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# Membrane distillation of saline with phenolic compound using superhydrophobic PVDF membrane incorporated with $TiO_2$ nanoparticles: Separation, fouling and self-cleaning evaluation

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A B S T R A C T

Superhydrophobic polyvinylidene fluoride (PVDF) membrane was used to study membrane distillation of saline with phenolic compound. Besides addition of $TiO_2$ nanoparticles, superhydrophobic surface of PVDF membrane was created by silane grafting after phase inversion. Three types of silane, (heptadecafluoro-1,1,2,2-tetrahydrodecyl) triethoxysilane, (tridecafluro-1,1,2,2-tetrahydrooctyl) triethoxysilane and (3,3,3-trifluoropropyl) trimethoxysilane were used in this work. All the membranes achieved superhydrophobicity (water contact angle > 150°), but pore size and porosity were significantly reduced by increasing silane concentration. Lithium chloride (LiCl, 2 wt%) was used to improve superhydrophobic PVDF/$TiO_2$ membranes. Membranes were further evaluated in terms of separation performance, fouling tendency and self-cleaning properties. In membrane distillation, all membranes retained 99.9% of the phenolic content in feed (100 g/L of gallic acid). However, superhydrophobic PVDF/$TiO_2$ membranes modified using (tridecafluro-1,1,2,2-tetrahydrooctyl) triethoxysilane (1 mL silane: 50 mL ethanol) and 2 wt% of LiCl was the only membrane that maintained the permeate flux near to 6 $kg.m^{-2}.h^{-1}$ without much flux decline after 8 h of operation. It also retained 99.9% of sodium chloride (NaCl) and gallic acid simultaneously at a higher feed temperature of 70 °C. Although some of these modified membranes were fouled by gallic acid, they could be easily cleaned under UV radiation.

## 1. Introduction

Phenolic compounds from natural plant sources are among popular ingredients in food, beverage and nutraceutical industries due to increasing health concerns. However, excessive phenolic content of wastewater produced from food, beverage and nutraceutical industries can affect the ecosystem adversely [1]. Leather manufacturing plants [2] and gallic acid processing plants [3,4] also release phenolic pollutants such as gallic acid into various water systems. Similar to other pollutants, the presence of phenolic compounds in different water sources causes membrane fouling which reduces separation performance and economics of the fouled membrane [5,6].

In the recent years, membrane fouling in membrane distillation has been extensively studied. This is because membrane distillation is not only useful for desalination, this technology is also applicable in producing water treatment, groundwater purification, juice concentration and more [7]. Unlike other pressure-driven filtration processes, membrane distillation is conventionally operated using a hydrophobic macroporous membrane which acts as the barrier between hot feed and cold permeate. The membrane only allows vapor transportation driven by a vapor pressure difference into cold solution. Similar to other filtration system, various compounds accumulate on the membrane surface after long operation hours. Fouling causes surface wetting and pore plugging, resulting a drop in permeate flux. The effects of different foulants such as $CaSO_4$, $CaCO_3$, humic acid, alginate acid and bovine serum albumin have been studied [8,9]. The organic foulants promoted accumulation of NaCl and $CaSO_4$ on the macroporous and hydrophobic polytetrafluoroethylene (PTFE) membrane [10]. Organic foulants even penetrated into PTFE membrane and migrated into the permeate side of membrane. When membrane distillation was applied to treat table olive mill wastewater [6,11–13], membrane fouling was reported as well. PVDF membranes with large pore size and great hydrophobicity offered high permeate flux, but pore plugging could diminish the permeate flux in long run. Commercial PTFE membrane with smaller pore size was reported to be more resistant to irreversible fouling [12]. Frequent cleaning on membrane could help to recover the permeate flux of a membrane distillation system used to concentrate orange juice [14]. In the coupled system of osmotic distillation and membrane distillation, pomegranate juice [15] and red grape juice [16] were even concentrated by heating the feed solution and cooling the hypertonic solution

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**Table 1**
The characteristics of PVDF membranes prepared using the varied ratio of silane to ethanol ratio in post-treatment.

| Membrane | Water contact angle (°) | Mean pore size (μm) | Porosity (%) | $LEP_w$ (bar) | Thickness (μm) |
|---|---|---|---|---|---|
| P-$TiO_2$[a] | 112.3 ± 1.4 | 0.44 ± 0.02 | 46.0 ± 0.8[b] | 0.64 ± 0.03 | 247.0 ± 3.8 |
| P-SI-100 | 151.2 ± 0.5 | 0.42 ± 0.07 | 40.6 ± 1.0 | 0.65 ± 0.01 | 204.4 ± 1.7 |
| P-SII-100 | 158.7 ± 1.5 | 0.44 ± 0.04 | 39.5 ± 0.4 | 0.67 ± 0.02 | 198.2 ± 1.5 |
| P-SIII-100 | 163.2 ± 1.6 | 0.39 ± 0.04 | 40.7 ± 0.6 | 0.67 ± 0.02 | 200.5 ± 5.0 |
| P-SII-50 | 162.7 ± 0.4 | 0.40 ± 0.02 | 38.8 ± 0.8 | 0.67 ± 0.02 | 200.7 ± 1.2 |
| P-SIII-50 | 163.2 ± 1.1 | 0.35 ± 0.02 | 40.4 ± 0.6 | 0.59 ± 0.01 | 195.5 ± 2.5 |
| P-SII-25 | 165.3 ± 1.0 | 0.34 ± 0.02 | 40.2 ± 0.8 | 0.70 ± 0.03 | 201.5 ± 1.3 |

[a] The membrane (P-pure) was studied in our previous work [26].
[b] The porosity was studied using ethanol as the tested solution.

simultaneously. Although permeate flux of this coupled system was higher than osmotic distillation, similar flux decline pattern was observed due to membrane fouling and wetting [15].

In order to prevent membrane wetting and the subsequent formation of liquid filled pores with great mass transfer resistance, hydrophobic membranes made of polytetrafluoroethylene (PTFE) and polyvinylidene fluoride (PVDF) were commonly applied in membrane distillation [17]. PTFE possesses great hydrophobicity, thermal and chemical resistance, while PVDF can dissolves in common solvents for phase inversion processing. Membrane researchers further modified these membranes into superhydrophobic membranes to avoid membrane fouling by reducing direct contact between foulant and membrane surface at Cassie-Baxter state. The coating of $TiO_2$ and $SiO_2$ nanoparticles on membrane surface induced formation of hierarchical structure for superhydrophobic surface [18,19]. However, permeate flux of these modified membranes was slightly lower than the unmodified membrane unless fouling occurred on the unmodified membrane. Such coating hindrance could be avoided using electrospun coating [20] or $CF_4$ plasma modification [21]. Up to date, electrospinning is more popular than other methods such as phase inversion in the synthesis of superhydrophobic membranes. A higher permeate flux was commonly reported after incorporating carbon nanotubes [22] or inorganic particles grafted with fluorocarbon group into the electrospun membranes [23]. This is because electrospun membranes showed excellent porosity (> 85%) for effective mass transfer in air filled pores [24]. Using the traditional phase inversion techniques, fabrication of superhydrophobic membranes was rarely reported to our best knowledge. Most of the researchers focused on pore forming agents to improve the porosity of hollow fibers [25]. Blending superhydrophobic silica nanoparticles into PVDF membrane which was prepared via phase inversion resulted in less satisfactory surface wettability in term of water contact angle compared to other works [20]. Blending hydrophobic nanoparticles into PVDF membrane caused the formation of dense structure in phase inversion, limiting mass transfer as reported in our previous works [5]. Although the post-treatment of PVDF membrane blended with $TiO_2$ showed less reduction in porosity, superhydrophobicity was not successfully attained in our previous work [5].

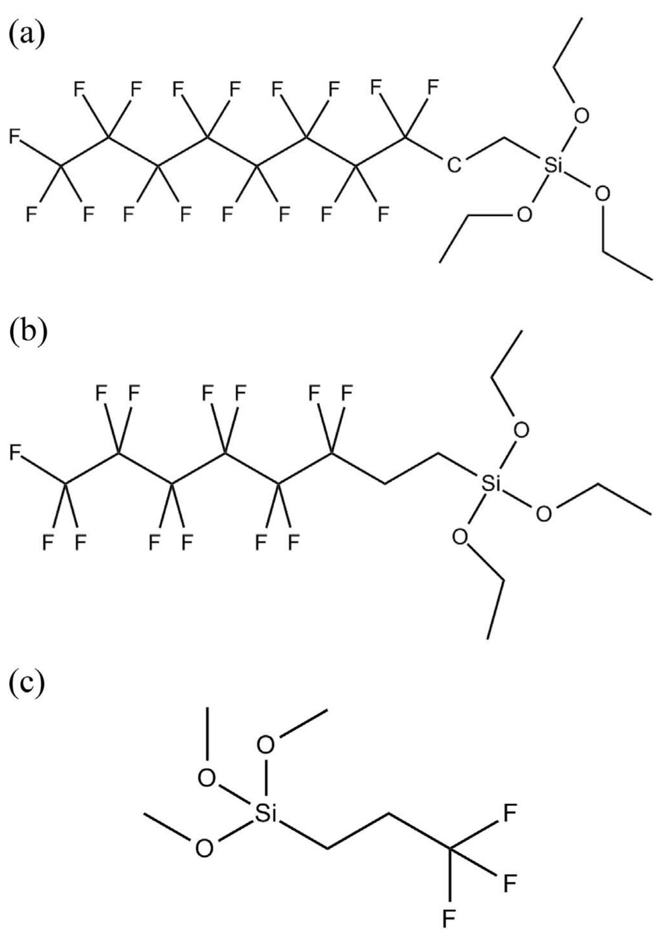


**Fig. 1.** Chemical structure of (a) type I silane ((heptadecafluoro-1,1,2,2-tetrahydrodecyl) triethoxysilane), (b) type II silane ((tridecafluro-1,1,2,2-tetrahydrooctyl) triethoxysilane) and (c) type III silane ((3,3,3-trifluoropropyl) trimethoxysilane).

In this work, superhydrophobic PVDF membrane was successfully fabricated and used to study membrane distillation of saline with phenolic compound. Besides the addition of $TiO_2$ nanoparticles, superhydrophobic surface of PVDF membrane was created by silane grafting after the phase inversion. The influence of silane chemistry on membrane characteristics and separation performance of superhydrophobic membrane in membrane distillation of saline containing phenolic compound were investigated. Pore former, LiCl was also subsequently used to improve the separation performance of PVDF/$TiO_2$ membrane. More importantly, the fouling and self-cleaning of this superhydrophobic membrane were further studied.

## 2. Materials and methods

### 2.1. Materials

PVDF powder (Solef® 6010 PVDF) was purchased from Solvay Solexis (France) and must be dried at 100 °C before being used. *N*-methyl-2-pyrrolidone (NMP) (> 99.5%) supplied by Merck (Darmstadt, Germany) was used as the polymer solvent. $TiO_2$ nanoparticles (21 nm primary particle size, > 99.5% trace metal basis) for dope solution were supplied by Sigma-Aldrich (St. Louis. MO, USA). The other additives of dope solution such as ortho-phosphoric acid ($H_3PO_4$) (> 85%), lithium chloride (LiCl) and acetone were obtained from Merck (Darmstadt, Germany). Ethanol acquired from Merck (> 99.9%, Darmstadt, Germany) was used as the coagulation bath and the silanation solvent. The silanes used in the post modification were detonated as type I silane ((heptadecafluoro-1,1,2,2-tetrahydrodecyl) triethoxysilane), type II silane ((tridecafluro-1,1,2,2-tetrahydrooctyl) triethoxysilane) and type III silane ((3,3,3-trifluoropropyl)

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