Evolution of Membrane Surface Properties for Membrane Distillation: A Mini Review

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ABSTRACT

To date, the membrane development for membrane distillation (MD) application is growing in line with the increasing volume of various types of wastewaters discharged into environment. MD is a liquid-vapor separation process and a hydrophobic membrane is used to retain the liquid. Theoretically, the hydrophobic membrane can achieve 100% rejection of non-volatile components that dissolved in feed liquids. As a result, MD has received significant attention in water recovery from saline water as well as wastewaters. Nevertheless, in addition to the scaling problem due to salts, the hydrophobicity property of membrane becomes a concern when dealing with challenging wastewaters which contain various types of low surface tension components such as oils, grease, alcohols, organics and surfactants. The membrane pore wetting due to salts deposition fouling and low surface tension components subsequently reduces the flux and even fails the liquid-vapor separation process. This article briefly discusses the transformation of MD membrane from hydrophobic to superhydrophobic and omniphobic which purposely to enhance the flux and eliminate the membrane pore wetting.

Keywords: Water recovery, membrane distillation, membrane modification, flux, wetting

1.0 INTRODUCTION

According to the United Nations Water, 80% of wastewater is discharged into the environment without being treated or reused, and 40% of the world’s people are affected by water scarcity [1]. Membrane technology has been examined as one of the promising technologies to treat various types of wastewaters for reusing purpose whereby the water reuse plays a critical role in circular economy and it is a potential strategy to achieve the Sustainable Development Goals [2]. Membrane distillation (MD) has been proven can highly recover the water because MD is not an osmotic limiting process [3, 4]. MD is a liquid-vapor separation process whereby only water vapor is allowed to transport through membrane and theoretically 100% rejection of non-volatile components [5]. The MD is basically categorized into four different configurations, they are direct contact membrane distillation (DCMD), air gap membrane distillation (AGMD), sweeping gas membrane distillation (SGMD) and vacuum membrane distillation (VMD). These MD configurations are differentiated based on the techniques of freshwater recovery of water vapor on permeate sides. Compared to pressurized membrane filtrations such as reverse osmosis and nanofiltration, MD shows

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lower fouling, less scaling, stable permeate flux and higher rejection \cite{6, 7}. Although MD requires thermal energy to elevate the temperature of feed waters, the low grade energy such as solar, geothermal and waste heat are sufficient to operate the MD \cite{8–10}. Recently, MD has been integrated with other membrane separations such as reverse osmosis, membrane bioreactor and forward osmosis to further enhance the water recovery \cite{11–14}.

A porous hydrophobic membrane which retains liquid phase and permits water vapor to permeate is used as the liquid-vapor separator. In the early stage of MD development, the materials used for MD membranes include polytetrafluoroethylene \cite{15}, polypropylene \cite{16}, polyvinylidene fluoride \cite{17} and polyethylene \cite{18}. These polymeric materials are excellent repellents towards liquid water as well as saline waters. However, the real seawater and brackish waters as well as wastewaters discharged from various industries contained the highly concentrated inorganic salts \cite{19, 20}, low surface tension contaminants, oils, organics and nutrients can promote membrane pore wetting which eventually deteriorate permeate quality and reduce flux \cite{21–24}. As a result, the basic hydrophobic membranes have been modified to become superhydrophobic \cite{25, 26} and omniphobic \cite{27} to minimize the membrane pore wetting. This short review discusses the evolution of MD membrane in improving the flux and minimizing the membrane pore wetting.

2.0 PHASE INVERSION MEMBRANES

The first patent on MD was filed by Bodell in 1963 and the membrane material was the silicone rubber \cite{28}. Other plastic materials which have a relatively high permeability to water vapor such as cellulose propionate, polyvinyl fluoride and cellulose acetate were also proposed for MD in a few decades ago \cite{29}. During the 1980s, polytetrafluoroethylene (PTFE), polypropylene (PP) and polyvinylidene fluoride (PVDF) were found as the most promising for MD application; and the membrane with high porosity 70 – 80% can be obtained from phase inversion fabrication method \cite{30, 31}. Figure 1 illustrates the phase inversion membrane fabrication technique.

In the early MD development for desalination application, membrane wetting is unavoidable due to salt crystal formed on the membrane surface \cite{32, 33}. A deposit stayed on the hydrophobic membrane surface can cause the pores adjacent to the deposit to be filled with liquid which leads to a wetting of this part of the membrane \cite{34, 35}. Even a low surface porosity can hinder the formation of scale inside the membrane pores, surface wetting is still possible and this also reduces the MD flux significantly \cite{36}. Schneider et al. \cite{37} recommended the membranes with wetting angles that greater 90° only are suitable for MD process in which PTFE, PVDF and PP meet this specification. Additionally, a recommended maximum pore diameter of approximately 0.5–0.6 μm can avoid the membrane wetting. While El-Bourawi et al. \cite{38} suggested that the pore sizes of the membranes used in MD should range between 10 nm and 1 μm to ensure no pore wetting. Lawson and Lloyd \cite{39} advised that the process equipment and process solutions must be free from the contamination of detergents or other surfacting agents because these materials can greatly reduce the surface tension of process liquid and the wetting angle. Nevertheless, these requirements have limited the applications of MD extended to the real wastewaters.
Consequently, various new techniques have been explored to minimize the membrane wetting. Peng et al. [40] adopted a hydrophilic layer on the hydrophobic PVDF membrane and revealed that the membrane can prohibit the membrane wetting even 25% ethanol was added to the brine feed solution. Gryta and Barancewicz [41] reported that the incorporation of PTFE particles into the PVDF membrane matrix with sponge-like structure has slow down the membrane wettability. Tian et al. [42] used bottom-up strategy to fabricate the PVDF membrane and the resultant membranes possessed high water contact angels (~144°) with rougher surfaces. Alberto et al. [43] coated the PVDF membranes with graphene oxide laminate and showed that the membranes were stable for at least 90 h with the feed solutions containing surfactants. Zou et al. [44] employed co-casting technique to fabricate the PVDF membranes and found the excellent rejections of brine with sodium dodecyl sulfate (SDS) surfactant and ginseng extracts were 99.9% for 120 h operation. Table 1 presents a few recent membrane fabrication strategies used to improve the performance of phase inversion membranes in MD. In general, the rejection performance of membranes is close to 99% which is satisfactory. However, the water contact angle (WCA) and liquid entry pressure (LEP) affect the MD flux and operating time as well as membrane wetting differently. DCMD fluxes increased and the operation extended when WCA and LEP of the modified membranes increased [42, 45–50]. On a contrary, AGMD flux increased when WCA decreased for the membrane fabricated from PVDF dope solution blended with activated carbon [51].
Table 1 Modification strategy of phase inversion membrane preparation for MD

| Membrane fabrication strategy | WCA (°) | LEP (kPa) | Configuration | Flux (kg/m² h) | Rejection (%) | Reference |
|-------------------------------|---------|-----------|---------------|---------------|--------------|-----------|
|                               | Pristine| Modified  | Pristine      | Modified      | Pristine     | Modified  |          |
| Bottom-up: PVDF dope solution was cast on a clean glass plate and the nascent film was covered by a piece of non-woven substrate. | 71.7 | 144 | - | - | DCMD | ~25<sup>a</sup> | 41.4<sup>a</sup> | 99 | 99 | [42] |
| Co-casting: PES, which acted as a sacrificial layer was immediately cast on PVDF nascent film. | ~75 | 125–133 | - | - | DCMD | 5–10<sup>b</sup> | 20–25<sup>b,c</sup> | > 99.95 | > 99.95 | [45] |
| Co-casting: ATBC or PEG400 was used as a sacrificial layer; sacrificial layer was cast on PVDF layer. | - | 120–130 | - | - | DCMD | - | 29–37<sup>d,e</sup> | 25–30<sup>d,f</sup> | >99.9<sup>d,g</sup> | [44] |
| Blending: 10 wt% PVDF dissolved in DMAC solvent with 1 wt% iron nanoparticle | ~97.7 | 99.2 | ~17 | ~23 | DCMD | ~11 | ~15 | > 99 | > 99 | [46] |
| Blending: 5 – 9 wt.% activated carbon in 12 wt.% PVDF and 79 – 88 wt.% DMAC | ~110 | 80–92 | - | - | AGMD | 0.02 | 0.08–3.2 | > 97 | > 97–99.9 |
| Texture casting substrate: Tempered glass, stainless steel mesh, Sandpaper and PDMS | - | 64.6±3.2<sup>i</sup> | 40±10<sup>m</sup> | 120±10<sup>i</sup> | AGMD | ~8.5<sup>m</sup> | ~19<sup>i,n</sup> | 97<sup>m,n</sup> | ~93<sup>i,n</sup> | [47] |
| Dip-coating: PEI hollow fiber membranes were immersed into PDMS solutions. | 82.5±0.94 | 103.8 ± 0.26 | 250 | 400 | SGMD | ~14<sup>q</sup> | ~17<sup>i</sup> | ~99.4<sup>q,p</sup> | ~99.9<sup>q,p</sup> | [48] |
| Dilute solution coating: PVDF hollow fiber membranes were immersed into the ZIF-71 /PVDF/PEG/DMF solutions at different compositions. | 94.5 | 115.5–136.5 | ~170 | 168–215 | VMD | 11–14<sup>q</sup> | 19–28<sup>q</sup> | > 99.9 | > 99.9 | [50] |
| Graft copolymerization: Introduced EA monomer onto the backbone of PVC. | 79±1 | 90.5±1<sup>c</sup> | - | - | VMD | 2.52 | 37.5 | - | - | [49] |

<sup>a</sup> Feed temperature at 70°C; <sup>b</sup> 18–20 wt.% PVDF and 15–21 wt.% PES; <sup>c</sup> same flux when operated for 48 h; <sup>d</sup> 120 h operation; <sup>e</sup> feed contained 35 g/L NaCl; <sup>f</sup> feed contained 35 g/l NaCl and 0.1 M SDS; <sup>g</sup> feed contained 35 g/L ginseng extract; <sup>h</sup> after nanoparticle leaching and stability tests; <sup>i</sup> glass casting substrate; <sup>j</sup> stainless steel mesh casting substrate; <sup>k</sup> sandpaper casting substrate; <sup;l</sup> PDMS casting substrate; <sup>m</sup> commercial PVDF membrane; <sup>n</sup> 40 h operation; <sup>o</sup> 135 h operation; <sup>p</sup> dye rejection; <sup>q</sup> 60 h operation; <sup>r</sup> 14 wt.% PVC; <sup>s</sup> 18 wt.% PVC; ATBC: Acetyl tributyl citrate; DMAC: N, N-Dimethylacetamide; LEP: Liquid entry pressure; PDMS: Polydimethylsiloxane; PEG: Polyethylene glycol; PEI: Polyetherimide; PES: Polyethersulfone; PVDF: Polyvinylidene fluoride; SDS: Sodium dodecyl sulfate; WCA: Water contact angle; ZIF: Zeolitic imidazolate framework.
3.0 ELECTROSPUN NANOFIBER MEMBRANES

Due to relatively low flux in phase inversion membranes, electrospun nanofiber membranes have been attractive for MD since 2008 [52]. The nanofiber membrane consists of fibers overlap with each other to form a thin film with highly interconnected open pore structure. Figure 2 shows a basic electrospinning method to fabricate the nanofiber membrane. The fluxes obtained from electrospun nanofiber membranes were as high as or even greater than those of commercial membranes, and the rejection of salt was approximately 98.7 – 99.9% which was applicable for a 25-day operation in an AGMD process [52]. Su et al. [53] revealed that the composite nanofibrous PVDF and PVDF-co-HFP membranes have exceeded the flux limit of commercial PTFE membrane. Essalhi and Khayet [54] reported that the increase of electrospinning time has decreased the maximum size of inter-fiber space because the thickness of nanofiber membrane increased correspondingly. As a result, the void volume fraction and LEP increased with increasing the electrospinning time. In spite of that, the MD flux decreased with increasing the electrospinning time because higher flux is preferable in thinner membrane. Although the nanofiber membrane made from hydrophobic polymer possesses high WCA, the application of nanofiber membrane alone in MD process is still limited due to its LEP is too low. Prince et al. [55] documented that a single PVDF nanofiber layer cannot be used in their AGMD desalination system because the penetration of salt increased dramatically to more than 90% within the first few minutes even WCA of the membrane was 145.7°. The LEP of nanofiber PVDF membrane increased approximately 8.7 times higher when the nanofiber membrane was supported by a casted membrane (i.e. phase inversion membrane) and a support layer. The triple layer membrane made the AGMD desalination process continuously operated at least 40 h without membrane wetting occurred. While the phase inversion membrane was wetted after 10 h of operation. On the other hand, Liao et al. [56] applied heat-press post-treatment on the fresh nanofiber membrane and this treatment has increased the LEP of membrane. The MD fluxes obtained from heat-pressed nanofiber membrane were also higher than that untreated nanofiber membrane as well as commercial phase inversion membrane even the pore size, WCA and porosity were slightly reduced due to structural compaction [56, 57]. The properties of PVDF nanofiber membrane shifted from hydrophobic to superhydrophobic when the content of clay nanocomposites blended in the spinning solution increased [58]. No flux declination was observed within 8-h DCMD operation when superhydrophobic membrane was tested. Table 2 compares the recent MD performance between electrospun nanofiber membranes and phase inversion membranes. The MD flux obtained by nanofiber membranes is relatively higher than that attained using phase inversion membranes even either WCA or LEP decreased in the nanofiber membranes [59–62].

4.0 SUPERHYDROPHOBIC MEMBRANES

By definition, the superhydrophobic surface possesses high WCAs that exceed 150° and low water sliding angles which is typically below 10° [63]. In 2009, Ma et al. [64] have found that superhydrophobic glass membranes with WCAs exceeded 160° showed better anti-fouling ability and higher flux than conventional hydrophobic membranes in their AGMD desalination system.
Starting from 2012, both polymeric phase inversion and electrospun nanofiber membranes [58, 65, 66] have been actively modified to attain the superhydrophobic property to recover freshwater from saline water. The application of superhydrophobic membranes has also been expanded to feed solutions containing organics such as humic acid and surfactants [76, 68]. Table 3 shows a few samples of latest researches of superhydrophobic membranes fabricated from different strategies and their comparisons with pristine hydrophobic membranes. In overall, the WCA and LEP as well as MD flux are improved significantly in superhydrophobic membranes. Although some authors reported that the MD flux gained by superhydrophobic membranes is somewhat lower than that attained by hydrophobic membranes, the superhydrophobic membranes can work for longer operation with good permeate quality [69–71]. Compare to hydrophobic membranes, the superhydrophobic membranes exhibited good anti-scaling, anti-fouling, anti-wetting, high and stable flux, good salt rejection and suitable for long-term MD desalination operation [72–76].

5.0 OMNIPHOBIC MEMBRANES

The performance of MD could be deteriorated if fed with challenging wastewaters that comprise of various types of inorganics and organics including surfactants discharged from oil and gas industries. For instance, membrane wetting occurred when the feed solution contained cationic surfactant $N,N,N$-trimethyl-1-dodecanaminium bromide even the membrane used was superhydrophobic [68].
### Table 2 Comparison of electrospun nanofiber membrane (ENM) and phase inversion membrane (PIM) in MD

| Composition of ENM | WCA (°) | LEP (kPa) | Configuration | Flux (kg/m²h) | Rejection (%) | Reference |
|--------------------|---------|-----------|---------------|---------------|---------------|-----------|
| 5 wt% PVDF in DMF/Acetone with 0.004 wt% LiCl | 135 | 136 | - | 35 | DCMD | 10.6 | 20.3 | - | - | [56] |
| 15 wt% PVDF-HFP in 68 wt% DMF and 17 wt% acetone with 0.005 wt% LiCl and 1.5 AC wt% | 140.8±0.8 | 142.7±0.6 | 234±3 | 136±4 | DCMD | 41.8 | 45.6 | ~100 | ~100 | [59] |
| 0.5 wt% CNT and 20 wt% PVDF-HFP in 8 wt% and 2 wt% acetone | - | 140.7±2.2 | - | 50±2.0 | DCMD | 13.2 | 18.7 | >99.99 | >99.99 | [77] |
| 18 wt% SAN in DMF/Acetone 70/30 | 124.4±0.8 | 148.6±0.65 | 119.7±3.1 | 101.6±2.5 | DCMD | 15–21 | 21–27 | 98.15 | 97.10 | [60] |
| 17.5 wt% PVDF-HEP in 16.5 wt% acetone and 66 wt% DMF with 1–2 wt% AlFu MOF | ~100 | 135±0.3 | - | 110 | DCMD | 15.64 | 22.7 | >99.9 | >99.9 | [78,79] |
| 20 wt% PVDF-HEP in DMF/Acetone (8/2, v/v) with 0.5 wt% ZIF-71 | 142.6±2.4 | 134±1.2 | 90 | 90±2 | DCMD | ~13.8 | 20 | >99.9 | >99.9 | [61] |
| 17 wt% PVDF in DMAC/Acetone (7/3 to 3/7, w/w) | 130.3±0.05 | 134–148 | - | 23.9–107.6 | DCMD | ~9 | 10–13 | - | - | [80] |
| 15 wt% PVDF in 38.75 wt% DMAC and 38.75 wt% acetone with 7.5 wt% PTFE micro powder | - | 122.4±0.3 | - | 80 | AGMD | ~14 | 27.7 | - | >97% | [81] |
| 15 wt% PVDF in DMF | 130±4 | 230±18 | 82±17 | DCMD | ~23 | 61.8 | >99.99 | 99.94 | [62] |
| 18 wt% PVDF/TBAC in acetone/DMF and 3 wt% FA | 107±5 | 135±4 | 180 | 210 | DCMD | 0 | 53.43 | 97.7 | 99.6 | [82] |

* Commercial Millipore Durapore membrane; † L/m²h is equivalent to kg/m²h; ‡ commercial PTFE supplied from Porous Membrane Technology (Ningbo, China); § commercial PTFE-PVDF/PET microporous membrane purchased from Shanghai Mingjie New Material Co. Ltd; ¶ 210 min operation; ‰ commercial PTFE membrane supplied by Membrane-Solutions Co., China; † 10–14 h operation; ① rejection of COD; ② rejection of color; ③ 46 h operation; ④ 3 h operation; ⑤ commercial PVDF membrane purchased from Aquastill (Sittard, Netherlands); ‡ ⑦ commercial PVDF0.22 membrane; ⑧ commercial PVDF membrane purchased from Millipore; ⑨ 50 h operation; ⑩ dye rejection; ⑪ commercial PVDF membrane supplied by Haining yanguan zhongqi filtration equipment Co., Ltd.; ⑫ wetted after 3.5 h operation; ⑬ 5 h operation; AC: Activated carbon; AlFu MOF: Aluminium fumarate metal organic framework; CNT: Carbon nanotube; DMAc: N, N-Dimethylacetamide; DMF: N,N-dimethylformamide; FA: Fluorinated acrylate copolymer; LEP: Liquid entry pressure; SAN: Styrene-acrylonitrile; TBAC: Tetrabutylammonium chloride; ZIF-71: Zeolite imidazole ester skeleton; PVDF: Polyvinylidene fluoride; PVDF-HFP: Polyvinylidene fluoride-co-hexafluoropropylene; PTFE: Polytetrafluoroethylene; WCA: Water contact angle.
### Table 3: Comparison of superhydrophobic membrane (S-pho M) and pristine hydrophobic membrane in MD

| Membrane fabrication strategy | WCA (°) | LEP (kPa) | Configuration | Flux (kg/m² h) | Rejection (%) |
|------------------------------|---------|-----------|---------------|---------------|---------------|
|                              | Pristine | S-pho M   | Pristine | S-pho M | AGMD | Pristine | S-pho M | Pristine | S-pho M |
| Three-stage method of superhydrophobic mixed-matrix nanofiber membrane: Synthesize ODA-rGO by a hydrothermal technique; reducing mean pore size by LiCl; and hot-pressed. | 130±1 a | 158±1 b | 134±2 e | 89.6±1.2 b | AGMD | ~8 a | 16.3 c | 99.99 a,e | 99.99 k,d |
| Combination of immersion precipitation with rolling embossing method | ~88 a | ~151 e | 128 a | 397 e | VMD | ~5 a | ~27 e | > 99.6 a | > 99.9 e |
| Dip-coating process: PVDF ENM was dipped in the PDMS-based graft copolymer/TiO₂/ethyl acetate/isopropanol sol | 118.2±2.2 a | 150.4±3.1 b | 241±17 a | 88±8 e | DCMD | ~19 a | 34.5 b | 99.9 a,b | 99.9 b |
| Coating and UV-curing: PVDF membrane was coated by 5 wt% PFPE solution and cured by UV lamp | 133.4±0.9 a | 162.6±0.4 g | - | - | VMD | 19.2 a,i | 13.9 e,k | - | > 99.99 e,k |
| Sequential electrospinning and electrospraying with low-viscosity CB/F-POSS/PDMS-HFP | 132 e | 162 b | 102 e | 176.8 b | DCMD | 22.72 a | ~40 b | ~100 a | ~100 b |
| Spray-coating of hydrophobic SiO₂ | 98.5±2.1 a | 170.8±1.3 e | - | - | DCMD | 10.3 a,l | 19.1 e,i | ~99.6 a,l | 99.99 g,l |
| Acid pre-treatment and silanization with FAS | 132.2±1.9 e | 154.6±2.2 b | 67±3 e | 143±4 b | VMD | 7.5 c,m | 11–12 b,m | ~83 e,o | 99.99 h,o |
| PU electrospinning and PDMS polymeric microsphere electrospraying with pore-size tuning with mechanical strain | > 160 e | > 160 b | 19.6 e | 12.2 b | DCMD | 10.13 c,o | 36.59 h,o | 99.99 c,o | 99.96 h,o |
| One-step electrospinning and immersion in ethanol coagulant to form microsphere/nanofiber membrane | 138.5±1.8 e | 156.6±1.4 b | 131±20 e | 149±10 b | DCMD | 21.2 c,p | 33.9 h,p | > 99.95 c,p | > 99.95 b,p |

a Pristine PIM; b modified ENM; c durable for 40 h operation; d durable for 60 h operation; e pristine ENM; f durable for 4 h operation; g modified PIM; h permeate conductivity less than 2.5 μS/cm; i permeate conductivity exceeded 5 μS/cm; j 10 h operation; k 30 h operation; l 8 h operation; m 15 h operation; n 22 h operation; o 5 h operation; p 40 h operation; CB: Carbon black; ENM: Electrospun nanofiber membrane; FAS: Fluoroalkylsilane; F-POSS: Fluorinated polyhedral oligomeric silsesquioxanes; LEP: Liquid entry pressure; ODA-rGO: Octadecylamine-reduced graphene oxide; PDMS: Poly(dimethyl siloxane); PFPE: Perfluoropolyether; PIM: Phase inversion membrane; PU: Polyurethane; PVDF-HFP: Polyvinylidene-hexafluoropropylene; WCA: Water contact angle.
In 2014, Lin et al. [89] proposed omniphobic microporous membranes that can resist the wetting due to water and low surface tension substances. A hydrophilic glass fiber membrane was coated with silica nanoparticles, followed by surface fluorination and polymer coating. Afterwards, several fabrication techniques of omniphobic membrane have been developed, such as particle-free approach [90], scalable approach [91], self-roughened approach [92], solvent-thermal induced roughening method [93], etc. A re-entrance structure is created to improve the roughness of membrane surface by employing nanoparticles [94–98] and fluorination [97, 99]. Table 4 shows a comparison between omniphobic membranes and pristine hydrophobic membranes in MD performance. Omniphobic membranes can retain a numerous low surface tension liquids in which contact angles (CAs) exceeding 100°. Basically, the omniphobic membranes can improve the MD flux [100–102]. Some researchers showed that the flux gained by omniphobic membranes is slightly lower than that obtained by pristine hydrophobic membranes, but the omniphobic membranes allowed for longer operation with high rejection of salts as well as low surface tension materials [93, 94, 103]. The omniphobic membranes exhibited good anti-fouling, anti-scaling and anti-wetting in MD processes for salts, surfactants and organic matters [100, 103–106].

6.0 CONCLUSIONS AND FUTURE RESEARCH DIRECTIONS

Membrane distillation (MD) has been receiving rapid attention in water recovery since four decades ago. The MD membrane has been developed from basic hydrophobic, followed by superhydrophobic and omniphobic in either phase inversion or electrospun membrane. The remarks of future research directions include:

- Harvesting freshwater from real wastewaters and produced waters from oilfields using omniphobic membranes could be an interesting research subject in the future.
- Rigorous investigations of modified membranes for long-term MD applications are required, especially the stability of adhesion of nanoparticles and coating layers as well as the modified structures.
- Sustainable and natural materials are recommended to replace the current chemicals used to modify the membrane surface. Thus, to reduce the hazards and cost.

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Table 4 Comparison of omniphobic membrane (O-pho M) and pristine membrane in MD

| Membrane fabrication strategy | CA (°) | LEP (kPa) | Configuration | Flux (kg/m² h) | Rejection (%) | Reference |
|-------------------------------|--------|-----------|---------------|---------------|--------------|-----------|
| Immersion of PVDF membrane in DA, AgNO₃/ethanol and PFDT | 142.7±2.9<sup>a</sup> | 165±1<sup>a</sup> | 80±10 | 0<sup>··</sup> | 21.5±1.2<sup>··</sup> | - | >99.99<sup>i</sup> | [105] |
| | 0<sup>b</sup> | 157±1<sup>b</sup> | 220±20 | 0<sup>··</sup> | 187±15 | DCMD | - | >99.99<sup>i</sup> |
| | 122±2<sup>c</sup> | > 150<sup>c,d,f</sup> | 102±1<sup>c</sup> | - | 187±15 | - | - | - |
| | 100±1<sup>d</sup> | - | - | - | - | - | - | - |
| | 134±3<sup>e</sup> | - | - | - | - | - | - | - |
| PDA/PEI co-deposition, silicification by TMOS solution and PDTS coating | 121.89<sup>a</sup> | > 150<sup>a</sup> | 243.4 | 49.22<sup>k</sup> | 94.1<sup>f</sup> | - | >99 | [100] |
| | 71.86<sup>d</sup> | 157.31<sup>d</sup> | 326.4 | - | 13.6<sup>p</sup> | - | >99 | [100] |
| Blending of CA and SiNPs, and next immersion in PDTS | 125.7<sup>a</sup> | 155.6<sup>a</sup> | 151.49<sup>a</sup> | 134±3<sup>e</sup> | 119 | - | - | - |
| | 79<sup>a</sup> | 135<sup>m</sup> | 140.64<sup>b</sup> | 107.5<sup>r</sup> | 223 | 33<sup>y</sup> | - | ~100<sup>j</sup> | [107] |
| | 120<sup>n</sup> | 120<sup>n</sup> | 119.59<sup>d</sup> | 107.5<sup>r</sup> | 151.6<sup>a</sup> | 20<sup>y</sup> | - | - | [107] |
| | ~30<sup>n</sup> | ~30<sup>n</sup> | 107.5<sup>r</sup> | ~30<sup>n</sup> | 119.1<sup>a</sup> | ~135<sup>m</sup> | - | - | [107] |
| Blending of PVDF-HFP and APTES, in-situ growth of SiNP and silanization | ~130<sup>a</sup> | ~110<sup>d</sup> | 151.49<sup>a</sup> | 151.49<sup>a</sup> | 119 | ~33<sup>y</sup> | ~35<sup>y</sup> | - | ~100<sup>j</sup> | [104] |
| Electrospaying of Ti-CNFs on PVDF membrane | ~125.0<sup>a</sup> | 172.0±1.8<sup>a</sup> | 190.5±3.5<sup>a</sup> | 240±5 | ~10<sup>y</sup> | ~35<sup>y</sup> | - | - | [101] |
| | ~119.0<sup>a</sup> | 170.5±3.5<sup>a</sup> | 160.1±2.6<sup>d</sup> | 300 – 400 | - | - | - | - | [101] |
| | 80±1.0<sup>d</sup> | 165±0.3<sup>d</sup> | 162.5±4.5<sup>r</sup> | 240±5 | - | - | - | - | [101] |
| | 86.5±1.5<sup>v</sup> | 151.0±1.5<sup>r</sup> | 153.5±3.5<sup>v</sup> | 240±5 | - | - | - | - | [101] |
| | 108.5±2.0<sup>v</sup> | 118.0±3.0<sup>r</sup> | 118.0±3.0<sup>r</sup> | 240±5 | - | - | - | - | [101] |
| Immobilization of TiO₂@PDA@Cu composite nanoparticles on PVDF membrane surface via chemical bonding; fluorosilanization treatment with PFDTs | 127.0±1.0<sup>a</sup> | 168.0±2.0<sup>a</sup> | 168.0±2.0<sup>a</sup> | 250 | ~13<sup>a</sup> | ~35<sup>a</sup> | - | - | [102] |
| | 70.8±3.0<sup>d</sup> | 157.1±1.5<sup>d</sup> | 157.1±1.5<sup>d</sup> | 250 | - | - | - | - | [102] |
| | 101.1±2.5<sup>i</sup> | 160.0±1.2<sup>i</sup> | 158.3±1.5<sup>i</sup> | 250 | - | - | - | - | [102] |
| | 86.6±3.5<sup>v</sup> | 152.5±0.3<sup>b</sup> | 152.5±0.3<sup>b</sup> | 250 | - | - | - | - | [102] |
| | 66.3±2.6<sup>b</sup> | 131.5±1.8<sup>b</sup> | 131.5±1.8<sup>b</sup> | 250 | - | - | - | - | [102] |
| Blending of PVDF-HFP, FA and ZnO | 135.3±1.2<sup>a</sup> | 161.4±3.4<sup>a</sup> | 161.4±3.4<sup>a</sup> | 101±11 | ~12<sup>··</sup> | ~22<sup>d</sup> | ~90<sup>y</sup> | >99.9<sup>d</sup> | [108] |
| | ~0<sup>··</sup> | 131.5±1.8<sup>b</sup> | 131.5±1.8<sup>b</sup> | 101±11 | - | - | - | - | [108] |
| Method                                      | Contact Angle (°) | DCMD (℃) | Stability (%) |
|---------------------------------------------|-------------------|----------|---------------|
| Solvent-thermal induced roughening method   | 131±2.9           | 216±29   | >99           |
| Heat-press treatment and dip-coating in a   | 380               | 700      | >99           |
| Teflon solution                             |                   |          | >99           |

- a water;   b mineral oil;   c SLS;   d SDS;   e DTAB;   f CTAB;   g measured flux after 6 h operation;   h stable flux for 12 h operation;   i 3.5 wt% NaCl feed solution contained 150 ppm DTAB;   j salt rejection;   k normalized flux (%) during 60-180 min operation with addition of 0.1 mM SDS in 35g/L NaCl feed solution;   l normalized flux (%) after 420 min operation with addition of 0–0.3 mM SDS in 35g/L NaCl feed solution;   m methanol;   n castor oil;   o decane;   p fed with 3.5 wt% NaCl for 30 operation;   q fed with 3.5 wt% NaCl contained 0.2 mM SDS for 120 h operation;   r ethanol;   s after 0.1 mM SDS was added at 120 min operation;   t after 0.4 mM SDS was added with 480 min operation;   u NaCl solution;   v Tween-20;   w soybean oil;   x dodecane;   y after 0.4 mM SDS/CTAB/Tween-20 was added at 1200 min operations;   z SDBS;   α measured flux after 26 h operation;   β vegetable oil;   γ measured flux before 450 min operation;   δ measured flux after 810 min operation;   ε surfactant contaminated saline water;   η measured flux after 60 min operation with SDS in feed solution;   ι measured flux after 360 min operation with SDS in feed solution;   ζ measured flux after 150 min with mineral oil in feed solution;   η measured flux after 420 min operation with mineral oil in feed solution;   θ glycerol;   μ EG;   ν silicone oil;   ξ toluene;   η measured flux after 1 h operation with 0.1 mM SDS in feed solution;   ζ measured flux after 90 h operation with 0.4 mM SDS in feed solution;   APTES: 3-Aminopropyl-triethoxysilane;   CA: Cellulose acetate;   CA(°): Contact angle;   CNFs: Carbon nanofibers;   CTAB: Cetyl trimethyl ammonium bromide;   Cu: Copper;   DA: Dopamine;   DTAB: Dodecyl trimethyl ammonium bromide;   EA: Epoxy acrylic;   EG: Ethylene glycol;   FAS: 1H,1H,2H,2H-perfluoroctyltriethoxysilane;   LEP: Liquid entry pressure;   PDA: Polydopamine;   PDTS: Perfluorodecyl-triethoxysilane;   PEI: Polyethyleneimine;   PFDT: Perfluorodecanethiol;   PFDTS: 1H,1H,2H,2H-perfluorodecyltriethoxysilane;   PFTS: Perfluorodecyltrimethoxysilane;   SDS: Sodium dodecyl sulfate;   SDBS: Sodium dodecyl benzene sulfonate;   SiNPs: Silica nanoparticles;   SLS: Sodium lauryl sulfate;   Ti or TiO₂: Titanium oxide;   TMOS: Tetramethyl orthosilicate;   ZnO: Zinc oxide
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