

Bio-Inspired Surface Engineering of Low-Content PVDF/TA Janus Membranes for Surfactant-Stabilized Water-in-Oil Emulsion Separation

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Research Article

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2 Janus Membranes for Surfactant-Stabilized Water-in-Oil
3 Emulsion Separation

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Abstract

In contrast to conventional tannic acid (TA) surface modifications, which often exhibit poor adhesion on hydrophobic polyvinylidene fluoride (PVDF) substrates. This study demonstrates, for the first time, a simple and rapid vacuum-filtration approach to fabricate and modify environmentally friendly Janus PVDF membranes for water-in-oil emulsion separation. These membranes were initially prepared via phase inversion and subsequently modified on a single surface with TA and NaIO₄ to achieve asymmetric wettability. The TA functional groups facilitated strong polar and hydrogen-bonding interactions, enabling efficient trapping and coalescence of water droplets while allowing preferential oil permeation through the hydrocarbon-affinitive pores. The fabricated membranes achieved separation efficiencies of beyond 99% for water-in-hexane and water-in-toluene emulsions, and water flux of 1545 and 1875 L m⁻² h⁻¹, respectively. It also maintained stable performance over seven consecutive filtration cycles, demonstrating excellent recyclability. In addition, the fabrication process is environmentally friendly and scalable, underscoring the strong potential of these Janus PVDF membranes for sustainable industrial water-in-oil separation applications.

Keywords: Janus membrane, phase inversion, polyvinylidene fluoride, water-in-oil emulsion, separation efficiency, water flux.

1. Introduction.

In everyday human life, industrial operations, textile wastewater containing toxic dyes requires efficient treatment prior to discharge, as many dyes are carcinogenic or suspected carcinogens [1]. Moreover, unintended crude oil and petrochemical leaks have increased, releasing pollutants that significantly harm the environment and cause financial losses [2, 3]. Recently, natural resources have been explored as coagulants due to their advantages, such as lower cost, renewable availability, and reduced sludge volume [4]. At the same time, global water consumption has risen substantially [5], and wastewater effluents containing organic and inorganic contaminants are expected to double compared to 2010 levels by 2025 [6]. The disposal of waste oil, paint, and coating barrels (WOPCBs) after use remains a serious social and environmental problem [7]. Among treatment methods, membrane filtration has emerged as one of

the most competitive technologies for separating water/oil mixtures, particularly highly emulsified, surfactant-stabilized emulsions, owing to its simple operation and excellent separation efficiency [8–11]. Recent studies have also demonstrated that polymer-blend composite membranes can provide high permeability and excellent oil rejection for surfactant- stabilized oil-in-water emulsions [12]. Filtration membranes are typically modified to have opposite wettability to water and oil in order to increase separation efficiency [13-15]. Making membranes that are hydrophobic, oleophilic and non-wettable to water but wettable to oils, is comparatively simple since water has a substantially higher surface tension than oils [16-19]. These hydrophobic, oleophilic and wettability membranes allows them to capture water droplets from the oil flow. However, there is a problem with oil fouling for these membranes particularly for more viscous oils, which can cause a significant drop in flux. Hydrophilic-oleophobic membranes are able to reject oils and maintain their surfaces clean, in contrast to hydrophobic-oleophilic membranes. However, oleophobic surfaces tend to be more resistant to water because water has a significantly higher surface tension than oils, making the hydrophilic-oleophobic combination extremely difficult to achieve. Designing heterogeneous molecular architectures with both hydrophilic and fluorinated moieties is essential for producing hydrophilic-oleophobic surfaces [20-23]. In contrast, the hydrophilic moieties create the hydrophilicity through a favorable interaction with water, and the fluorochains reject oils. It was attached on the membrane surface because it also facilitates the anti-oil fouling function by causing water to replace oils, and under-oil superhydrophilicity can be used as an alternative to the hydrophilic–oleophobic property for oil–water separation. Additionally, it can facilitate the demulsification of water-in-oil emulsions due to its stronger affinity for water than for oils [24-26]. One clear benefit of under-oil superhydrophilicity over hydrophilic–oleophobic properties is that it does not require the inclusion of a fluorinated moiety. While cellulose-based porous materials [27, 28], desert sand [29], fumed silica-coated metal felt [30], metal organic frame (MOF)-coated stainless-steel mesh [31], poly 2-dimethyl-amino ethyl methacrylate coated fabric [32], and electro-spun membrane of deacetylated cellulose acetate appear to be among the under-oil super-hydrophilic materials [33]. Achieving the under-oil superhydrophilicity remains difficult because it is thermodynamically unfavorable to substitute a solid–water contact (higher interfacial tension) for the solid–oil interface (lower interfacial tension). Herin, it was reported about a simple

efficient strategy for imparting super-hydrophilicity to a phase inverted polyvinylidene fluoride by coating with TA and addition of strong oxidizing agent, NaIO₄. It was chosen as the integrated component since TA is a naturally occurring polyphenol that has long been recognized for its tanning qualities in the leather and ink industries. The remarkable ability of TA for binding to any material that resembles mussel byssus on its surface, enhancing the hydrophilicity and antifouling qualities of membrane. It is primarily responsible for its allure in more contemporary fields of membrane research [34-36]. It allows the big phenolic hydroxyl groups to engage with hydrophobic materials via the creation of hydrogen bonds with crosslinking and the most important hydrophobic interaction. The surface binding capacity is varied with TA [37, 38]. However, the inherent oleophilicity and hydrophobicity of polymer membranes can be readily changed by cross-linking TA in the hydrophobic area using straightforward modification procedures. Taking advantage of asymmetric Janus membranes in the separation efficiency and flux enhancement [39-43].

This study demonstrate, a simple and rapid vacuum-filtration approach to fabricate and modify environmentally friendly Janus PVDF membranes for water-in-oil emulsion separation. Initially, the membrane was fabricated through phase inversion, followed by TA/ NaIO₄ coating on one face of the membrane. Various mixtures of water-in-oil emulsions were then effectively separated using the fabricated membrane. The TA coating on the membrane surface mainly traps and coalesces dispersed water droplets in the emulsion through polar and hydrogen-bonding interactions. Therefore, it prevents water penetration, while oil readily permeates due to the hydrocarbon affinity with the oil-wetted pores. For surfactant stabilized water-in-hexane and water-in-toluene showed separation efficiency of beyond 99% in both conditions. In addition, the flux is 1545 and 1875 L m⁻² h⁻¹, respectively, under water-in-hexane and water-in-toluene. It exhibited excellent long-term stability and recyclability, highlighting its strong potential for industrial applications.

2. Experimental

2.1 Materials

Polyvinylidene fluoride (PVDF; MW 90,000 mg/mol), powder purchased from Guangdong Weng Jiang Chemical Reagent Co., Ltd., Tannic acid (TA) was purchased from Belgium. Sodium metaperiodate (NaIO₄) of 98% purity and Span-80 were made in USA. Honeywell Riedel-de Haen provided toluene (purity ≥99.7%) and methanol

(purity $\geq 99.8\%$). n-octane was provided by Dae-Jung reagents chemicals (purity $\geq 97\%$), while xylene (purity $\geq 96\%$), and hexane (purity $\geq 99\%$) were provided by Sigma-Aldrich xylene. Other oils include soy, purchased from Ding Guan company, mustard purchased from Taishan enterprise Co., Ltd and coco oil, supplied by Fssai company.

2.2 Characterization

A scanning electron microscope (SEM; JSM-IT100, JEOL Ltd., Japan,) was used to observe the surface morphologies of the asymmetric fabricated membrane. Energy-dispersive X-ray spectroscopy (EDS; 51-ADD0076, INCA x-act, England,) was used to know about the chemical composition of the membrane. Water content was analyzed by Karl Fischer moisture titrator (MKV-710, KEM). The oil concentration in the permeate and feed was obtained by using a wastewater treatment photometer (HI 839800, HANNA Instruments) to measure the chemical oxygen demand. Fourier Transform Infrared Spectroscopy (FTIR; Nicolet iS5, Thermo Scientific, USA), and water contact angle (WCA; OSA100, Ningbo NB Scientific Instruments Co Ltd., China).

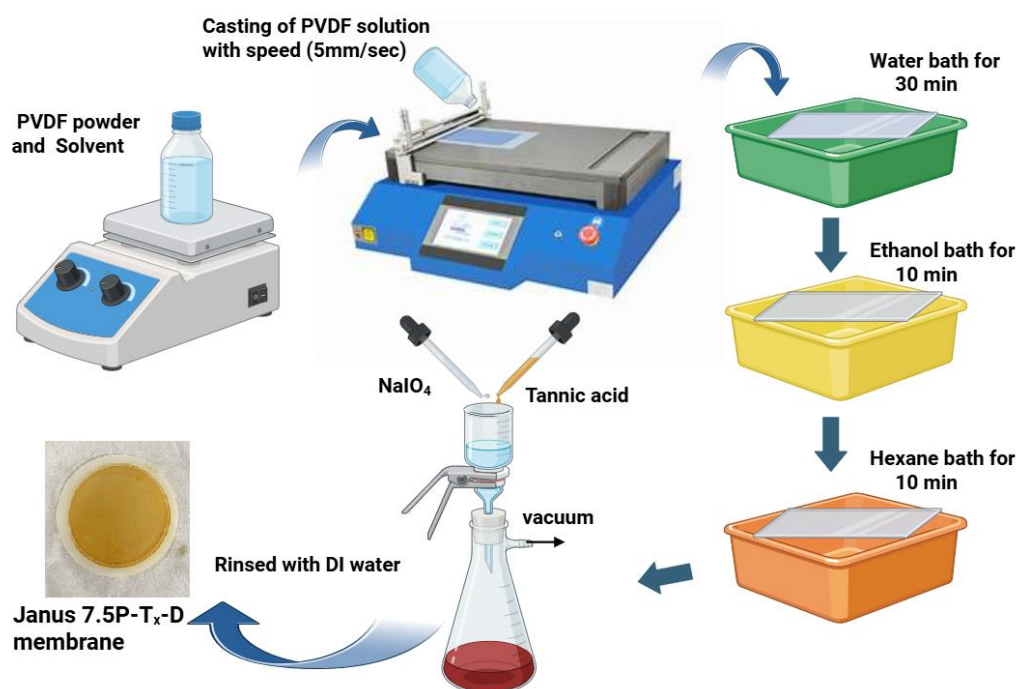


Figure 1. Schematic for the fabrication of Janus 7.5P-T_x-D membrane

2.3 Fabrication of membrane

The poly vinylidene fluoride (PVDF) membranes were prepared using two different solvents i.e. N-methyl-2-pyrrolidone (NMP) and dimethylacetamide (DMAc) to assess solvent influence on membrane formation at low polymer concentrations. PVDF powders content of 5, 7.5, 10, 15, and 20 wt. % were dissolved in the respective solvents, with the solvent fraction adjusted accordingly to maintain total composition at 100 wt. %. Each dope solution was magnetically stirred for 6 h to achieve complete dissolution and then degassed for an hour to remove air bubbles. The homogeneous solutions were casted on a nonwoven support fixed on a glass plate at a constant casting speed of 5 mm s⁻¹, and the nascent films were immediately immersed in a deionized-water coagulation bath for 30 min to induce phase inversion. Subsequently, the membranes were treated sequentially in ethanol and n-hexane baths (10 min each) to enhance solvent exchange and structural stabilization, followed by drying at room temperature. It was observed that membranes fabricated from NMP solutions containing less than 15wt. % PVDF exhibited severe shrinkage and mechanical instability. This behavior was attributed to instantaneous demixing during immersion in the nonsolvent bath, which generated large macro voids and weak membrane structures. In contrast, stable membranes could be formed at higher polymer concentrations (15 and 20 wt.%), where the slower phase inversion reduced structural defects. This phenomenon arises from the relatively lower polymer-solvent affinity of NMP compared to DMAc. Moreover, the stronger interaction between DMAc and the nonsolvent (water) accelerates phase inversion, thereby promoting uniform and coherent membrane formation even at lower PVDF contents Wu et al. [44].

So, for all the above content the PVDF membrane fabrication was possible. It owing to delayed demixing behavior that slowed phase separation and improved gel strength during membrane solidification, yielding uniform and mechanically stable films.

2.4 Preparation of TA and NaIO₄ aqueous solution

Different TA concentration was prepared by dissolving certain amount of TA powder in ethanol. While, NaIO₄ was dissolved in the deionized (D.I) water with a fixed concentration of 0.02 g mL⁻¹, as listed below (table 1). Figure S2 displays the average thickness of TA coating layer formed on the various Janus 7.5P-T_x-D

membrane

Table 1. Concentration of TA

Janus Membrane	Concentration of TA (g mL ⁻¹)
7.5P-T _{0.16} -D	0.16
7.5P-T _{0.20} -D	0.20
7.5P-T _{0.24} -D	0.24
7.5P-T _{0.28} -D	0.28

2.5. Preparation of Janus7.5P-T_x-D membranes

The PVDF membranes fabricated under the aforementioned conditions yielded seven distinct samples with varying pore structures. Among them, the composition designated as 7.5P-D (7.5 wt. % PVDF in DMAc) was identified as optimal, exhibiting a uniform pore size of approximately 0.2 μm. Other formulations were deemed unsuitable for subsequent surface modification due to poor structural uniformity (Figure S1). Prior to modification, the selected membrane was cleaned by immersion in ethanol for 2 min to remove residual contaminants and surface debris, followed by mounting in a vacuum filtration unit and pre-wetting with ethanol to ensure complete pore saturation. For the single-step surface modification, 3.5 mL of TA solution was introduced into the filtration cell and allowed to permeate through the membrane, with the filtration time varying according to concentration of TA (Table 2). Once the TA solution had completely passed through and deposited on the upper surface, 1.5 mL of NaIO₄ solution was added and allowed to react for 4 min to oxidatively crosslink the TA layer. This reaction formed a stable polyphenolic coating on the membrane's upper face. The modified membrane was then rinsed thoroughly with DI water to remove unreacted species and by-products. Through this TA/oxidant-assisted treatment, a Janus membrane was obtained, featuring a hydrophilic TA-coated top layer and an intrinsically hydrophobic PVDF base. Finally, the membrane was dried at room temperature and stored for subsequent characterization and separation testing.

Table 2. The time for TA modification

Janus membrane	Ratio C _T : C _{Na}	Time of TA modification (min)
7.5P-T _{0.16} -D	08:1	03
7.5P-T _{0.20} -D	10:1	04
7.5P-T _{0.24} -D	12:1	05
7.5P-T _{0.28} -D	14:1	06

2.6 Water-in-oil emulsion separation.

A surfactant stabilized water-in-oil (W/O) emulsion was prepared as a feed solution to evaluate the separation performance of the fabricated Janus membranes. The emulsion consisted of water, oil and Span-80 in a mass ratio of 1:50:0.1, and was homogenized by sonication for 90 min followed by overnight stirring to ensure uniform dispersion of water droplets within the oil phase. The separation experiment was conducted using a dead-end vacuum filtration setup with an effective membrane area of $1.9 \times 10^{-4} \text{ m}^2$ under a mild pressure of 800 mbar. During experiment, the Janus membrane was mounted such that its TA-modified (hydrophilic) surface faced the emulsion feed. Prior to every separation cycle, the membrane was pre-wetted with deionized water to establish a stable hydration layer that enhances anti-fouling and selective wettability. The prepared W/O emulsion was then poured into the feed reservoir, and the permeate was collected for compositional analysis to quantify any residual water content. The permeation flux (J) and separation efficiency (R) were calculated using equations. (1) and (2), respectively, as adopted from Ong et al. [45] and Wang et al. [46]:

$$J(\text{Lm}^{-2} \text{ h}^{-1}) = \frac{V \times \Delta t}{A} \dots \text{Eq. (1)}$$

Where V is the volume of permeance (L), A represents effective membrane area (m^2) and Δt is for the permeation time (h).

$$R(\%) = 1 - \frac{C_f}{C_o} \times 100 \dots \text{Eq. (2)}$$

Here, R denotes the water rejection coefficient, while C_f and C_o are the concentration of water in the permeation and emulsion feed.



Figure 2. The TA coating of Janus 7.5P-T_{0.24}-D membrane.

3. Results and discussion

3.1 Surface characterization of Janus 7.5P-T_x-D membranes

The surface morphology of the synthesized membranes was examined using SEM, as illustrated in figure 3. The pristine 7.5P-D membrane exhibited a relatively smooth and porous surface, with no discernible optical or particulate structures observed on its upper face, indicating a uniform phase-inverted morphology. Upon modification with TA and NaIO₄, notable morphological changes were observed, which varied depending on the TA concentration. At the lowest TA concentration of 0.16 g mL⁻¹, the upper surface of the Janus membrane appeared slightly rough with fiber-like textures. The limited deposition at this concentration resulted in insufficient coating coverage, leaving the pore structure largely unaffected and thus providing only modest selectivity enhancement. As the TA concentration increased, a progressive alteration of the surface topography was evident. The TA and NaIO₄ treatment induced surface densification and partial pore constriction, consistent with the interfacial oxidation and crosslinking behavior typically reported for plant polyphenol-derived coatings. This phenomenon parallels the well-known densification observed in polydopamine (PDA) coatings [47-49]. At 0.20 g mL⁻¹, the membrane, nominated as 7.5P-T_{0.20}-D, surface displayed a denser TA layer characterized by visible crack-like features, suggesting heavy deposition and partial pore coverage. The enhanced surface compactness improved selectivity, although this led to a reduction in permeation flux. When the TA concentration was further increased to 0.24 g mL⁻¹, a uniform and continuous coating layer was formed. It displays interconnected crack-like structures indicative of stable crosslinking for preparing membrane, nominated as 7.5P-T_{0.24}-D. The pores remained partially open,

allowing water permeation while maintaining structural stability thus representing an optimal balance between flux and selectivity. However, at the highest TA concentration of 0.28 g mL⁻¹, excessive deposition produced a brittle, thick coating layer membrane, nominated as 7.5P-T_{0.28}-D, with collapsed regions and wide crack openings, signifying over-oxidation and pore blockage. The bottom (uncoated) surface of the membrane, in contrast, retained its original morphology across all samples, confirming that the modification was confined to the upper face. The successful TA coating on the 7.5P-T_{0.24}-D membrane was further confirmed by EDS mapping, as shown in figure 4. A homogeneous distribution of oxygen (O) was detected across the modified surface, whereas the pristine membrane lacked this element. The elevated O content originated from the hydroxyl and quinone functional groups of TA, as reported by Liu et al. [50]. During oxidation, the phenolic -OH groups in TA were converted by NaIO₄ into o-quinone or p-quinone species containing C=O functionalities, thereby confirming the successful formation of a stable TA-based coating via the proposed one-pot modification method.

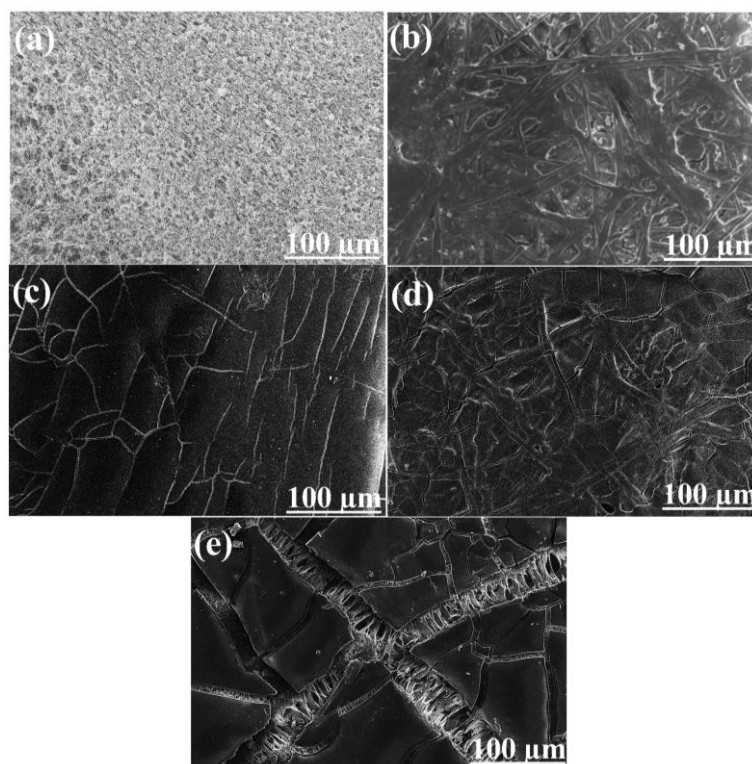


Figure 3. SEM image upper face of pristine and modified membrane (a) 7.5P-D (b) 7.5P-T_{0.16}-D (c) 7.5P-T_{0.20}-D (d) 7.5P-T_{0.24}-D (e) 7.5P-T_{0.28}-D

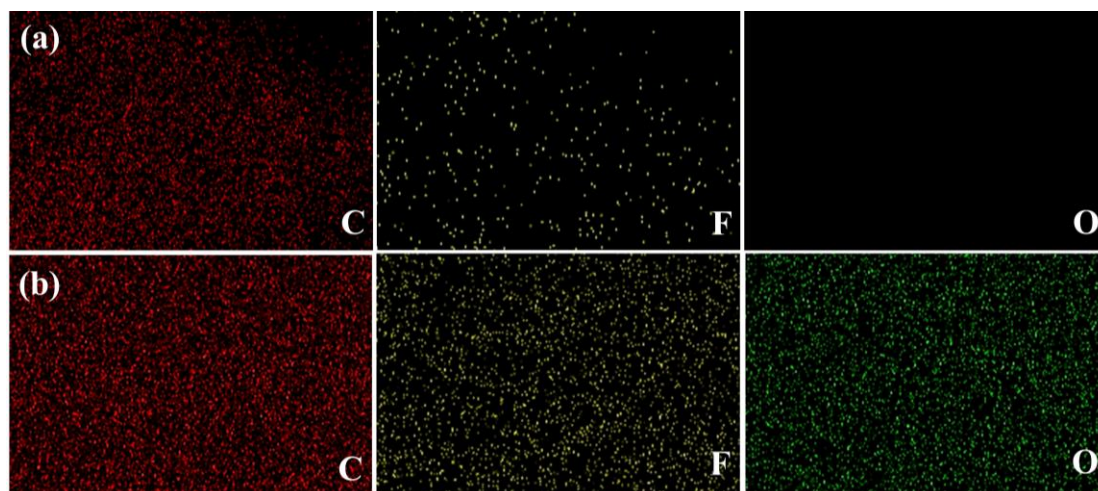


Figure 4. Elemental composition of the (a) Pristine of 7.5P-D and (b) Janus membrane of 7.5P-T_{0.24}-D

3.2 FTIR of the Janus 7.5P-T_{0.24}-D membrane

The chemical modification of the fabricated membrane support by TA was further confirmed by FTIR spectroscopy. The spectra of the pristine membrane and the modified 7.5P-T_{0.24}-D membrane are presented in figure 5. The modified membrane exhibited additional absorption features corresponding to the TA layer, including a broad stretching band at 3200–3500 cm⁻¹, which was notably observed in the spectra of all TA-modified membrane upper surfaces (see figure S4). This broad band is attributed to the –OH functional groups arising from the abundant phenolic hydroxyls present in TA [51-52]. In contrast, -OH broad stretching band was absent in the pristine 7.5P-D membrane. Furthermore, the FTIR spectrum of the 7.5P-T_{0.24}-D membrane displayed new absorption peaks associated with C=O stretching near 1820 cm⁻¹ and aromatic C=C stretching around 1680 cm⁻¹ [53-54], confirming the successful surface modification by TA. FTIR spectra of all other used concentrations are given in figure S3.

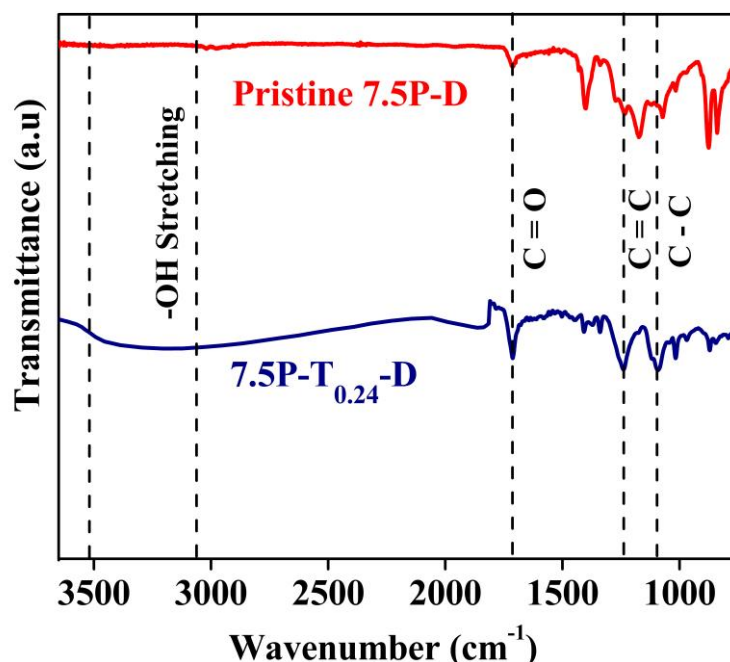


Figure 5. FTIR spectra of both the upper face of pristine 7.5P-D and TA modified 7.5P-T_{0.24}-D membrane

3.3 Janus membrane's wettability

The as-prepared membrane demonstrates dual characteristics similar to those of a lotus leaf. One surface is hydrophobic, while the other is hydrophilic. The pristine PVDF membrane exhibited strong hydrophobic. However, the WCA significantly decreased from 97.8° to 15.5° after depositing TA at different concentrations on the upper surface of the fabricated membranes. Remarkably, the WCA further dropped close to zero within just a minute. This dramatic decrease is attributed to the abundance of hydrophilic carboxyl groups introduced by the TA coating, which effectively enhanced the surface wettability and selectivity of the membrane, is consistent with the study of Wu, et al. [38]. Thus, a nature-inspired Janus membrane was successfully realized through this rapid modification method.

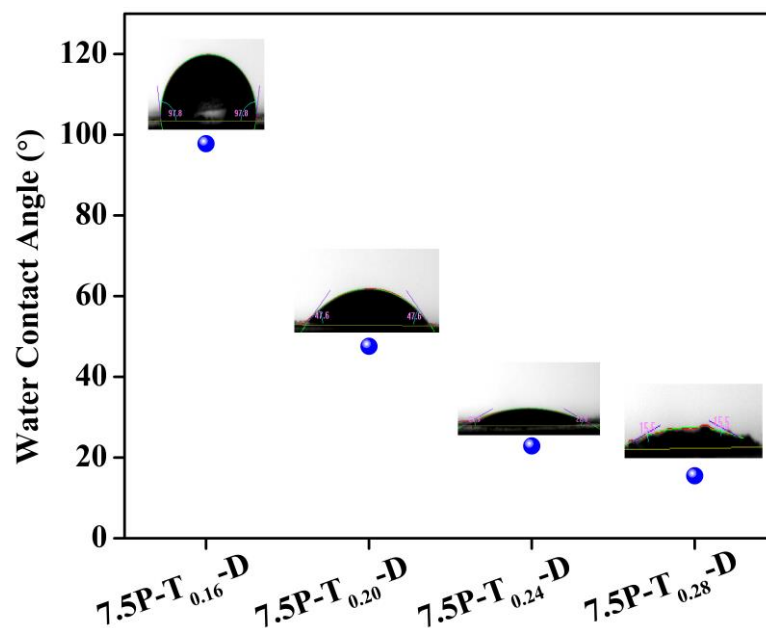


Figure 6. Water contact angle of the modified Janus 7.5P-T_x-D membrane with different concentration of TA

3.4 Impact of TA on the performance of the membrane

The practical applicability of the as-fabricated Janus membranes was evaluated through separation tests using membranes modified with varying concentrations of TA and NaIO₄. As shown in Figure 7, the separation efficiency increased with rising TA concentration, attributed to the abundant polar and hydroxyl functional groups that promoted strong hydrogen bonding and polar interactions, effectively trapping and coalescing water droplets while allowing oil to permeate. Consistent with the earlier characterization results, the enhanced hydrophilicity of the TA-coated surface led to a progressive increase in water permeability, with the flux rising from 868 to 1875 kg m⁻² h⁻¹ as the TA concentration increased. This trend indicates that moderate TA deposition improved surface wettability and transport, whereas excessive coating thickness and pore narrowing partially restricted permeability. The optimized 7.5P-T_{0.24}-D membrane demonstrated a favorable balance between flux and selectivity, outperforming several previously reported membranes for w/o emulsion separation, as shown in table 3.

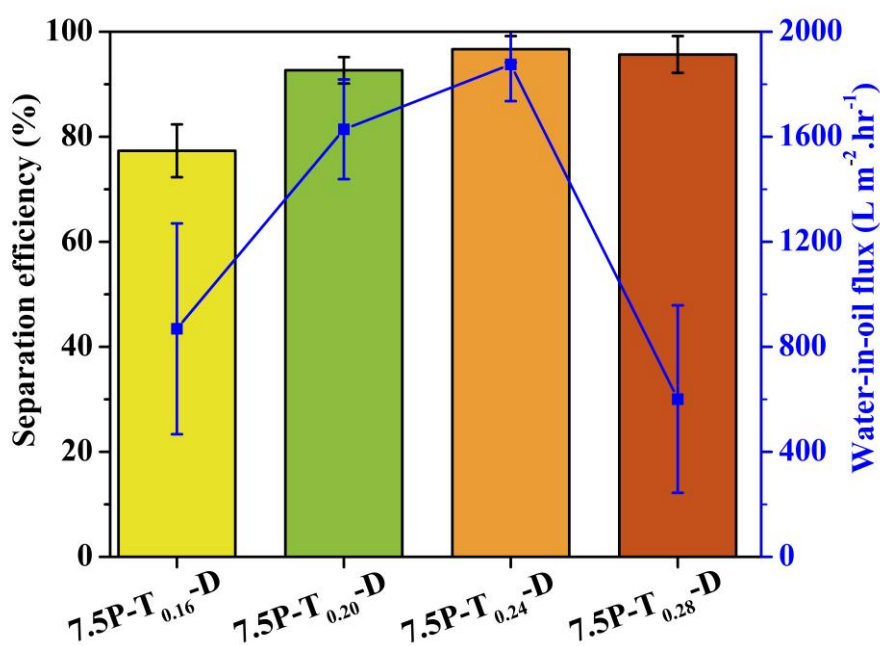


Figure 7. Performance of modified Janus 7.5P-T_x-D membrane in water in toluene

Table 3. Comparison of the Janus membrane to the previously reported membranes used for water-in-oil emulsion separation

Membrane	Materials	Emulsion	Surfactant	Water content	Separation Pressure	Separation efficiency	Flux (Lm ⁻² h ⁻¹)	Ref
Janus dCA/PVDF	CA/PVDF	Water-in-diesel	Span 80	1%	0.1 MPa	98.9 %	525	26
Au@ZIF-8 @PAN-TD	PAN, ZIF-8	Water-in-hexane	Span 80	1%	Gravity	95.6 %	650	54
PTFE@ZIF-8	PTFE, ZIF-8	Water-in-hexane	Span 80	1%	0.08 MPa	99.9 %	513	56
Agar/PVA	PVA, agar	Water-in-diesel	Tween 80	1/50	0.01 MPa	97.2 %	890	57
PVDF/SA	PVDF, stearic acid	Water-in-diesel	Span 80	1/50	0.05 MPa	99.9 %	200	58

Membrane	Materials	Emulsion	Surfactant	Water content	Separation Pressure	Separation efficiency	Flux ($\text{Lm}^{-2} \text{h}^{-1}$)	Ref
MTS@SH-SiO ₂	Methyl trichlorosilane (MTS), SiO ₂	Water-in-hexane	Span 80	1%	0.01 MPa	99.9 %	1500	59
Poly propylene	TA/DETA co-deposition	Water-in-hexane	Span 80	1%	0.01MPa	98 %	<1200	60
PVDF	PVDF/SA/CC	Water-in-hexane	Span 80	1/50	0.08 MPa	99.1 %	115	61
PVDF	PVDF/GO	Water-in-hexane	Span 80	1%	Gravity	>98 %	200	62
7.5P-T _{0.24} -D	PVDF/TA/NaIO ₄	Water-in-toluene	Span 80	2%	0.08 MPa	99.7 %	1875	This work
		Water-in-hexane				99.8 %	1545	

321

322 3.5 Various water in oil emulsion separation.

323 The separation performance of the optimized 7.5P-T_{0.24}-D Janus membrane was
324 further examined using various light oil systems to assess its versatility. The
325 TA-coated surface effectively trapped and coalesced dispersed water droplets through
326 strong polar and hydrogen-bonding interactions during the separation of water-in-oil
327 emulsions, preventing their passage while allowing the continuous oil phase to
328 permeate. The oil readily wetted the hydrophobic pores, creating a high water-entry
329 pressure that exceeded the applied transmembrane pressure, thereby ensuring
330 selective oil permeation. As shown in figure 8, the membrane exhibited excellent
331 separation efficiencies of 99.8, 99.7, 95.4, 92.1, 82.4, 79.7 and 78.2% for

water-in-hexane, water-in-toluene, water-in-octane, water-in-xylene, water-in-soybean oil, water-in-mustard oil, and water-in-coconut oil, respectively. The corresponding permeation fluxes were 1545, 1845, 1344, 1125, 339, 281 and 170 L m⁻² h⁻¹. The higher flux observed for toluene was consistent with the influence of oil density and viscosity reported by Hu et al., although hexane has the lowest viscosity [63]. The observed flux variations were primarily governed by the physicochemical properties of the oils, particularly viscosity and density differences.

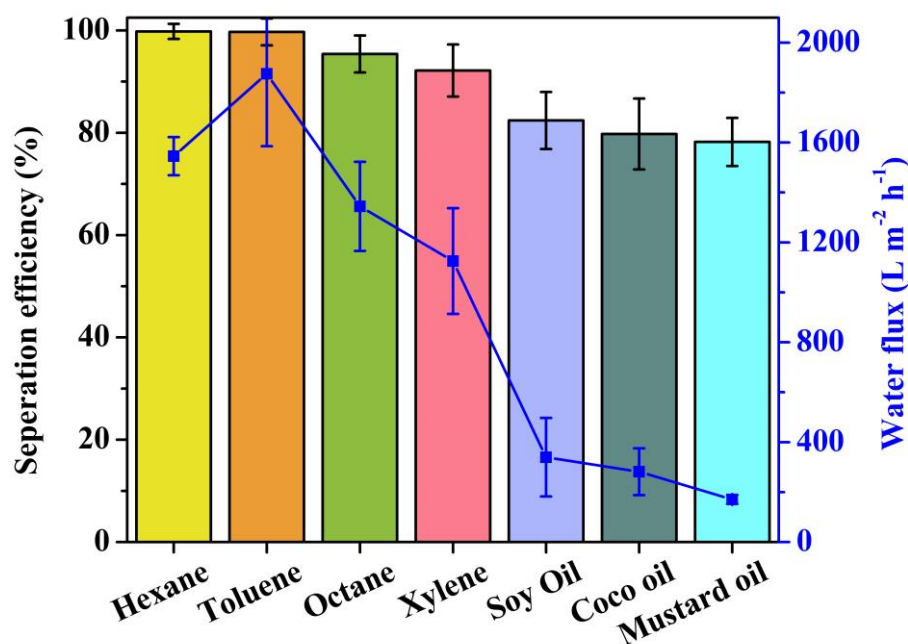


Figure 8. Performance of various oils with Janus 7.5P-T_{0.24}-D membrane

3.6 Stability of the Janus membrane.

The membrane maintained stable performance over seven consecutive separation cycles when tested with hexane and toluene emulsions, consistently achieving high separation efficiency and flux. This excellent reusability highlights the strong adhesion of the TA coating and the structural robustness of the Janus membrane, confirming its potential for long-term and practical water-in-oil emulsion separation applications.

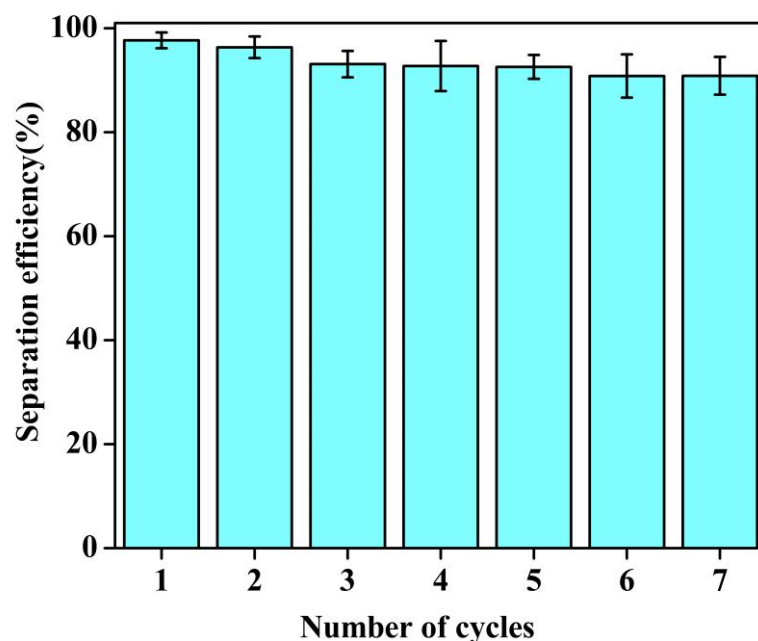


Figure 9. Cycle test for water-in-oil emulsions with Janus 7.5P-T_{0.24}-D membrane

3.7 Photographs of water-in-oil emulsions before and after separation using the Janus 7.5P-T_{0.24}-D membrane

In figure 10, several surfactant-stabilized water-in-oil emulsions are visually compared before and after separation using Janus membrane 7.5P-T_{0.24}-D. Each pair of bottles depicts the transparent permeate (right) and the milky emulsion feed (left). The emulsions from light oil hexane, toluene, octane, and xylene to heavier natural oils soybean, mustard, and coconut were effectively separated by the membrane. Excellent clarity of the recovered oil phase and great separation efficiency have been demonstrated by the membrane. The remarkable transparency of the recovered oil phases confirms that the membrane has higher separation efficiency and excellent capability across a wide range of oil types.

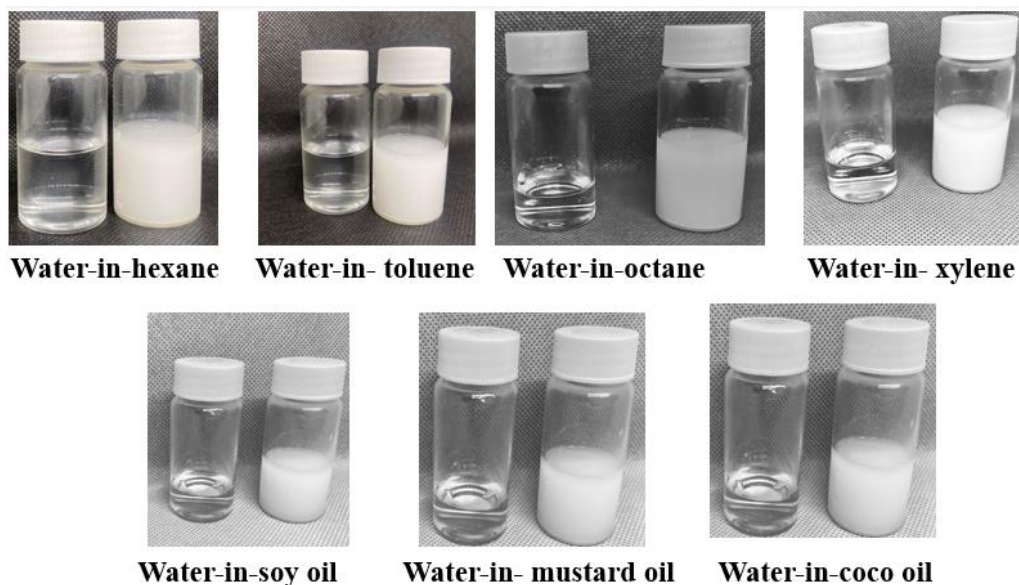


Figure 10. Recovery of various oils by Janus 7.5P-T_{0.24}-D membrane

Conclusion

In this study, hydrophobic PVDF membranes were prepared through phase inversion followed by surface modification to form Janus structures suitable for efficient water-in-oil emulsion separation. The strategy involved rapid deposition of TA on the upper membrane surface, followed by oxidative crosslinking using NaIO₄ with an optimized TA: NaIO₄ ratio of 12:1. The resulting membrane achieved a high separation efficiency of beyond 99% and maintained stable performance over seven reuse cycles. A maximum permeation flux of 1875 L m⁻² h⁻¹ was obtained, reflecting a well-balanced combination of high flux and selectivity. The Janus membrane's under-oil super-hydrophilic surface facilitated efficient demulsification, enabling excellent separation of various water-in-oil emulsions, particularly water-in-hexane and water-in-toluene, both exceeding 99% efficiency. These results demonstrate the membrane's strong potential for practical and scalable water-in-oil separation applications.

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