.; Tang, C.Y. In situ surface modification of thin film composite forward osmosis membranes with sulfonated poly(arylene ether sulfone) for anti fouling in emulsified oil/water separation. *J. Memb. Sci.* 2017, 527, 26–34, https://doi.org/10.1016/j.memsci.2017.01.002.

175. Wang, J.; Xiao, T.; Bao, R.; Li, T.; Wang, Y.; Li, D.; Li, X.; He, T. Zwitterionic surface modification of forward osmosis membranes using N-aminoethyl piperazine propane sulfonate for grey water treatment. *Process Saf. Environ. Prot.* 2018, 116, 632–639, https://doi.org/10.1016/j.psep.2018.03.029.

176. Pejman, M.; Dadashi Firouzjaei, M.; Aghapour Aktij, S.; Das, P.; Zolghadr, E.; Jafarian, H.; Arabi Shamsabadi, A.; Elliott, M.; Sadrzadeh, M.; Sangermano, M.; et al. In Situ Ag-MOF Growth on Pre-Grafted Zwitterions Imparts Outstanding Antifouling Properties to Forward Osmosis Membranes. *ACS Appl. Mater. Interfaces* 2020, 12, 36287–36300, https://doi.org/10.1021/acsami.0c12141.

177. Hegab, H.M.; ElMekawy, A.; Barclay, T.G.; Michelmore, A.; Zou, L.; Saint, C.P.; Ginic-Markovic, M. Fine-tuning the surface of forward osmosis membranes via grafting graphene oxide: Performance patterns and bio fouling propensity. *ACS Appl. Mater. Interfaces* 2015, 7, 18004–18016, https://doi.org/10.1021/acsami.5b04818.

178. Mendoza, E.; Buttiglieri, G.; Blandin, G.; Comas, J. Exploring the limitations of forward osmosis for direct hydroponic fertigation: Impact of ion transfer and fertilizer composition on effective dilution. *J. Environ. Manage.* 2022, 305, 114339, https://doi.org/10.1016/j.jenvman.2021.114339.

179. Hey, T.; Bajraktari, N.; Davidson, À.; Vogel, J.; Madsen, H.T.; Hélix-Nielsen, C.; Jansen, J. la C.; Jönsson, K. Evaluation of direct membrane filtration and direct forward osmosis as concepts for compact and energy-positive municipal wastewater treatment. *Environ. Technol.* 2018, 39, 264–276, https://doi.org/10.1080/09593330.2017.1298677.

180. Chen, L.; Gu, Y.; Cao, C.; Zhang, J.; Ng, J.W.; Tang, C. Performance of a submerged anaerobic membrane bioreactor with forward osmosis membrane for low-strength wastewater treatment. *Water Res.* 2014, 50, 114–123, https://doi.org/10.1016/j.watres.2013.12.009.

181. Pan, S.F.; Dong, Y.; Zheng, Y.M.; Zhong, L. Bin; Yuan, Z.H. Self-sustained hydrophilic nanofiber thin film composite forward osmosis membranes: Preparation, characterization and application for simulated antibiotic wastewater treatment. *J. Memb. Sci.* 2017, 523, 205–215, https://doi.org/10.1016/j.memsci.2016.09.045.

182. Ansari, A.J.; Hai, F.I.; Price, W.E.; Drewes, J.E.; Nghiem, L.D. Forward osmosis as a platform for resource recovery from municipal wastewater - A critical assessment of the literature. *J. Memb. Sci.* 2017, 529, 195–206, https://doi.org/10.1016/j.memsci.2017.01.054.

183. Zhao, S.; Zou, L.; Mulcahy, D. Brackish water desalination by a hybrid forward osmosis-nanofiltration system using divalent draw solute. *Desalination* 2012, 284, 175–181, https://doi.org/10.1016/j.desal.2011.08.053.