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Review

A Mini Review on Antiwetting Studies in Membrane Distillation for Textile Wastewater Treatment

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Abstract: The textile industry is an important contributor to the growth of the global economy. However, a huge quantity of wastewater is generated as a by-product during textile manufacturing, which hinders the ongoing development of textile industry in terms of environmental sustainability. Membrane distillation (MD), which is driven by thermal-induced vapor pressure difference, is being considered as an emerging economically viable technology to treat the textile wastewater for water reuse. So far, massive efforts have been put into new membrane material developments and modifications of the membrane surface. However, membrane wetting, direct feed solution transport through membrane pores leading to the failure of separation, remains as one of the main challenges for the success and potential commercialization of this separation process as textile wastewater contains membrane wetting inducing surfactants. Herein, this review presents current progress on the MD process for textile wastewater treatment with particular focuses on the fundamentals of membrane wetting, types of membranes applied as well as the fabrication or modification of membranes for anti-wetting properties. This article aims at providing insights in membrane design to enhance the MD separation performance towards commercial application of textile wastewater treatment.

Keywords: membrane distillation; membrane antifouling; textile wastewater; polymeric membranes

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1. Introduction

The textile industry is one of the largest contributors to the global economy. However, it also faces a trade-off between economic output and environmental sustainability as a huge amount of effluent wastewater can be generated during the textile manufacturing [1]. The composition of textile effluent is complex with variations depending on the processing techniques and raw materials, but is mainly composed of a wide variety of dyes, multiple ions, and various surfactants [2]. Current solutions to mitigate the environmental impact of textile wastewater include adsorption [3], coagulation/flocculation [4], chemical/biological oxidation where the majority of biological oxygen demand, total suspended solids, color, and organic molecules can be removed [5]. However, these applied techniques have limitations such as low process efficiency, high maintenance expense, and utilization of harmful chemicals. For example, due to the influences of various pollutants, pH, temperature, and organic concentrations in the textile effluent, the biological processes may even exacerbate the toxicity, posing increased risks to the environment [6]. For adsorption processes, it is difficult to regenerate spent adsorbents and the disposal of the adsorbents can result in environmental pollution [7]. Compared with those conventional techniques, textile wastewater treatment using membranes has been attracting lots of attention owing to no additional chemicals, high separation efficiency, ease of maintenance, small footprint, and adjustable modular design [8].

Membrane processes including microfiltration (MF), ultrafiltration, nanofiltration, and reverse osmosis (RO) have been gaining popularity in the remediation of textile wastewater due to their high separation efficiency, ease of maintenance, small footprint, and adjustable modular design [9–13]. Among them, membrane distillation (MD), a membrane separation process driven by the vapor pressure difference between the hot feed side and the distillate side, has been investigated as an effective yet simplified technology for treating textile wastewater. The textile industry generally discharges hot wastewater in the temperature range of 50 to 80 °C [14]. That means the thermally driven MD process can take advantage of the high temperature of textile effluents. Compared with pressure driven membrane technologies, the feed solutions for MD are processed under atmospheric pressure and alternative energy sources such as waste heat commonly available in textile industry can be utilized [15]. Theoretically, MD offers 100% retention for non-volatile dissolved substances, and has no limitation on feed concentration, which overcomes the main drawback of pressure-driven processes of inability to treat high salt concentration solutions and usually lesser quality of produced water. MD process relies on hydrophobic membranes as a barrier to allow vapor transport while rejecting liquids, and it thereby requires no additional pressure of the feed for mass transfer. [16]. In addition, being a thermally driven process, MD is relatively less susceptible to fouling compared with the pressure driven membrane processes. MD has been proven to produce high-quality water and to concentrate various types of dyes solutions. However, most studies used synthetic dye solutions with only few studies using real textile wastewater. Membrane wetting and fouling are considered as the most critical challenges preventing the commercial application of MD in treating textile wastewater and wetting phenomenon has been receiving increasing attention for practical implementation of MD processes. When wetting occurs, the membrane loses separation performance due to the feed solution passing through the membrane pores in liquid form. Particularly, MD membranes are prone to wetting caused by surfactants commonly present in textile wastewaters [17]. For example, Dow et al. [18] conducted a pilot trial and assessed direct contact MD (DCMD) towards achieving zero liquid discharge using real textile wastewater. Initial lab testing found rapid membrane wetting appeared due to presence of surface active materials present in textile wastewater such as surfactant and biomolecules. Pretreatment with a foam fractionation process was found to be effective for minimizing membrane wetting over the long trial period. Similar findings were also reported by Zhang et al. [19]. The standalone DCMD process using commercial polytetrafluoroethylene (PTFE) membrane has difficulty in directly treating textile wastewater due to wetting of the membrane. Fractionation or ozonation was an essential pretreatment step to avoid membrane wetting of using DCMD for the treatment of real textile wastewater. Therefore, a critical requirement of stable MD performance is the ability to resist pore wetting when the membrane is in contact with the liquid to be treated.

Until now, there are many comprehensive reviews on MD with topics such as membrane polymers and surface modifications, membrane fabrication technologies, configuration design, fouling control, and temperature polarization [20–24]. Most of these reviews focus on MD processes and new material developments, and it is now appropriate to consider the practicality of MD in a specific application. Herein, the aim of this article is to present a concise review on the state-of-the-art MD membranes for the treatment of textile wastewater with particular focus on membrane modifications for enhancing anti-wetting property. First, the recent advancements in applications of textile wastewater treatment by MD including membrane types and configurations are provided. Second, the wetting mechanism in MD is analyzed to provide insights for anti-wetting modifications. Finally, recent developments on improving anti-property of MD membranes are described in order to reveal the progress in the textile wastewater treatment.

2. MD Membranes and Textile Wastewater Treatment

2.1. MD Membranes

Polymers such as PTFE [25], polypropylene (PP) [26], and polyvinylidene fluoride (PVDF) [27] are the most widely applied membrane materials for the MD processes due to their intrinsic or modified hydrophobicity, low cost, and ease of scale-up [22]. According to the polymer physicochemical characteristics and applications, various fabrication processes including nonsolvent induced phase separation (NIPS), thermally induced phase separation (TIPS), melt extrusion stretching, sintering, and electrospinning, as well as the modification methods including graft polymerization, plasma polymerization, interfacial polymerization, and dip-coating technologies are applied to obtain membranes with desired separation properties for MD application [28]. For PTFE, the membranes are usually fabricated from sintering or melt-extrusion where a paste composed of PTFE powders and lubricating agents are extruded to form a film or hollow fiber followed by heating and expanding to obtain a microporous membrane [25]. In contrast, PVDF is soluble in solvents such as dimethylformamide, *N*-methyl-2-pyrrolidone, and dimethylacetamide. The obtained PVDF dope can be processed either by NIPS, TIPS, or a combination of TIPS and NIPS methods to form MD membranes [27]. PP can be fabricated into membranes by both melt-extrusion stretching and the TIPS processes. However, nitrogen is often needed as inert gas to avoid chemical oxidation of PP [29]. Recently, new types of the membranes including mixed matrix membranes, dual-layer membranes, 2-dimensional lamellar membranes, zeolite membrane, and metal–organic framework-based membranes have elicited a lot of attention for MD applications due to their unique physicochemical properties [30–36]. For example, the water vapor permeation through a lamellar graphene membrane is reported to occur via the high-density nanochannels in the overlapped graphene multilayers and a high-water flux of ~50 L/m^2 h for 4 cm^2 of effective membrane area could be obtained [35] This membrane also exhibited good antifouling properties by rejecting common oil and surfactants over 48 and 72 h, respectively.

2.2. Membranes Applied in Textile Wastewater Treatment

For textile wastewater treatment by MD process, some representative MD membranes for textile wastewater treatment are summarized in Table 1. An early attempt was carried out by Calabro and co-workers in 1991 where a commercial PP membrane was used to separate water from dye solution [37]. The effects of feed temperature and flow rate on the permeation flux and separation efficiency were investigated and the experimental results demonstrated the potential of MD for textile wastewater treatment to produce highly purified water. Banat et al. [38] used a tubular module with PP membrane for the treatment of the methylene blue/water mixture by applying vacuum to the permeate side (VMD). The results demonstrated that the dye molecules were concentrated in the feed side without being detected in the permeate. Different operating conditions including feed temperature, flow rate, and dye concentration were studied to illustrate their effects on the separation process. In addition, a numerical model was developed and validated using experimental data for better understanding. In another study, Khayet and co-workers investigated the PVDF membranes for the treatment of both cationic (Maxilon Blue 5G, Drimarene Yellow K-2R) and anionic sodium fluorescein dye solutions as well as membrane fouling caused by those dye molecules [39]. It was found that the cationic dye was more prone to penetrate into the pores of the PVDF membrane, whereas the anionic dye was subject to the repulsive forces from the membrane matrix. As a result, anionic dye could lead to the formation of a fouling cake on the membrane surface. Li and co-workers compared the separation performances of commercial PVDF and PTFE membranes in DCMD processes for textile wastewater treatment [40]. The PTFE membrane (0.22 um pore size) was demonstrated to exhibit higher permeation flux and rejection to contaminants than that of the PVDF membrane (0.22 um pore size) due to its enhanced hydrophobicity and superior wetting resistance. Consequently, in the 48-h continuous operation, the PTFE membrane exhibited 90% COD removal and 94% color removal when the feed solution was the discharge outlet of the dyeing vat wastewater. Dow et al. also used PTFE membranes (6.4 m^2) to assess the long-term performance of DCMD in the textile wastewater treatment in combination with pretreatment [18]. They first conducted a laboratory-scale long-term performance study where a 41-fold increase in the feed concentration and over 99.9% non-volatile sulfate rejection were obtained. Further, a plant integration assessment in a textile mill was conducted to demonstrate the feasibility towards zero discharge using MD process provided the saline streams are first isolated and waste heat integration was available.

Table 1. Summary of typical membrane materials as membranes used for textile wastewater treatment (AGMD refers to air gap MD).

Membrane Material	Pore Size (µm)	Feed	Configuration	Feed/Permeate Temperature (°C)	Flux (kg/m^2 h)	Rejection (%)	Reference
PVDF	0.09–0.14	Dye solution	DCMD	80/20	5.64	99.78	[14]
PVDF-Cloisite 15A	0.088	Industrial effluent	DCMD	90/25	13–22	COD: 89.6 Color: 95.3	[41]
PVDF	0.45	Dye solution	DCMD	60/20	17.45	100	[44]
PDMS/PVDF-HFP	0.49	Dye solution	DCMD	60/20	25.23	100	[43]
PTFE	0.22	Industrial effluent	DCMD	50/20	~17.5	99.8	[40]
PVDF	0.22	Sodium Fluorescei solution	DCMD	70/20	21.7	99.73	[39]
SAN4/HIPS	0.43	Industrial effluent	DCMD	52/12	23.56	COD: 99.28 Color: 100	[42]
PTFE	0.2	Rose bengal solution	AGMD	70/20	12.6	100	[45]
PP	0.2	Dye solution	VMD	40–60/vacuum	27.5–57	100	[46]
PTFE/agarose hydrogel	0.2	Dye solution	DCMD	60/21	24.7	99	[47]
PVDF/PVP	0.17	Dye solution	DCMD	60/20	8.92	99.54	[48]

Apart from the neat PP, PVDF, and PTFE membranes, other kinds of materials used as the MD membrane materials have also been increasingly investigated in the application of textile wastewater treatment. Mokhtar et al. [41] utilized a phase inversion method to prepare a hollow-fiber PVDF-Coloisite 15A nanocomposite membrane for industrial textile wastewater treatment in DCMD. The results exhibited promising removal efficiencies for chemical oxygen demand (COD) (90.8%), dye (95.3%), and total dissolved solids (TDS) (93.7%). However, there was a decline of almost 50% in the permeation flux in the first few hours owing to membrane fouling, wetting, and thermal polarization effect. Meshkani and co-workers prepared a dual-layer nanofibrous membrane using styrene-acrylonitrile (SAN4) polymer as the bottom layer and high-impact polystyrene (HIPS) polymer as the top layer via a gas-assisted electrospinning process [42]. As a result, increased mass production and deposition speed were rendered by the gas flow due to the additional stretching force. Compared with the PTFE control membrane (98.64%, 99.75%, and 99.35% for COD, color, and TDS removal, respectively), the prepared membranes exhibited 99.28%, 100%, and $\geq$99.79% removal for COD, color, and TDS, respectively, in the membrane permeate side as shown in Figure 1, demonstrating remarkable potential for industrial textile wastewater treatment. An et al. [43] modified the electrospun poly(vinylidene fluoride-co-hexafluoropropene) (PVDF-HFP) membrane with extra electrospraying of hybridizing polydimethylsiloxane (PDMS) polymeric microspheres onto the PVDF-HFP membrane as shown in Figure 2. The fabricated membrane showed increased surface hydrophobicity and roughness. Meanwhile, complete color removal was achieved in DCMD process for differently-charged dyes/water mixtures with pure water production that was 50% higher than that of commercial PVDF membranes.

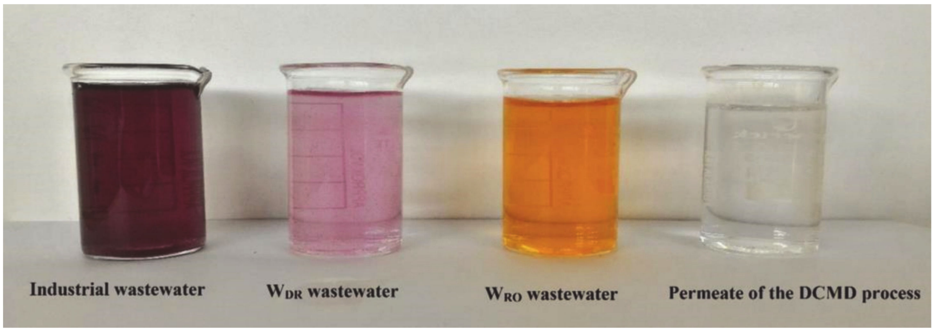

Figure 1. A visual comparison between the feed samples (industrial wastewater, wastewater by dispersing red dye (W_{DR}), and the wastewater by reactive orange dye (W_{RO})) and the permeate of the DCMD process using nanofibrous SAN4-HIPS membrane. Reproduced with permission from the authors of [42], published by Elsevier, 2020.

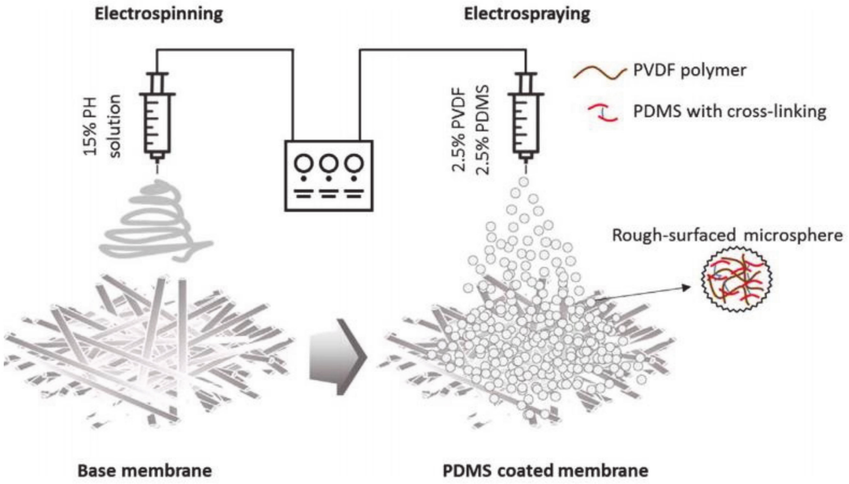

Figure 2. Fabrication of the hybrid PVDF-HFP/PDMS membrane using electrospinning technology. Reproduced with permission from the authors of [43], published by Elsevier, 2017.

2.3. *MD Configurations Applied in Textile Wastewater Treatment*

The most reported MD configuration for textile wastewater treatment is DCMD, where the hot feed and cold permeate water are in direct contact with the membrane [49]. Evaporation occurs at the interface between the feed and membrane, and then the water vapor transports through the membrane driven by vapor pressure difference. AGMD is another popular configuration by employing a thermal insulation layer, i.e., a cavity filled by air between the membrane downstream side and a chilled surface [50]. The vapor can thereby be condensed on the surface rather than dissolved in the cold circulating fluid. AGMD exhibited less energy loss than DCMD due to the insulating effect of the air gap [51]. In addition, AGMD can exhibit better separation performance in textile wastewater treatment. Leaper and co-workers [45] compared the separation performances of AGMD and DCMD using simulated solution composed of NaCl and either sunset yellow (SY) or rose bengal (RB) dyes and sodium dodecyl sulfate (SDS) surfactant. The AGMD exhibited higher color removal than DCMD. However, partial pore wetting was still observed after continuous operation for 70 h, which was caused by the membrane fouling by SDS and RB dye. In addition, special attention should be noted in the vacuum MD (VMD) process because the vacuum applied in the downstream side of the membrane could provide higher pressure difference across the membrane than DCMD and AGMD configurations. Meanwhile, different VMD process conditions also exert influence on the wetting. For example, the cross-flow VMD exhibited stable salt rejection over 50 h operation in desalination whereas pore wetting was observed in the submerged VMD process within the first 8 h in the same application [52].

3. Membrane Wetting Phenomena in MD

3.1. Wetting Behavior in MD

Although the MD process exhibits potential for the purified water production from the textile effluents, membrane pore wetting is still one of the most critical challenges limiting its practical implementation [53,54]. The membrane wetting is a phenomenon that the hydrophobicity of membrane decreases during the MD separation process and the feed liquid can thereby penetrate into the membrane pores instead of vapor [54–56]. Gryta et al. proposed that different degrees of membrane wetting are present during the MD operation, and they can approximately be divided into four types: non-wetted, surface-wetted, partially-wetted, and fully wetted [57]. As depicted in a continuous evolution in Figure 3, it was investigated that the membrane wetting can cause variations in permeation flux and deterioration of the membrane selectivity based on the wetting types. In the surface wetting phenomenon, the interface of liquid/vapor can be located inward of the membrane pores. This leads to increases in the temperature polarization as well as scaling inside the pores, which decreases the throughput by restraining diffusion of solutes from the bulk solution to the wetted pores [58,59]. For the partial wetting pores, some of the pores are fully wetted and the membrane throughput can be decreased (blue solid line) as occurred in surface wetting or increased (blue dash line) once the liquid transport through the membrane is dominated over the vapor transport. However, in this case, a rapid decrease after the initial increase of flux can be observed owing to the simultaneous intrusion of foulants and consequent blockage of pores depending on the characteristics of the foulants. Furthermore, when the pores of the membrane are fully wetted, the transport of solutes takes the form of viscous flow rather than vapor flow. The membrane no longer exhibits selectivity since the components in the feed can reach the permeate side indiscriminately. As a result, the total flux of the membrane increases (or decreases if the pores are blocked by foulants) and the rejections are lost.

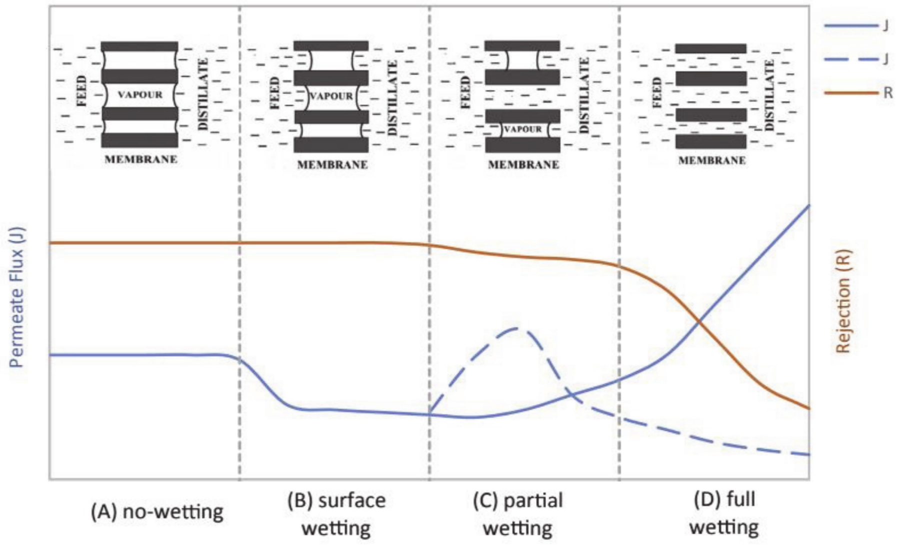

Figure 3. Evolution of permeation flux and rejection with different wetting degrees. (**A**) non-wetted, (**B**) surface-wetted, (**C**) partially-wetted, and (**D**) fully-wetted. Reproduced with permission from the authors of [17], published by Elsevier, 2018.

3.2. Causes of Membrane Wetting

In MD processes, the hydrophobicity of the membranes and surface tension of the feed enable a convex meniscus at the membrane pores, i.e., an interface of the liquid without entering the membrane pore. It reaches a stable state where the pressure differences caused by the surface tension at the interface and the partial pressures of vapors across the membrane are in equilibrium [17]. In other words, the vapors are in equilibrium with the liquid meniscus. The surface tension holds back the hydraulic pressure. Otherwise, pore wetting happens by allowing the feed liquid to penetrate the membranes and thus continuous water flow is formed locally through the pores. There are several causes to break the balance during the MD processes, including membrane fouling, surfactants in the feed, membrane degradation, and capillary condensation [54,60–62]. The primary cause of the MD membrane wetting is fouling. Fouling deposits and scaling on the membrane surface, as well as in the pores, can lead to reduction of the liquid entry pressure (LEP; the pressure (Pa) required to force the solution to pass through pores of membrane) as the deposits are commonly hydrophilic. Consequently, the amount of water in the permeate side as well as its purity can be decreased due to the membrane wetting and deposition that blocks the pores. Another cause is the existence of surfactants related to the reduction of the surface tension of the feed solution, which allows the passage of the feed in liquid state instead of building a convex meniscus at the pore. This will be a main reason causing membrane wetting when using MD for textile wastewater treatment as surfactant is commonly present in textile wastewaters. Additional causes include capillary condensation and membrane damage. Capillary condensation means the decline of the saturation pressure for vapor and the pores filled by condensed liquid from the vapor [63]. That also results in the pore wetting and consequent loss of membrane selectivity. Membrane degradation in terms of its chemical structure can also affect the wetting property of membranes, especially during the long-term MD application. It has been reported that chemical oxidative degradation on the membrane surface renders hydrophilic groups such as hydroxyl, carbonyl, and unsaturated groups. As a result, those groups reduce the surface hydrophobicity of the membrane and give rise to penetration of the feed solution and membrane wetting [61].

Depending on the types of dyeing processes, the textile effluent is a multicomponent mixture containing various dyes, surfactants, leveling agents, softeners, and salts. Therefore, the above-mentioned causes for membrane wetting can be found in MD process during textile wastewater treatment [13]. For example, surfactants in the feed can form attractive interactions with the membrane via hydrophobic–hydrophobic interactions, with the hydrophilic end of the surfactants that are able to attract water [64–66]. Consequently, MD membranes can be modified by the surfactants to exhibit reduced hydrophobicity, which increases the incidence of pore wetting. In addition, the existence of organic compounds in the feed such as dyes, oils, and alcohols also play a critical role in wetting occurrence owing to the decline of surface tension of the feed solution [17,67]. For the inorganic components, scaling of salts on the membrane surface due to concentration polarization, can also lead to a decline of MD membrane hydrophobicity and subsequently induce water intrusion into the pores [68–70].

4. Approaches for Antiwetting in Textile Wastewater Treatment

So far, various methods to mitigate or control the membrane wetting in textile wastewater treatment have been investigated with particular focus on attaching a hydrophilic layer on the hydrophobic membrane to avoid the interactions of the membrane materials with the foulants in the feed. Other efforts are made in the integration of pre-treatment steps, configuration optimization, and controlling operating conditions for anti-wetting purpose. The following section will discuss those approaches applied in the textile wastewater treatment by MD in detail.

4.1. Dual Layer Membrane

Essentially, the nature of membrane materials and the compositions of the feed determine the interaction between them [71]. Thus, material with desired antiwetting properties is beneficial to control membrane wetting. Although significantly improved antiwetting properties have been achieved by applying dual layer membranes containing hydrophilic surface layer and hydrophobic layer [72–75], there are only a few applied in the textile wastewater treatment. Polymers including PVDF, PP, and PTFE are the dominating materials for MD membranes. Therefore, current studies are mainly focused on hydrophilic layer attachment on those polymers for obtaining enhanced antiwetting property.

Lin et al. prepared a hydrophilic-modified membrane for DCMD application by simply holding an agarose hydrogel layer and the PTFE layer together by a frame as shown in Figure 4 [47]. The obtained composite membrane was first examined using aqueous NaCl solution containing SDS, Tween20, and Tween85 as the feed. It was shown that the wetting phenomenon was not observed until concentration of surfactant was above critical micelle concentration, where the surfactant in the form of micelle could penetrate through the hydrogel layer. Consequently, the authors proposed that the repellence of hydrophobic moiety of the surfactant by the hydrogel phase could render the agarose hydrogel covered membrane with enhanced antiwetting property. The treatment of polyester fabric dyeing wastewater using this composite membrane was compared with that of the bare PTFE membrane. The bare PTFE membrane exhibited the flux decreasing rapidly from over 30 to 7.9 L/m^2 h and the electrical conductivity increasing from 4.6 to 8.7 μS/cm over 24 h operation. In contrast, both the flux and the electrical conductivity of the agarose-covered PTFE membrane remained unchanged for 24 h, proving the effectiveness of using agarose layer in MD process for stable dyeing wastewater treatment. Similarly, García et al. employed a polyurethane coated PTFE membrane for a pilot trial of textile effluent directly from the plant [76]. The custom-made MD membrane exhibited good antiwetting property by rejecting surfactant as illustrated in Figure 5, whereas the permeate electrical conductivity of the control PTFE membrane was rising during the MD process. The hydrophilic-coated MD membrane exhibited viability for real textile wastewater treatment.

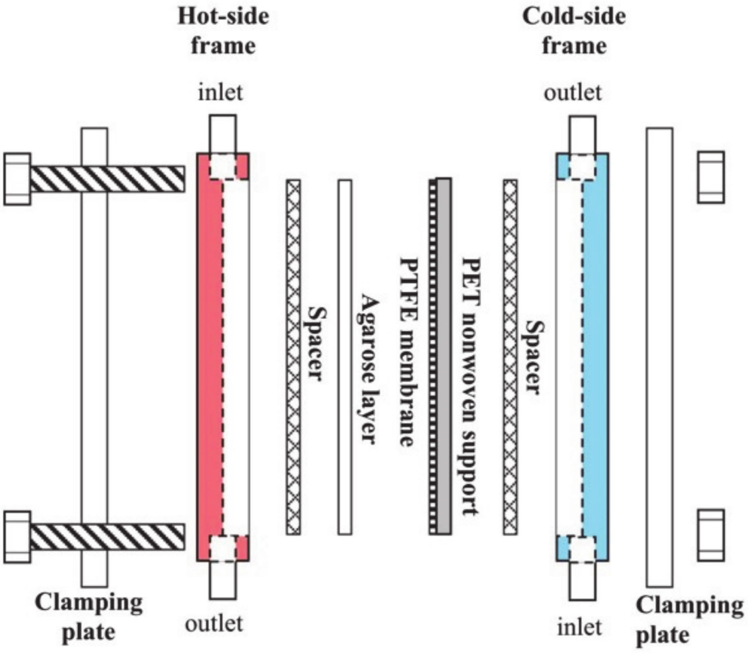

Figure 4. The architect of the module with dual layer (agarose hydrogel layer and hydrophobic layer) membrane. Reproduced with permission from the authors of [47], published by Elsevier, 2018.

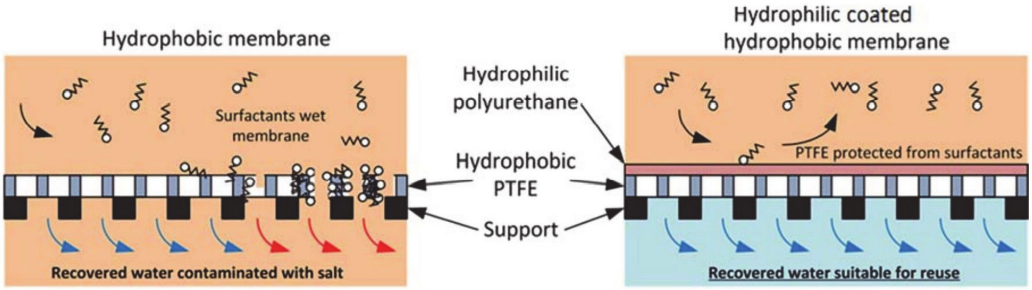

Figure 5. Wetting of the hydrophobic PTFE membrane and antiwetting property of the dual layer membrane. Reproduced with permission from the authors of [76], published by MDPI, 2018.

4.2. Surface Modification

The wettability of the MD membranes can be significantly affected by the surface chemistry and geometrical structures as they impact the surface free energy of the membranes [77,78]. To date, there have been two types of modifications applied on the MD membranes: chemical and physical modifications. Chemical modification can provide a reduction in surface free energy. Therefore, it is commonly realized by attaching low surface energy molecules or functional groups on the membrane surface [56,79–81]. Physical modification usually aims to increase the surface roughness by methods such as plasma treatment, layer-by-layer assembly, template replication, phase separation, electrospinning, or thermal treatment [82–87]. As a result, those surface modifications can effectively render superhydrophobicity with the MD membranes for antiwetting purpose. However, when used in MD application, superhydrophobic membranes are not able to avoid the fouling of organic molecules. As such, omniphobic membranes are intrinsically preferred as they provide superior wetting resistance to liquids containing oils, alcohols, and SDS. So far, enhancing the stability of omniphobic membranes, simplifying the complicated fabrication procedures, and lowering the cost of large-scale fabrication are the main challenges impeding the development of omniphobic membranes [88].

Although there are a number of surface modified membranes for desalination by MD, only very few studies have reported data for textile wastewater treatment. An et al. prepared a negatively charged superhydrophobic PDMS/PVDF membrane by coating PDMS microspheres onto the PVDF-HFP using electrospinning [43]. Compared with commercial PVDF and electrospun PVDF-HFP membranes, the deposited layer of PDMS particles exhibited a structure of three-dimensional hydrophobic barrier with a significant enhancement in surface hydrophobicity (contact angle, CA = 155.4°) and roughness (Ra = 1285 nm) as shown in Figure 6. The PDMS/PVDF membrane exhibited a 50% higher productivity without wetting than the commercial PVDF and electrospun PVDF in treating synthetic dyeing wastewater. For antiwetting property, it was reported that the negatively charged surface of the membrane easily repelled the sulfonate groups of the dyes, which led to a formation of a dye–dye structure on the E-PDMS membrane surface instead of within the membrane pores. As a result, this membrane exhibited stable dye removal without fouling or wetting during a 24-h test.

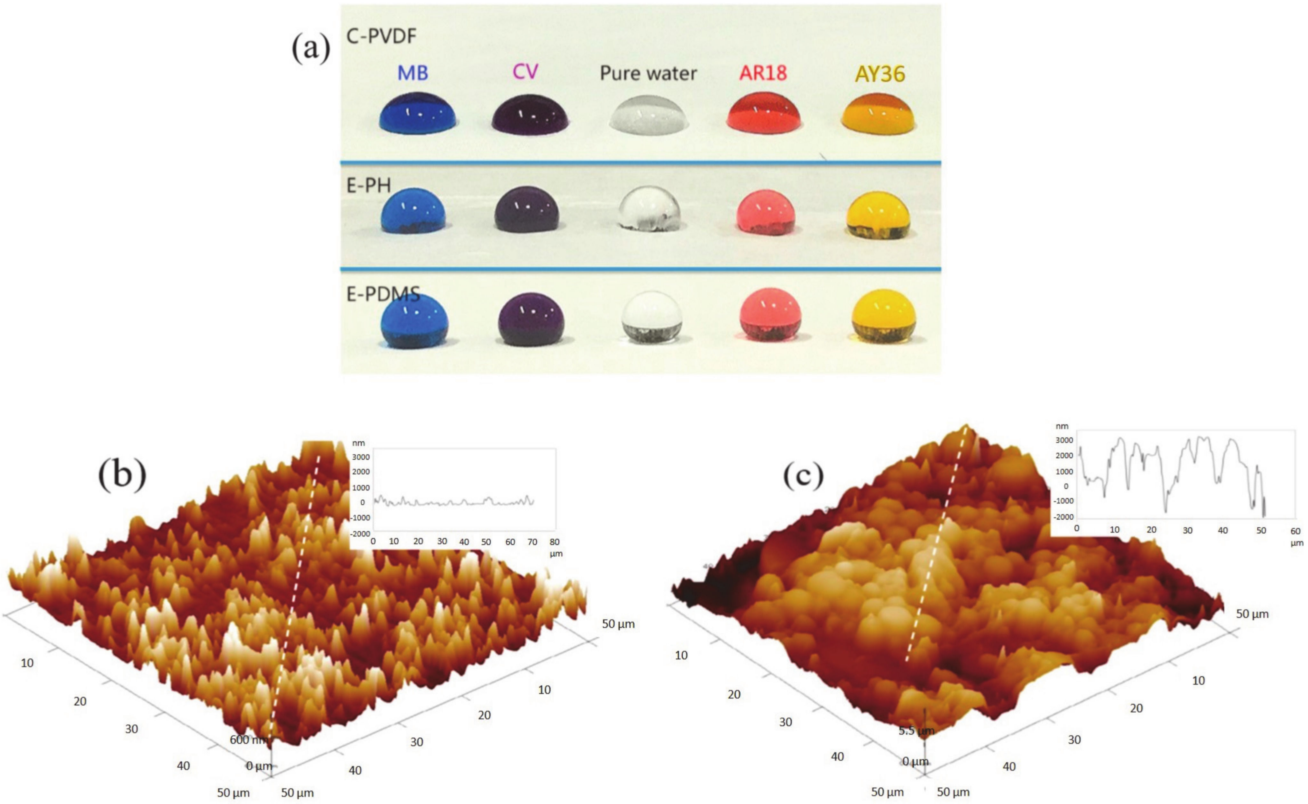

Figure 6. (**a**) Images of droplets of pure water and methylene blue (MB), crystal violet (CV), acid red 18 (AR18), and acid yellow 36 (AY36) on C-PVDF (commercial PVDF) membrane, E-PH (electrospun PVDF-HFP) membrane, and E-PDMS (electrospun PVDF-HFP/PDMS) membrane. Surface roughness of (**b**) C-PVDF and (**c**) E-PDMS. Reproduced with permission from the authors of [43], published by Elsevier, 2017.

4.3. Hybrid Treatment Process

Apart from the investigation of membrane materials, other manipulations such as a robust pretreatment of the textile wastewater or integrated processes with other membrane processes can also improve the wetting resistance and the stability of the MD process for textile wastewater treatment [89]. Basically, these processes aim at removing the contaminants and foulants that could lead to the membrane wetting before they contact the membranes [90,91]. In previous studies, different methods have been applied to enhance the antiwetting property of the MD process towards various feeds [22,49,92–96]. For example, accelerated precipitation softening (APS) of the concentrate from a RO process was employed by pH adjustment with sodium hydroxide along with calcite seeding [97]. Then, a MF process was further applied to the feed to remove the seeds followed by DCMD for the desalination process. Compared with the sharp decline of flux in desalting the RO concentrate by DCMD, only 20% of permeation flux was observed within 300 h running after the implementation of the APS treatment. In another study, ultrasonication was integrated with the MD process to mitigate PTFE membrane scaling [98]. Specially, the membrane module was immersed vertically in an ultrasonic bath equipped with transducers, which could provide an ultrasonication of 20 kHz (frequency) and 260 W (acoustic power). As a result, the ultrasonic wave enabled refreshment at the liquid–membrane interface with reduced concentration polarization, thereby restraining membrane scaling caused by $CaSO_4$ as well as the flux decline.

As introduced in the introduction section, foam fractionation and anaerobic/aerobic digestion were applied as a new strategy on the untreated effluent. Then, the pretreated textile effluent was then subject to long-term pilot testing using MD process. The MD process was stable without membrane wetting over the entire 3-month trial duration and the final water recovery of this trial was 91.6%. In addition, an assessment for zero liquid discharge was provided to demonstrate that the MD process would be viable when it is coupled with RO process. Li et al. [99] used a forward osmosis (FO)/MD hybrid process during textile wastewater treatment where the draw solution (DS) for FO was also employed as the feed solution for the MD process as shown in Figure 7. Thus, the MD process was designed to recover or reuse the DS of the FO process for continuous and stable separation performance. Economic analysis was further conducted on this hybrid process and the results showed the lowest total cost of about 0.17 USD for treating 500 mL textile wastewater. A photocatalysis and MD-integrated process was developed for reactive dye solutions (Red 180 and Orange 16) [100]. The hybrid processes contain the photocatalytic process for the dye solutions with 1 g/L ZnO catalyst loading under ultraviolet irradiation and subsequent MD processes. As a result, the MD process effectively separated the catalyst particles from the treated solution while the hybrid process exhibited complete color removal efficiency for both of the dye solutions.

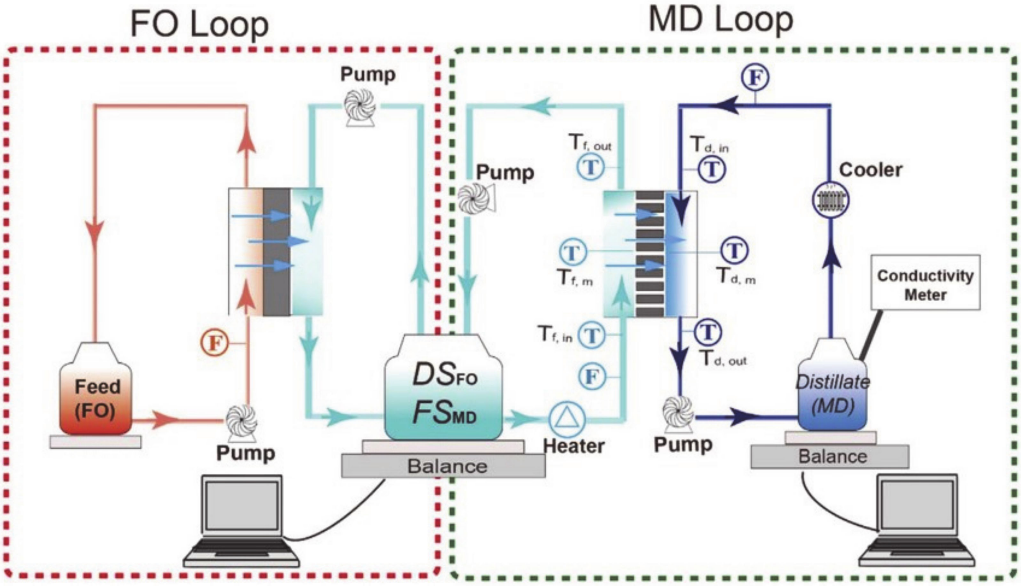

Figure 7. Schematic diagram of the FO-MD hybrid system for textile wastewater treatment. Reproduced with permission from the authors of [99], published by Elsevier, 2020.

5. Conclusions and Perspective

A summary of membranes for textile wastewater treatment by MD processes and the antiwetting modifications that are applied was described. Due to the complex composition of effluents from textile industry, textile wastewater treatment has aroused a lot of attention in the recent years. Hydrophobic polymers such as PVDF, PTFE, and PP have been widely used as the membrane materials for the textile wastewater treatment. However, membrane wetting is still limiting the stability of the MD processes. Previous literature shows that the wetting phenomenon can be caused by membrane fouling, surfactants, membrane degradation, and capillary condensation. Depending on the different degrees of membrane wetting, the permeation flux in the MD process can be either increased (partial wetting and full wetting) or decreased (surface wetting), while the purity of water in the permeate side usually declines. Several antiwetting modifications are discussed to inhibit the membrane wetting by preparing hydrophilic-hydrophobic dual layer membrane, modifying the surface chemistry and roughness of the membrane, and conducting pretreatment or hybrid process that removes the wetting agents from the feed solution.

Despite the prospect of MD processes for textile wastewater treatment, further study of the membrane wetting by textile effluent is still in need. It is important to investigate the various interactions between the membrane surface and the complex components in textile wastewater for further understanding. In addition, more effort towards efficient fabrication techniques should be made to manipulate the physicochemical properties of membranes towards better wetting-resistance ability. Developments of novel membrane materials such as graphene nanosheets with robust antiwetting and antifouling properties are also required to potentially enhance the separation performance in real textile wastewater treatment. In addition, the design of advanced modules needs further investigation to inhibit the decline of permeate quality upon the occurrence of membrane wetting.

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Abbreviations

MD	membrane distillation
DCMD	direct contact membrane distillation
AGMD	air gap membrane distillation
VMD	vacuum membrane distillation
RO	reverse osmosis
MF	microfiltration
FO	forward osmosis
DS	draw solution
PTFE	polytetrafluoroethylene
PP	polypropylene
PVDF	polyvinylidene fluoride
SAN4	styrene-acrylonitrile
PVDF-HFP	poly (vinylidene fluoride-co-hexafluoropropene)
PDMS	polydimethylsiloxane
COD	chemical oxygen demand
TDS	total dissolved solids
NIPS	nonsolvent induced phase separation
TIPS	thermally induced phase separation
LEP	liquid entry pressure
CA	contact angle
SY	sunset yellow
RB	rose bengal
SDS	sodium dodecyl sulfate
MB	methylene blue
CV	crystal violet
AR18	acid red 18
AY36	acid yellow 36
APS	accelerated precipitation softening

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