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

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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

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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 semipermeable 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 (μ_s , μ_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 μ_d should higher than C_s and μ_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].



Figure 3. The category of draw solutes and their types.

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 -CO₂ 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 Fe₃O₄ with surface decoration would generate the osmotic pressure needed to extract water through the FO membrane. Fe₃O₄ 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 MgSO₄) [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(Nisopropylacrylamide) (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 $Al_2(SO_4)_3$ 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 $CuSO_4$ and $MgSO_4$, 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 $BaSO_4$ and $Cu(OH)_2$ or $Mg(OH)_2$ precipitation chemically. These DS can be used effectively for brackish water solutions; $BaSO_4$ is simple to separate from precipitation mixtures with $CuSO_4$ or $MgSO_4$ DS using H_2SO_4 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 SiO₂ coated magnetic Fe₃O₄ NPs. The utilized external magnetic field as DS removal and H₂SO₄ with insoluble CaSO₄ as the by-product. However, the resulted water is not required to consume; the Al³⁺ and Cu²⁺ 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 SO₂ and NH₃ 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 SO₂ or CO₂. 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.NH₃-CO₂ DS.

 NH_3 -CO₂ 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 $(NH_4)_2CO_3$ and $H_2NCOONH_4$ as long as the ratio of NH_3 :CO₂ is more than one [77]. However, they can form CaCO₃ because of the Ca²⁺ 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 Ca²⁺ 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].

$NR_3 + CO_2 + H_2O$	$ \longleftrightarrow $	$HNR_3^+ + HCO_3^-$
Hydrophobic		Hydrophilic
P-SDS		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 temperaturedependent 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 benchscale 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 $> 1 L m^{-2} h^{-1}$ 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.

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 μ m and 0.15 μ m) [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, MgCl2 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 MgCl2 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 TiO₂ [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 standalone 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.

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Conflicts of Interest

The authors declare no conflict of interest.

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