

A Mini-Review on Thin-Film Composite Hollow Fiber Membranes for Forward Osmosis Applications

Wafaa Kh. Al-Musawy¹, Mustafa H. Al-Furaiji¹, Mustafa Mohammed Aljumaily^{2*}, Khalid T. Rashid³, Hussein G. Salih³, Qusay F. Alsahy^{3*}, Hasan Sh Majdi⁴, Alberto Figoli⁵

¹ Environment and Water Directorate, Ministry of Science and Technology, Baghdad, Iraq

² Department of Civil Engineering, Al-Maarif University College, Ramadi, Iraq

³ Membrane Technology Research Unit, Chemical Engineering Department, University of Technology – Iraq, Alsinaa Str. 52, Baghdad 10066, Iraq

⁴ Department of Chemical Engineering and Petroleum Industries, Al-Mustaqbal University College, Babylon 51001, Iraq

⁵ Institute on Membrane Technology, National Research Council, 87030 Rende, Italy

* Corresponding author's e-mail: qusay_alsahy@yahoo.com

ABSTRACT

Forward osmosis (FO) is an emerging technology that has been extensively studied in the last decade as an efficient method for desalination and water treatment. FO presents many benefits over traditional desalination technologies such as reverse osmosis and distillation. Nevertheless, there are many decisive challenges; the great significance one is the new modification and advances in the preparation of the TFC membranes that must be achieved to enhance the FO performance. Therefore, preparing a suitable TFC membrane with a low structural parameter, low tortuosity, and high porosity are preferred in preparing the TFC membranes to get higher water flux and lower salt flux. This paper reviewed the recent development and advances in using TFC hollow fiber membranes in FO applications. Within that, the most widely applied monomers to prepare the thin polyamide layer (PA) in TFC membranes and the additives that are added during the preparation of the PA layer and their effect on the performance of the TFC membranes have been discussed. Moreover, an effort is made to generate a TFC membrane properties and performance trend according to the results of the water permeate flux and reverse salt flux of the modified TFC FO membranes and the future perspectives and concluding remarks on the FO membrane are evaluated.

Keywords: forward osmosis; hollow fiber; thin-film composite; membranes; structural parameter; polyamide.

INTRODUCTION

Nowadays, supplying suitable freshwater has become very important for human life and different purposes due to the rapid population growth (Alayan et al. 2020). One of the leading technologies that have been used in the treatment of water is membrane technology owing to its competent removal of pollutants from contaminated water (Al-Furaiji et al. 2021; Alayan et al. 2021). Reverse osmosis (RO) is an example of membrane technologies which has been used for treating saline water (Kalash et al. 2020). RO uses

operating pressure or hydraulic pressure to exceed and oppose the osmotic pressure of the saline water to produce purified water (Kadhom et al. 2019). The hydraulic pressure is the driving force for the mass transfer through a semi-permeable membrane in the reverse osmosis process (Martinetti et al. 2009). Compared to reverse osmosis, the forward osmosis (FO) process has the benefits of operating at low or no pressure as it uses osmotic pressure as the driving force for the process (Cath et al. 2006). Forward osmosis uses the difference in the osmotic pressure between a low-concentrated feed solution and a high-concentrated draw

solution resulting in pure water to transfer through the membrane from feed solution to draw solution. The other advantages of the FO process over the other filtration process types are the high rejection for salts and other contaminants, low fouling, ease of building and the very simple equipment used (McCutcheon et al. 2006). The forward osmosis process has been investigated in many applications in water treatment fields. One of the best researches of seawater desalination by the FO process has been successfully demonstrated using ammonia-carbon dioxide as a draw solution (McCutcheon et al., 2005). The draw solution was economically recovered and regenerated in the process; high water flux and high salt rejection were achieved. Also, FO has been recognized as a cost-competitive and a potential alternative for wastewater treatment technologies (Lutchmiah et al. 2014). Forward osmosis is not the ultimate process in wastewater treatment but relatively a high-level pretreatment step before an ultimate desalination process. The treatment of the high salinity oil field-produced water can be a big challenge due to its complexity and its high osmotic pressure makes it challenging to handle it using the conventional membrane processes. FO has the ability to treat these complex wastewater streams using high osmotic pressure draw solutions like $MgCl_2$ (Al-Furaiji et al. 2018). It can be integrated with the membrane distillation process to extract clean water from produced water with high salinity even more than eight times that of seawater (Shaffer et al. 2013; Al-Furaiji et al. 2018). Also, it can be used to provide potable water in a situation where water is scarce or not available using hydration bags. In the hydration bags, a sugar draw solution is used in a sealed bag made of a semi-permeable FO membrane where the diluted draw solution can be consumed as a sweet drink containing minerals and nutrients. This application is beneficial in recreational, military and emergency relief situations (Cath et al. 2006). The preparation of a suitable forward osmosis membrane is a crucial issue for the development of forward osmosis operations. These suitable membranes have to provide high water flux, low reverse salt flux, controlled concentration polarization and low fouling (Alihemati et al. 2020).

Nanofiltration (NF) membranes prepared from polybenzimidazole were tested in forward osmosis process. The NF membranes can be a good candidate for FO applications, especially when using divalent ions draw solutions such as $MgCl_2$ and $CaCl_2$ (Wang et al. 2007). Another attempt was made by

Jaffer et al. 2020 by preparing polyphenylsulfone hollow fiber Nanofiltration membranes at different polymer concentrations. These membranes were examined in forward osmosis process and showed high performance in water desalination (Jaafer et al. 2020). Latterly, polyvinyl chloride hollow fiber membranes were prepared and their performance was tested in nanofiltration and forward osmosis process. The prepared PVC hollow fiber membranes showed promising results in the desalination of saline water (Behboudi et al. 2021). The most recent technique that has been used is the preparation of a suitable thin-film composite (TFC) layer coating a nonselective porous substrate membrane (Tian et al. 2013). TFC layer provides high water permeability and good solute rejection values. It is formed by the interfacial polymerization (IP) reaction of two selected monomers on the substrate surface of the membranes (Raaijmakers and Benes 2016). The active layer and the support layer in TFC membranes can be synthesized separately by using different materials as well as different methods, which can enhance the TFC membrane performance. There are different factors that can affect in preparation of the support layer including; the selection of a suitable polymer material with different concentrations, a suitable solvent (for preparing the dope solution), the medium of precipitation and using different additives (Alihemati et al. 2020). On the other hand, the parameters that affect preparing the TFC layer by the IP reaction includes the monomers type, the monomers concentration, solvent type, the IP reaction time and the additives used in the solution (aqueous solution or organic solution) (Kadhom et al. 2021). Generally, the TFC layer can be synthesized on the various configurations including flat sheet, spiral-wound, hollow fiber, and tubular (Lim et al. 2019). This paper presents a general review on the recent development and advances of thin-film composite hollow fiber membranes for forward osmosis operations. It discussed the various membrane configurations that can be used in the FO process. Then it reviewed the most recent papers on using TFC membranes in FO experiments, focusing on hollow fiber configuration.

BASIC STRUCTURE AND CHEMISTRY OF THE FO MEMBRANE

Concentration polarization (CP) is a phenomenon that can occur in any membrane

separation procedure, regardless of whether it is pressure- or osmotically-driven (Lee et al. 2020). CP is caused by the selective transport of species via a semi-permeable membrane, which results in a concentration difference at the membrane-solution interface (She et al. 2016). CP develops in osmotically driven membrane processes like FO and PRO as a result of the concentration differential between draw and feed solutions passing through an asymmetric FO membrane (McCutcheon and Elimelech, 2008). The two types of CP that develop throughout the FO process are known as internal concentration polarization (ICP), which takes place inside the membrane support layer, and external concentration polarization (ECP), which can be found at the surface of the membrane active layer (Gray et al. 2006). Water flux and recovery in FO are mostly regulated by transmembrane osmotic pressure (McCutcheon, McGinnis, and Elimelech 2005). According to considerable study on CP, it was discovered that the presence of CP on both sides of the FO membrane significantly lowers the effective transmembrane osmotic pressure, making it one of the main causes of diminishing water flux and recovery across semi-permeable membranes (Akther et al. 2015).

Active layer

The selective transport barrier known as the active layer serves as this layer's primary determinant of FO performance, including water flux and reverse solute flux (Thompson et al. 2011). In other words, it isolates the water molecules while rejecting other solutes and contaminants from the feed solution... The active layer is often made to be more exclusively selective towards water. Due to the difference in concentration of the solution at the membrane surface from that of the bulk solution, external concentration polarization (ECP) occurs at the surface of the membrane's active layer (Linares et al. 2014). In contrast to pressure-driven membrane processes, which can only produce concentrative ECP, osmotic pressure-driven membrane processes can produce both concentrative and dilutive ECP depending on the orientation of the membrane (Pendergast et al. 2011). Counter-current is suggested by many studies of modeling outcomes of FO desalination by the coupling of hydrodynamics and mass transfer equations (Li et al. 2016). Water molecules diffuse across the membrane, causing

the water flow (Wang et al. 2021). The speed of this transport is significantly influenced by the strength of the driving force and the selectivity of the FO membrane for water molecules (Heo et al. 2016). The osmotic potential gradient and the inherent water selectivity of the FO membrane work together to define the water flux since the driving force for water separation is calculated by the osmotic pressure differential. Consequently, the following equation can be used to represent the water flux (J_w):

$$J_w = A \pi \Delta \quad (1)$$

where: $\Delta\pi$ – the difference in osmotic pressure between the feed and draw solutions, A – the permeability of pure water.

The pure water permeability can be used to calculate the intrinsic selectivity of water molecules, as shown in Eq (A) (Kim et al. 2017). As a result, a rise in A causes the flux of pure water to increase correspondingly (Geise et al. 2011). Similar to this, the difference in solute concentration between the two separated solutions drives the diffusion of the solute through a FO membrane (Aljumaily et al. 2022). Solute selectivity of a FO membrane can therefore be determined by the proportional constant of the solute transport equation (Eq. 2):

$$J_s = B \cdot \Delta C \quad (2)$$

where: C – the solute concentration difference between the feed and draw solutions, J_s – the solute flux, B – the solute permeability, and B is the solute flux (Linares et al. 2014).

Also studied has been conducted to reveal the impact of cross flow velocity through the FO membrane on flux (Eddouibi et al. 2021). When equal circumstances of feed and draw solutions were used, $MgCl_2$ obtained a little higher water flow than $NaCl$ (Wang et al. 2016). Although ECP caused a small flux increase when cross flow velocity rose from 0.25 to 1.0 m/s, the greatest fluxes for both draw solutions were achieved at 1.0 m/s (Li et al. 2017). Reverse diffusion happens when a solute goes from a draw solution to a feed solution, whereas forward diffusion happens when a solute move from a feed solution to a draw solution. In order to reject undesirable salts and contaminants from the feed solution as well as prevent the solute from the draw solution from leaking into the feed, B should be kept to a minimum.

Support layer

In order to give the active layer of a FO membrane mechanical strength, the support layer is often thicker, more porous, and more tortuous (Cath et al. 2013). This leads to an undesirable behavior called diffusion hindrance of draw solute through the membrane support layer, which lowers the performance of the membrane. As a result, it is crucial to quantify this transport phenomenon in order to assess the effectiveness of the FO membrane. The structural parameter (S) can serve as a generic yardstick for defining the properties of the support layer. The S is inversely proportional to porosity and is determined by the product of support layer thickness t_s and tortuosity:

$$S = \frac{t_s \tau}{\varepsilon} \quad (3)$$

According to Eq. 3, a thinner, more porous, and less tortuous support layer can attain a higher osmotic pressure of the draw solution at the interface between the active and support layers, leading to a higher water flux, because it has a lower S value. Despite having a reduced susceptibility for fouling than other membrane processes, FO has significant difficulties maintaining a high enough transmembrane flux, which has prevented its commercialization (She et al. 2016; Aljumaily et al. 2020). The membrane separation industry was completely transformed by the discovery of phase inverted asymmetric membranes, which consist of a dense active layer constructed on top of a porous support layer made of polyethersulfone (PES) or polysulfone (PSf). Nevertheless, the initial tests revealed that ICP might reduce the water flux by more than 80% (Cath et al. 2013). Several experiments have recently been conducted to further our understanding of ICP and the different ways its impacts might be reduced to improve FO performance. Because (1) the terms in Eq. 3 are difficult to directly measure and (2) it is difficult to correctly predict diffusion behavior within the support layer, the structural parameter (S) of a FO membrane is often calculated experimentally. However, some research has made an effort to determine the S using measurement techniques. For instance, X-ray microscopy (XRM) was used to determine the characteristics of the support layer in TFC membranes. Unfortunately, when compared to traditional test-based methodologies, this strategy was unable to obtain appropriate estimations of S since it was unable to take into account different physicochemical factors. Given

that certain physicochemical characteristics of the support layer substrate that are not taken into account in Eq. (3), such as hydrophobicity, can influence the diffusion of water and solute molecules, it appears that experimental methods are currently more appropriate for the measurement of precise S values.

MEMBRANE MODULES

The membranes have to be arranged into membrane modules for practical consideration. The membrane modules in use today are small units that can be used to pack or hold membranes for different membrane processes. During these processes, the feed solution enters the module at a specific flow rate and specific content. In contrast, the draw solution enters the other side of the membrane carrying the permeated water with it. The main properties of the best membrane modules are:

1. Its high packing density;
2. A reasonable control of CP and fouling;
3. Low cost of operation;
4. Cost-efficient production.

The membranes module can be classified into two types: flat sheet membranes (A plate and frame module and spiral-wound module) and tubular membranes (Hollow fiber module and tubular module). The hollow fiber (HF) modules contain a large number of thin fibers compacted together in the module; the free-ends of these thin fibers are stuck with epoxy resins, silicon rubber or polyurethanes (Alsahy 2012). The performance of the membrane inside the module is similar to the performance of the individual fibers before module preparation, which demands a module that reduces the hydraulic pressure loss, minimizes the CP and maximizes the area of the membrane. The fiber wall is either asymmetric or thin-film composite depending on the process in which the hollow fiber membranes are intended to use. The hollow fiber module contains two components: the shell side and the lumen side. The shell side represents the space between the membrane housing and the outer surface of the fibers, while the lumen side represents the space inside the fibers (Alsahy et al. 2011). Relative to other membrane types, hollow fibers offer the advantage of so large packing density that might reach up to 30,000 m²/m³, resulting in a high effective

surface area. Hollow fiber configuration is usually applied when the feed solution is almost clean, such as in seawater desalination and gas separation (Lee et al. 2020). HF configuration can be successfully used in forward osmosis application as it can be manufactured in such a way that the feed solution and draw solution flow on both membrane's sides (McCutcheon and Elimelech 2008). The benefits and drawbacks of membrane modules are listed in Table 1.

THIN FILM COMPOSITE HF MEMBRANE FOR FORWARD OSMOSIS PROCESS

Typically, the TFC membrane in the form of a film consists of two layers of materials: the first layer is called the support layer, while the second layer is the active layer (selective layer). The membrane support layer mainly gives the necessary mechanical strength of the membrane, whereas the selective layer provides the selectivity of the membrane by rejecting the unwanted impurities and allowing only water to pass. The active layer is typically prepared by a process called interfacial

polymerization coated over the top or the bottom of the support layer. Interfacial polymerization was known firstly in 1965 by Mogan. In the IP process, an ultra-thin layer is prepared via interfacial tangled between two reactant monomers (one is in the aqueous phase, while the other is in the organic phase) on the top of a porous polymeric support layer, resulting in a good selectivity to dissolved ions (Lau et al. 2012). Almost all TFC membranes prepared for desalination applications are specially generated by the IP reaction of m-phenylene diamine (MPD) in the aqueous phase with trimsoyl chloride (TMC) in the organic phase (Raaijmakers and Benes 2016).

A high water permeability, better selectivity, low structural parameter, low tortuosity, and high porosity are preferred in preparing the TFC membranes to get higher water flux and lower salt flux (McCutcheon and Elimelech 2008). The active layer prepared by IP reaction can be either generated on a flat-sheet or HF support layer. In HF membranes, the selective layer can be prepared either on the shell side of the lumen side of membranes. Table 2 shows the benefits and drawbacks of each of the configurations in HF membranes.

Table 1. Benefits and drawbacks of various membrane modules

Membrane module	Benefits	Drawbacks
Plate and frame	<ul style="list-style-type: none"> • Modest membrane surface/ volume ratio; • Well-built equipment. 	<ul style="list-style-type: none"> • Vulnerable to plugging at flow stagnation points; • Potentially hard to clean; • Costly.
Spiral-wound	<ul style="list-style-type: none"> • Compacted; • Best membrane surface/volume ratio; • Less expensive than hollow fiber and tubular elements modulas. 	<ul style="list-style-type: none"> • Vulnerable to plugging by particles; • Severely fouled. membranes are hard to clean; • Require a chemical cleaning.
Tubular	<ul style="list-style-type: none"> • Easy to clean mechanically or chemically if membranes fouled; • Able to treat large suspended solid feed with less pre-treatment; • Best hydrodynamic control; • Specific tubes can be replaced easily. 	<ul style="list-style-type: none"> • High volume required per unit area of the membrane; • Reasonably expensive; • High pressure (1500 psig).
Hollow fiber	<ul style="list-style-type: none"> • Compacted; • Outstanding membrane surface/ volume ratio; • Cost-effective. 	<ul style="list-style-type: none"> • Vulnerable to plugging by particles; • Fouled membrane. modules are difficult to clean; • Require a chemical cleaning.

Table 2. Comparison of shell selective and lumen selective HF membrane

Feature	Shell selective	Lumen selective
Advantages	<ol style="list-style-type: none"> 1. The surface area is more effective. 2. Fouling- clogging tendency is low. 3. Feed pressure drop is lower. 	<ol style="list-style-type: none"> 1. The conduct IP in a bundle is easy. 2. Burst pressure- tolerance is higher of the capillary fiber.
Disadvantages	<ol style="list-style-type: none"> 1. The conduct IP in a bundle is difficult. 2. When conducting IP in a bundle, the fiber intersection induces defects. 3. When conducting IP in a continuous operation, the bundle contact induces defects. 	<ol style="list-style-type: none"> 1. The surface area is less effective. 2. When treating exciting water, the fouling- clogging tendency is higher.

Commonly, these developed (TFC HF) membranes have great attention nowadays because of their advantages, which include:

1. They operate at low pressure.
2. Have great chemical stability.
3. Are not biodegradable.
4. They have a higher water flux.
5. Have higher rejection of unwanted materials, such as salts, silica, and organics.
6. Moreover, the operating temperature range of these membranes is 0 to 40 °C and a PH range of 2 to 12.

INTERFACIAL POLYMERIZATION REACTION TO PREPARE TFC MEMBRANES

Interfacial polymerization is a polycondensation reaction in which a reaction takes place rapidly between two monomers (aromatic diamine and acid chloride), eventually forming a linear polymer chain or a dense polymer layer (termed as polyamide). In membrane science, polyamide is a thin film layer of a thickness of 100-200 nm. Primarily, the polyamide layer is prepared on the top of a highly porous supporting membrane. The two monomers usually have a high reactivity that allows for film formation in even minutes or seconds. Therefore, the nature of the monomers determines the nature of the formed layer. However, the interface (membrane surface) plays a significant role in controlling the localized polymerization reaction. Generally, one of the reactant monomers is prepared in the aqueous phase,

while the other is prepared in the organic phase. Different types of reactive polymers are widely used as monomers that are:

1. Aliphatic-aromatic diamine, for example, p-phenylenediamine (PPD), piperazine (PIP), and m-phenylenediamine (MPD).
2. Acidic chloride monomers, for example isophthaloyl chloride (IPC), 5-isocyanatoisophthaloyl chloride (ICIC) and trimesoyl chloride (TMC) (Tsai et al. 2017).

MPD and TMC are the most widely applied monomers to prepare the thin polyamide layer in membrane science. The hexane phase is the most commonly used solvent in the organic phase (Raaijmakers and Benes, 2016). The properties of the formed polyamide layer are affected by the monomers' concentration, the monomers' reactivity, the contact time of the monomer solution and the number of reactive groups on each monomer (Tsai et al. 2017). At the high organic monomer concentrations, the monomer diffusion of the aqueous phase will increase. The high reactivity of monomers and the low solubility in the other phase are essential to obtain a dense film and the solubility of one of the monomers in the other phase could influence the film's morphology. The interfacial polymerization reaction is sensitive to humidity, purity of nascent reactants and temperature. The interfacial polymerization reaction is presented in Figure 1. In an attempt to improve the performance of the TFC membrane, some additives can be added to the reactant monomers during the IP reaction to modify

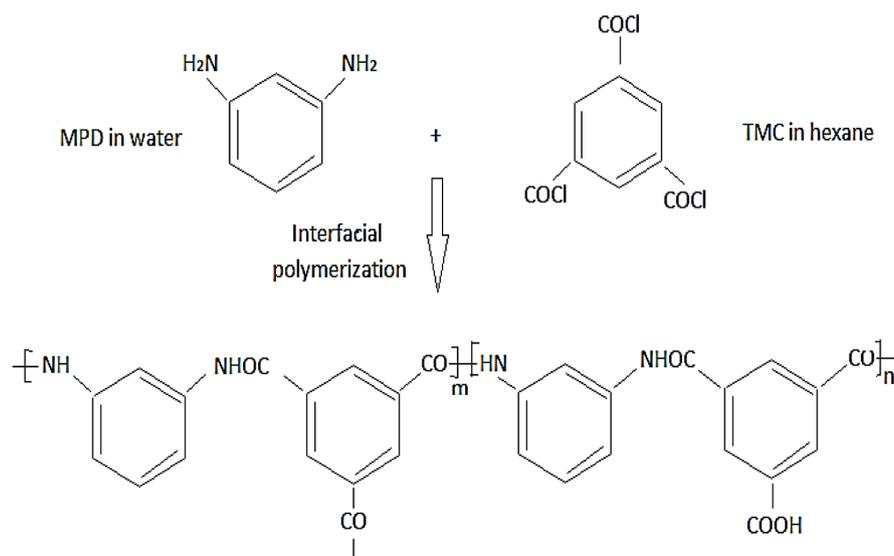


Figure 1. The IP reaction between TMC and MPD

the selective layer. These additives are added during the preparation of the aqueous solution or the organic solution. The apparent effect of the additives is to precise the mechanism of the IP reaction, improve the IP performance, change the surface roughness, hydrophobicity, surface charge, cross-linking density and antifouling properties of the TFC layer produced (Gohil, Ray, and Technology 2017). Akther et al., used graphene oxide (GO) as an additive to enhance the antifouling properties, increase the smoothness, enhance the selectivity and hydrophilicity of the membrane surface of commercial cross-linked polyvinyl alcohol (PVA) hydrogel FO membranes (Akther et al. 2020). Jang et al, added GO to modify the active layer. It reacts with MPD where it contains $-COOH$ and $-OH$ functional groups and it acts as an amide linking ring for IP reaction. High water flux, low salt flux were obtained and polyamide thickness was increased (Janga et al. 2020). HCl acid is a byproduct of the interfacial polymerization reaction. Therefore, the acceptors of acid have been used as additives with concentrations varied from 0.01 to 5%, such as sodium hydroxide (NaOH), N, N-dimethyl piperazine, trisodium phosphate, and mostly triethylamine (TEA). The polycondensation rate can be influenced by controlling the state of diamine or monomers dissociation with the addition of triethylamine. TEA acid acts as a surface-active agent which advances the wettability of the surface of the aqueous

phase monomer as well as helps the monomers' composition in regularly joining to the surface of the support membrane. In fact, the thickness of the membrane increased with adding triethylamine, but the radius of membrane pores is not affected (Gohil, Ray, and Technology 2017). Jia et al., studied the effects of phase transfer catalysts (PTCs) (sodium dodecyl sulfate (SDS), dodecyl trimethyl ammonium chloride (DTAC) and dodecyl dimethyl betaine (BS12)) on PA molecular aggregation (cross-linking), the performance of FO separation and the surface morphology of the membranes were systematically examined. PTCs can affect the polyamide morphology and the state of the molecular chain aggregation, which can enhance water flux (Jia et al. 2021). The polysulfone support showed a dual-layer symmetric structure in which the internal and external diameters were $900\ \mu\text{m}$ and $1350\ \mu\text{m}$, respectively. Also, it can clearly be seen that (Figure 2) the thickness of the polyamide layer increased from $152\ \text{nm}$ to $193\ \text{nm}$ with increasing the percent of SDS.

RECENT ADVANCES ON TFC HOLLOW FIBER MEMBRANES FOR FO

Wang, (2010) prepared a thin-film polyamide layer on a commercial polyethersulfone (PES) substrate HF membrane as shown in Figure 3. Two configurations were tested, where the thin

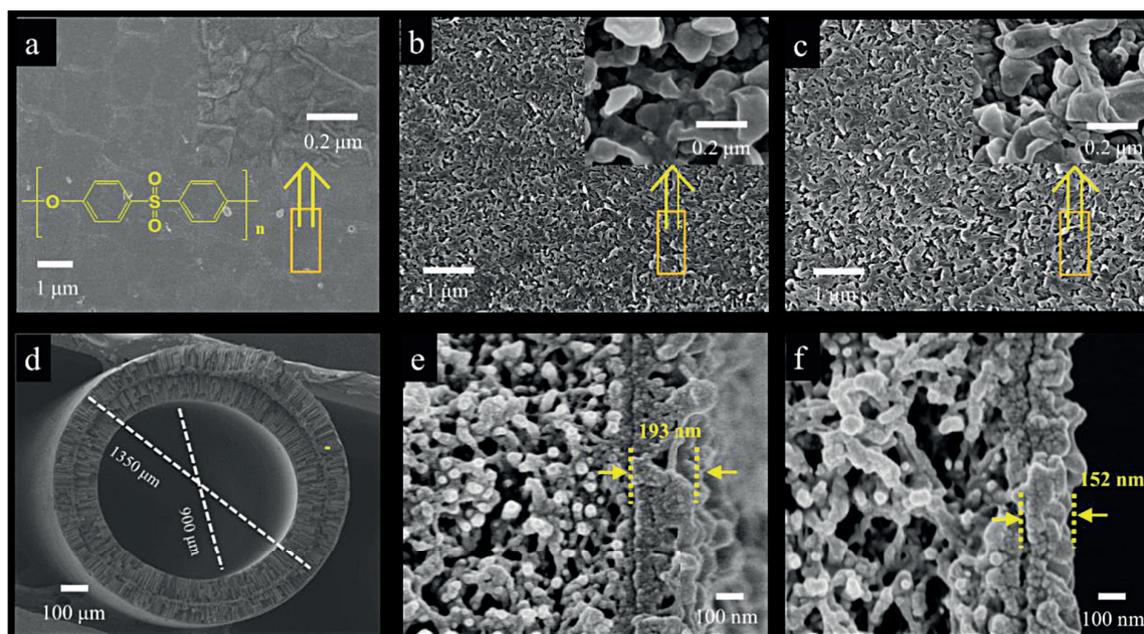


Figure 2. SEM images of surface (a) (b) (c) and cross-section (d) (e) (f) of the FO membranes, (a, d): Support layer; (b, e): polyamide layer with 0.05% SDS; (c, f): polyamide layer with 1% SDS (Jia et al. 2021)

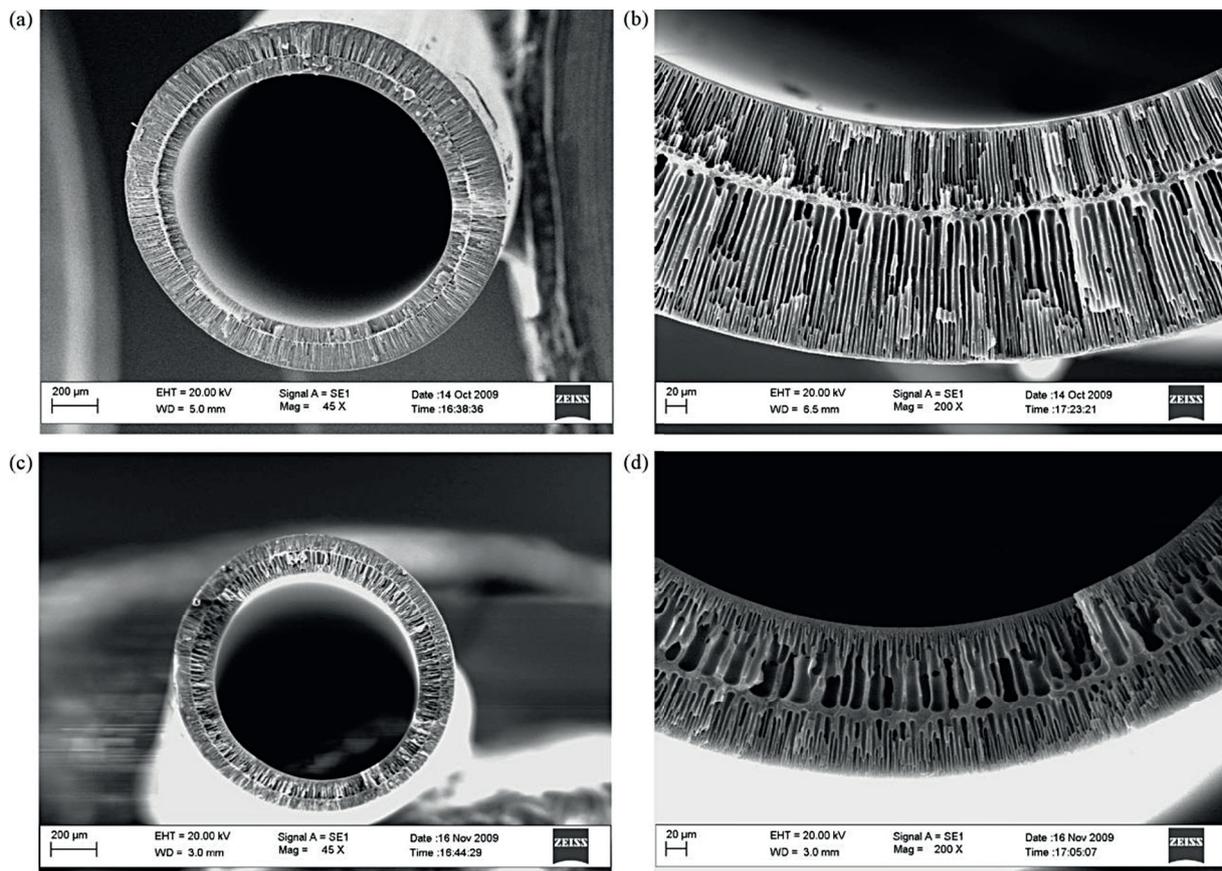


Figure 3. Morphology of PES FO hollow fiber substrates, (a) A cross-section at 45 \times , (b) A enlarged at 200 \times , (c) B cross-section at 45 \times , and (d) B enlarged at 200 \times (Jia et al. 2021)

layer was formed on both the outer surface (A-FO) and the inner surface (B-FO) of the HF membrane. The prepared TFC membranes were highly porous and narrow pore size distribution, where the selective layers have good separation specifications, good mechanical strength and a hydrophilic membrane. A-FO HF membrane achieved 12.9 LMH water flux and 5.03 GMH salt flux. However, for the B-FO hollow fiber membrane, the water flux obtained was 32.2 LMH, while the salt flux was 3.7 GMH (Jia et al. 2021).

Shi (2011) investigated the effects of the substrate structure of five PES HF membranes on the synthesis of the active layer for FO application. TFC-PES membranes were synthesized in two steps: a phase inversion (PI) to prepare a UF-like support membrane and an interfacial polarization to prepare RO-like ultra-thin selective layer (Gray et al. 2006). The results displayed that the substrate configuration is so significant for configuration RO like thin-film to obtain a good semi-permeable skin layer. This study suggested that the molecular weight of the substrate (less than 300 KDa) is important to achieve a good semi-permeable

skin layer (Shi et al. 2011). Fam et al. (2013) improved the forward osmosis performance, where two polyamide (PA) thin-film composite forward osmosis membranes have been examined and compared with the commercial cellulose triacetate (CTA) forward osmosis membranes. The experiments showed that TFC membranes showed interesting results in terms of selectivity and permeability (Fam et al. 2013).

Han (2016) developed a double skin polyamide layer structure in forward osmosis (for water reuse) and pressure retarded osmosis (for power generation) membrane. The double selective layers were configured by double IP on a polyethersulfone (PES) fiber substrate (named as *d*TFC-PES) on the shell side and lumen side. A typical ridge and valley polyamide structure was noticed (Figure 4) on both internal and external surfaces of the PES hollow fiber support with an estimated thickness of about 375 nm and 470 nm, respectively. The reason behind testing this configuration was to decrease the negative impact of ICP as well as the effect of fouling. The actual wastewater brine contains several organic foulants,

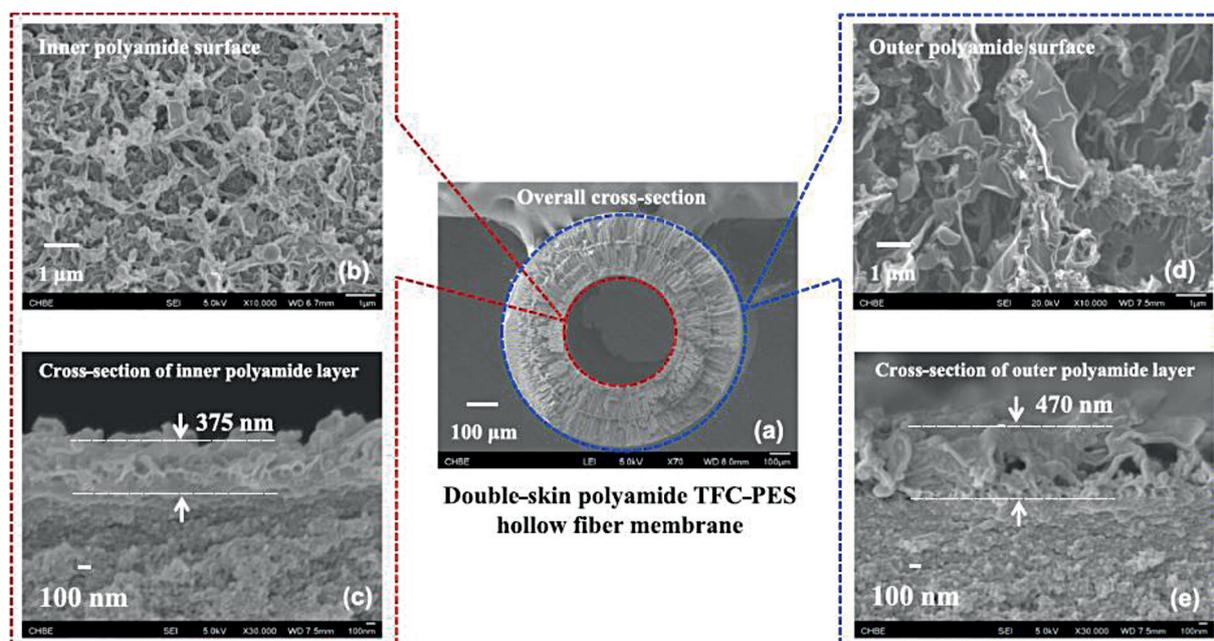


Figure 4. SEM images of the dTFC-PES hollow fiber membrane: (a) overall cross-section of the membrane, (b) surface of the inner polyamide selective skin, (c) cross-section of the inner polyamide skin, (d) surface of the outer polyamide antifouling layer, and (e) cross-section of the outer polyamide antifouling layer

and inorganic salts were used as a feed solution. Due to the fact that both sides of the membrane are polyamide layers, which are preventing the foulants and the inorganic salts from penetrating into the substrate, making fouling only happens on the membrane surface. Under the pressure retarded osmosis (PRO mode), the water flux of the membrane marginally decreased to about 70% of its initial value at a significant feed recovery of around 80%. However, flushing by either a commercial Genesol-704 or DI water can repair the water flux back to the initial value of 87% or 98% recovery (Han et al. 2017).

Shibuya, (2017) synthesized a hollow fiber FO membrane using polyketone as a substrate. The effect of hollow fiber diameter on the FO process performance was studied using two different inner diameters: 347 μm and 609 μm . The active layer (TFC) was prepared on the shell side of the polyketone HF membrane. Results showed that the hollow fiber membrane having a small diameter exhibited a higher water flux and better mechanical properties than those having a larger diameter. On the other hand, the pressure drop of the bore side is higher, that can results in a high energy consumption by the pump (Shibuya et al. 2017). Ren (2017) prepared and tested thin film composite hollow fiber membranes for the forward osmosis process using two different commercial substrate membranes from Koch

Membrane System with different inner and outer diameters HFM-A (467 and 906 μm) and HFM-B (1023 and 1818 μm). The PA layer was formed on the lumen side of these membranes via IP reaction. Results demonstrated that the thin film composite HF FO membranes with practical efficiency could be formed at scale with relative ease using the available commercial ultrafiltration modules. These membranes exhibited a good FO performance in term of water flux as well as salt rejection in comparison with commercial FO membranes (Ren et al. 2017).

Lim (2019) fabricated a thin and porous PES substrate at different air gap distances as support for a hollow fiber FO membrane. Then, a polyamide thin layer was coated on the outer surface of the HF support membrane using vacuum-assisted IP reaction. Results revealed that the molecular weight cut off of the substrate surface should be less than 88 kDa to achieve a defect free outer polyamide layer with a smooth surface roughness. The FO performance testing of the prepared membranes showed that the high water flux of 30.2 LMH and a specific revers salt flux of (0.13 g/L) using DI water as feed solution and 1 M NaCl as draw solution could be achieved. Also, these membranes provided better efficiency in terms of fouling resistance, cleaning efficiency, and the potential to scale up (Lim et al. 2019).

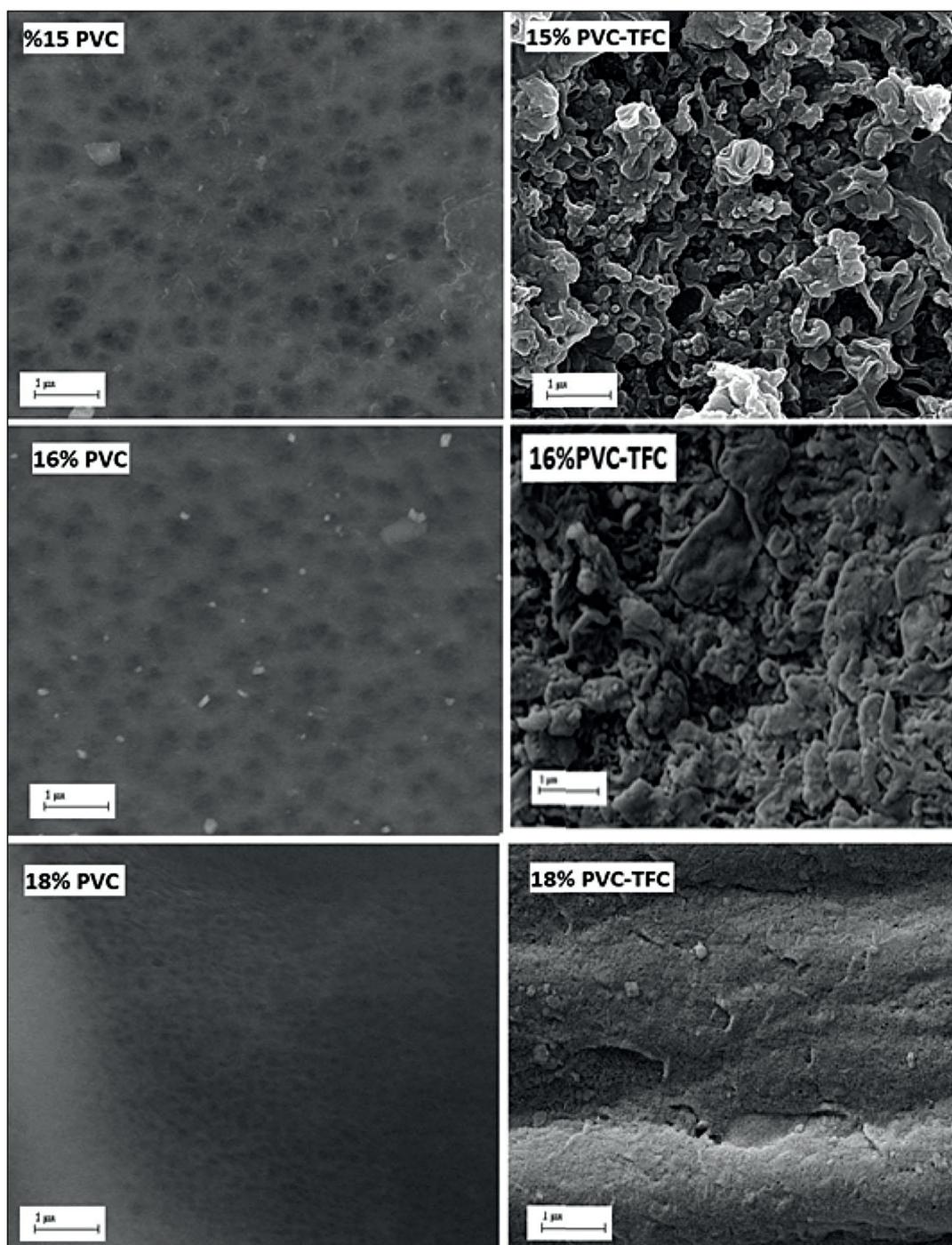


Figure 5. SEM surface images of the PVC hollow fiber membranes before and after interfacial polymerization (IP) reaction (Sugawara, Amamiya, and Yamaguchi 2022)

Liu, (2019) modified an active polyamide layer when a novel TFC FO hollow fiber membrane was prepared via IP reaction. The effect of adding different materials to the aqueous phase and to the organic phase in the IP reaction was studied. Dimethyl sulfoxide (DMSO), phase transfer catalysts (PTCs), acetone, lithium chloride (LiCl) were added as additives in the organic phase or aqueous phase system. It was found that

changing the reactants' compositions in the IP reaction could improve the FO efficacy of the TFC HF membranes. PTCs can improve the IP reaction and increase the surface area by transferring the monomer in the water phase to move into the interface region. The addition of LiCl could increase the FO membrane's water flux; however, it can also rise the reverse salt flux. DMSO was considered as the best additive as it resulted in

enhancing the water flux without changing the J_s/J_w ratio, which is the amount of salt lost in gram per liter of water permeated (Liu and Yu 2019). Saeedi-Jurkuyeh and Jonidi-Jafari, (2019) used four different concentrations of polysulfone (15%, 16%, 17%, and 18%) for preparing and characterizing the TFC FO membranes to remove two organic micro pollutants (benzene and phenol) from aqueous solution. They detected that the water flux and salt flux reduced with rising

the concentration of polysulfone (PSU) polymer and the composite 16% PSU TFC and 17% PSU TFC had higher efficiencies. Also, more amounts of organic micropollutants could be removed when the draw solution concentration increased. The results presented that TFC-FO membranes were capable of removing micro-pollutants from their aqueous solutions(Saeedi-Jurkuyeh and Jonidi Jafari 2019). Al-Musawy (2021), synthesized polyamide thin layer on the outer surface of

Table 3. Summary of the TFC membranes’ properties in the literature

Membrane type	Support layer material	Polymer concentration	Active layer	Membrane porosity (%)	Membrane thickness (µm)	Structural parameter (mm) (FO mode)	Ref.
PES HF #A-FO PES HF #B-FO PES HF #C-FO PES HF #E-FO	PES	Commercial	Polyamide (MPD+TMC)	84 75 82 69	190 180 205 180	0.63 0.58 0.63 0.58	Shi et al, 2011
TFC Polyketone HF-A TFC Polyketone HF-B	Polyketone	Commercial	Polyamide (MPD, CSA, SDS, TEA, and HMPA+ TMC)	73.6 78.0	66.5 142	250 521	Shibuya et al, 2017
TFC PPSU TFC sPPSU TFC sPPSU	PPSU 1.5% sPPSU 2.5% sPPSU	Commercial	Polyamide (MPD+TMC)		96 180 198	0.746 0.163 0.24	Zhong et al, 2013
PS HFM-A PS HFM-B	PSUPS	Commercial	Polyamide (MPD+TMC)		220 400	539 651	Ren, 2017
PVC HF	PVC	Commercial	Polyamide (MPD+TMC)			210	Ren, 2018
Chiel		Commercial	Polyamide (MPD+TMC)			N/A	Majeed et al, 2014
TFC-PES HF(0%CaCl ₂) TFC-PES HF(0%CaCl ₂) TFC-PES HF(0%CaCl ₂) TFC-PES HF(0%CaCl ₂)	PES		Polyamide (MPD+TMC)		371 257 329	422+-15 (PRO) 430+-11 457+-18	Wan et al, 2018
PAN TFC-TIP PAN TFC-HIP	PAN	12wt%PAN	Polyamide (MPD+TMC)	82.1	400 148	387 376	Kwon et al, 2018
TFC-FO HF TFC-FO/ PTC0.02 HF TFC-FO/LiClO.2 HF TFC-FO/ DMSO0.8 HF TFC-FO/ Acetone0.2 HF	Identical kind of HF UF membrane		Polyamide (MPD+TMC)				Liu and Yu, 2019
15wt% PS TFC1 16wt% PS TFC2 17wt% PS TFC3 18wt% PS TFC4	PS	15wt% PS 16wt% PS 17wt% PS 18wt% PS	Polyamide (MPD+TMC)				Jurkuyeh and Jafari, 2018
PES-HF-TFC	PES		Polyamide (MPD+TMC)				Ng et al,2019
PA-PVA-PVDF PA-PVDF PA-PVA-PSf PA-PSf		34%PVDF 20%PSf	Polyamide (MPD, TEA, CSA, SDS+ TMC)	63 64 78 76		1403 1252 989 n/d	Yabuno et al, 2019
RGO/CNT HF	PVB+CNT		Reduced graphene oxide			202	Fan et al. 2020
TFC PES HF (SM-1) TFC PES HF (SM-2) TFC PES HF (SM-3)	PES	20%PES			0.225 0.225 0.195	295.72± 15.1 1456.59±102.4 884.10 ±31.8	Lim et al, 2020

Table 4. Summary of the TFC membranes’ performance in the literature

Membrane type	Feed solution	Draw solution	Mode of operation	FS flowrate	DS flowrate	Water Flux J_w (LMH)	Salt Flux J_s (GMH)	J_s/J_w (G/L)	Ref.
PES HF #A-FO PES HF #B-FO PES HF #C-FO PES HF #E-FO	DI water	0.5M NaCl	PRO mode FO mode PRO mode FO mode PRO mode FO mode PRO mode FO mode	1.5 L/min 0.45 L/min 1.5 L/min 0.45 L/min 1.5 L/min 0.45 L/min 1.5 L/min 0.45 L/min	0.45 L/min 1.5 L/min 0.45 L/min 1.5 L/min 0.45 L/min 1.5 L/min 0.45 L/min 1.5 L/min	47.7 18.6 49.4 16.7 43.6 18.7 49.0 17.6	3.5 2.0 3.9 1.2 2.8 1.6 4.0 1.2	Shi et al, 2011	
18wt% TFC	DI water	2M NaCl				7.5	0.37	0.05	Han et al, 2012
TFC polyketone HF-A TFC polyketone HF-B	DI water	1M NaCl	PRO mode	0.15 L/min	-	50 33	11.78 5.8		Shibuya et al, 2017
TFC PPSU TFC 1.5 mol% sPPSU TFC 2.5 mol% sPPSU	DI water	0.5 M NaCl	PRO mode FO mode PRO mode FO mode PRO mode FO mode	0.1 L/min	0.2 L/min	22.64 12.37 49.39 22.51 37.71 17.98	7.73 2.69 11.0 5.49 6.98 2.63	0.34 0.22 0.22 0.24 0.19 0.15	Zhong et al, 2013
Polysulfone PS HFM-A Polysulfone PS HFM-B	DI water	1M NaCl	PRO mode FO mode PRO mode FO mode			17.49 9.08 16.54 6.18	5.52 11.38 2.13 0.75		Ren, 2017
Aquaporin	DI water	1M NaCl	PRO mode FO mode	-	-	21.0 13.2	3.6 1.7	0.18 0.14	Ren, 2018
Toyobo HF-A Toyobo HF-B Toyobo HF-C	DI water	1M NaCl	PRO mode FO mode PRO mode FO mode PRO mode FO mode	0.03 L/min 0.04 L/min 0.04 L/min	0.6 L/min 0.6 L/min 0.6 L/min	8 4 9 5 15 8	N/A 0.7 N/A 0.35 N/A 0.59	N/A 0.18 N/A 0.07 N/A 0.07	Shibuya, 2015
Chiel	DI water	1M NaCl	PRO mode FO mode			19.0 10.0	88.8 3.60	0.47 0.36	Majeed, 2014
TFC-PES HF	DI water	1.2M NaCl	PRO mode	0.2 l/min	0.2 l/min				Wan et al, 2018
PAN TFC-TIP PAN TFC-HIP	DI water	1M NaCl	PRO mode FO mode PRO mode FO mode			44.5 34.2 17.0 12.9		0.19 0.17	Kwon et al, 2018

PVC hollow fiber membrane as shown in Figure 5. The support layer was prepared utilizing the dry/wet-spinning process from dissolving 15, 16, and 18% of PVC in DMAc solvent. The polyamide layer was synthesized by IP reaction between MPD and TMC. The prepared PVC-TFC fibers exhibited excellent performance in terms of water flux (25.3 LMH) and salt flux (7.5 GMH) for the 15% PVC-TFC membrane. Increasing the polymer concentration of the support layer led to a decrease in the FO performance (Sugawara et al. 2022).

CONCLUSIONS

Only a few studies have been made on modifying the HF membranes for forward osmosis application. These studies are steadily expanded to adjust the HF membranes and make them efficient for FO operations. The efforts include the physicochemical characterization of the active layer of HF membranes which can benefit on modifying the support layer and enhance the hydrophilicity. Thus, there is a need to expand the researches by using numerous methods to improve the support layer of these membranes. Using hydrophilic polymers in preparing the HF membrane is a big challenge to investigate their behavior as support for the PA layer and then their performance in the FO operation. Moreover, studying different IP reaction conditions such as contact time of the monomers and the concentration of the monomers especially for the hollow fiber module is of great importance to produce high-efficiency FO membranes. Also, Nanofiltration membranes can be a good candidate for FO membranes. Therefore, there is a big need to investigate the development of nanofiltration membranes to make them more suitable for forward osmosis applications.

The development of a suitable draw solution compound has gained great attention in water desalination researches. The biological and functional characteristics of draw solution are rarely described, so depending on the membrane characteristics (water flux and salt flux), the draw solution must have low viscosity, low molecular weight, high osmotic pressure, high chemical stability, high thermal stability, low re-concentration energy requirement, low toxicity, complete ion dissociation and compatible with the forward osmosis membrane. Therefore, studying different draw solutions with TFC hollow fiber

FO membranes would give more insight on understanding the mechanism of water and solute transfer across the TFC membranes in the hollow fiber configuration.

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