

Electronic Supplementary Information
Robust Superhydrophobic V₂O₅/CoFe₂O₄@SiO₂/PDMS Composite
Membrane for Membrane Distillation

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1. Materials

All the chemicals and solvents with reagent grade quality were purchased commercially and used without any further purification. Nylon microfiltration membrane (pore sizes: 0.2, 0.45, 0.8 μm) was obtained from Axiva Sicheem Pvt. Ltd. and PTFE microfiltration membrane (pore size: 0.22 μm) was purchased from Sterlitech Co., USA. SYLGARD™ 184 PDMS Silicone Elastomer Kit was obtained from Dow Chemical Company. Ferric chloride hexahydrate (97%), cobalt chloride hexahydrate (99%), vanadium pentoxide (AR, 99.5%), sodium borohydride, sodium chloride (AR, 99.9%), sodium dodecyl sulphate (AR, 99%) and hexane (99%) were purchased from Siscon Research Laboratories Pvt. Ltd. Bis[3-(triethoxysilyl)propyl] tetrasulfide, S 22.3% (typical) was purchased from Thermo Scientific, tetraethyl orthosilicate from Sigma Aldrich, sodium hydroxide from Fisher Scientific, ethylene glycol and hydrogen peroxide from Qualigens, hexadecane from Spectrochem, and ethanol from Analytical CS reagent.

2. Characterization

Membrane surface morphology was measured by Scanning electron microscopy (FEI, Quanta 200) with energy dispersive X-ray (EDX) analysis. Attenuated total reflectance Fourier transform infrared spectroscopy (ATR-FTIR) spectra were recorded on (Shimadzu IR Tracer-100) to characterize the chemical structure of the prepared powder sample and membranes. The samples were placed on the sample holder and all spectra were recorded from 4000 cm^{-1} to 400 cm^{-1} . The membrane thickness was measured using a thickness gauge (Mitutoyo, 0-25mm) with an accuracy of 1 μm . Thermogravimetric analysis of membranes was carried out with TGA 4000 PerkinElmer instrument with a heating rate of 10°C/min under nitrogen atmosphere. Inductively coupled plasma optical emission spectrometer (ICP-OES) (Model: iCAP 7600, Make: Thermo Fisher Scientific) was used for determination of the concentration

of iron metal present in the permeate solution. Atomic force microscope (AFM, Park Systems Corporation, XE7) was employed to measure the surface roughness of the composite membrane.

Static and dynamic water contact angles were measured by a contact angle goniometer (sessile drop) KYOWA DMS-40, using half-angle method fitting and by FAMAS add-in software with the solvent droplet range of 0.5–2 μ L on the membrane's surface. Water contact angles with at least 5 different positions at a membrane surface were tested and the average value was taken as the final contact angle value. Advancing contact angle (θ_A) and receding contact angle (θ_R) were determined by gradually increasing and decreasing the droplet volume on the membrane surface, respectively, to assess surface wettability and hysteresis.

2.1. Surface Free Energy

The surface free energy (SFE) of the membranes was assessed via the extended Fowkes model, also referred to as the Kitazaki-Hata theory.¹⁻² By partitioning the SFE majorly into three components including dispersive, polar, and hydrogen bonding this model offers a comprehensive picture of molecular interactions of the solid surface. The SFE of the material is equal to the sum of the dispersive, polar and hydrogen bonding components.

$$\gamma^{total} = \gamma^d + \gamma^p + \gamma^h$$

γ^d = dispersive component, γ^p = polar component, γ^h = hydrogen bonding component

Contact angles were measured using three solvents with varied polarities and surface tensions including water (~72.8 mN/m), ethylene glycol (~47.7 mN/m), and hexadecane (~27.6 mN/m)] at 25°C on the composite membranes. Using the measured contact angle values and applying the Kitazaki-Hata equation, the dispersive, polar, and hydrogen bonding components of the surface energy were calculated. The extracted values are summarized in Table S2.†

2.2. Porosity calculation

Porosity (ε) of the membranes were evaluated by using the gravimetric method.³ In brief, membrane specimens with a size of 1.5 x 1.5 cm² were cut from each membrane, weighed, and then immersed in isopropanol for 25 seconds. Afterward, each square was taken out, gently dabbed on tissue paper to remove surface residue, and weighed again to determine the mass of the liquid absorbed by the pores. The membrane porosity was then calculated by using the following equation

$$\varepsilon = \frac{\frac{W_w - W_d}{\rho_i}}{\frac{W_w - W_d}{\rho_i} + \frac{W_d}{\rho_p}} \times 100$$

Where W_w and W_d represent the weight of wet and dry membranes (g), respectively, ρ_i is the density of isopropanol in g/cm³, and ρ_p represents the polymer density in g/cm³.

2.3. Liquid entry pressure

The LEP of the composite membranes was determined using a custom-made setup. Initially, the membrane was placed in a sealed membrane filter holder (Stainless Steel Filter Holder, diameter 25 mm). Deionized water was poured onto the membrane holder, followed by connected to an N₂ gas cylinder. A barometer positioned between the cylinder, and the holder allowed for precise pressure measurement. The pressure was gradually increased until the first water drop appeared, and this pressure was recorded as the LEP of the membrane. This process was repeated three times and the average value was reported as the final LEP.

2.4. Stability tests

The thermal, mechanical and chemical stability of the BV-CS-P was investigated under extreme conditions including hot saline water (70°C) for 144 h, sonication for 90 min, and acidic (HCl solution, pH = 1) and basic (NaOH solution, pH = 14) conditions at room

temperature for 3 h respectively and a sequential acid-base soaking for 4 h. The water contact angle of the membrane was then measured at room temperature.

2.5. Nano indentation-Mechanical test

The surface hardness (H) and elastic modulus (E) of the membranes were precisely determined through depth-sensing nanoindentation using a Nios Scanning Nanohardness Tester (Ostec Instruments, Moscow, Russia) equipped with a high-precision Berkovich pyramidal indenter. Prior to testing, the membranes were mounted onto glass substrates using water and left to dry at room temperature. Measurements were conducted in a load-controlled mode following a four-step procedure.⁴⁻⁵ Initially, a certain preload force was applied to establish contact between the indenter and the membrane surface. Once contact was confirmed, the load was increased at a constant rate of 500 $\mu\text{N/s}$ until reaching the maximum force over a span of 10 seconds. This peak load was held steady for 5 seconds to allow deformation to stabilize. The final stage involved unloading the sample at the same rate used during loading. Throughout the process, the instrument continuously monitored both the applied force and the displacement of the indenter to ensure precise and reliable data acquisition. Each membrane sample was tested with seven separate indentations to assess measurement repeatability.

The surface hardness (H) of a material, typically defined as the mean pressure exerted under the nanoindenter, can be calculated using the following equation:

$$H = \frac{P_{max}}{A} \quad eq. (1)$$

Where P_{max} is the maximum applied force and obtained directly from the load-displacement curve, and A is the projected contact area of the indenter's tip with the Nylon and BV-CS-P membranes.

The initial slope of the unloading segment of the curve (dP/dh) is used to determine the indentation modulus (M) of the material via the following relation:

$$S = \frac{dp}{dh} = \beta \frac{2}{\sqrt{\pi}} M \sqrt{A} \quad eq. (2)$$

Here, β is a correction factor based on the geometry of the nanoindenter tip. A Berkovich indenter was used in this study, with β value of 1.034.

The elastic modulus (E) of the material can then be calculated from:

$$\frac{1}{M} = \frac{1 - \nu^2}{E} + \frac{1 - \nu_i^2}{E_i} \quad eq. (3)$$

where ν is the Poisson's ratio of the sample, and E_i and ν_i are the Elastic modulus and Poisson's ratio of the indenter tip, respectively.

Using equations (1)–(3), both the hardness (H) and elastic modulus (E) of the Nylon and BV-CS-P membranes has been determined.

3. Membrane distillation performance

The desalination performance of BV-CS-P and other composite membranes were performed using the custom-made DCMD setup with an effective membrane area of 4.9 cm². The membrane was placed in a customized DCMD module. Each side of the membrane, one support mesh (1 mm thick) was used to hold the membrane in a planar shape and reduce the membrane deformation due to the pressure difference between feed and permeate flow. Feed saline water was heated at 70°C by the heating circulator (HWC 110, Borg Scientific) and circulated to the DCMD module using the inbuilt circulator with a constant flow rate of 92 ml/min. The permeate side temperature was maintained at 20°C using a refrigerated water bath circulator (CRC 105, Borg Scientific). Four types of saline solutions including 0.3 g/L, 3.5 g/L, 7 g/L NaCl, and sea water (Besant Nagar beach, Tamil Nadu, India) were used to measure the MD performance. Additionally, the salt solution made from 3.5 g/L NaCl is mixed with 35 mg/L SDS and 100 mg/L rust (prepared by precipitation of [Fe(OH)₃] using 100 ml of 3 M of

NaOH and 1 M of FeCl_3 [$\text{Fe}(\text{OH})_3$] respectively were tested to study the anti-fouling and anti-scaling properties. To ensure uniform dispersion and simulate real time fouling conditions, $\text{Fe}(\text{OH})_3$ was subjected to 30 min. sonication in distilled water before use in the anti-scaling tests. The conductivities of feed and distilled water were measured in an interval of half an hour by the conductivity meter (HI2003-02, Hanna Equipments), while salt rejection was calculated using the following equation.

$$\text{Salt Rejection (\%)} = \left(1 - \frac{C_p}{C_f} \right) \times 100\%$$

Where C_p and C_f is the concentration of NaCl in permeate and feed side.

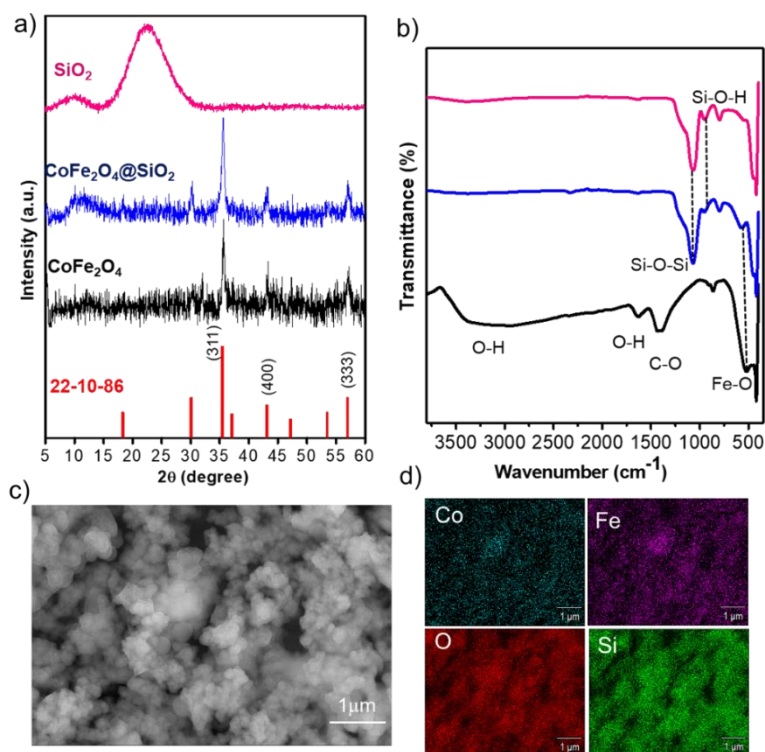
Two temperature and pressure sensors were integrated to measure the liquid temperature and pressure at feed and permeate side respectively. One digital flow meter (FTB336D, Omega) and one mount panel flow meter (FL50003A-V, Omega) were installed in the pipelines to continuously monitor the flow rate in the feed and coolant loops.

The flux generated from the direct contact membrane distillation performance was recorded in an interval of half an hour and calculated from the equation

$$\text{Flux } (J_w) = \frac{Q}{\Delta t \times A}$$

Where Q = increase in water in permeate side (litre), Δt = operation time interval (h),
 A =effective membrane area (m^2).

166 4. Characterization of CS ($\text{CoFe}_2\text{O}_4 @ \text{SiO}_2$)

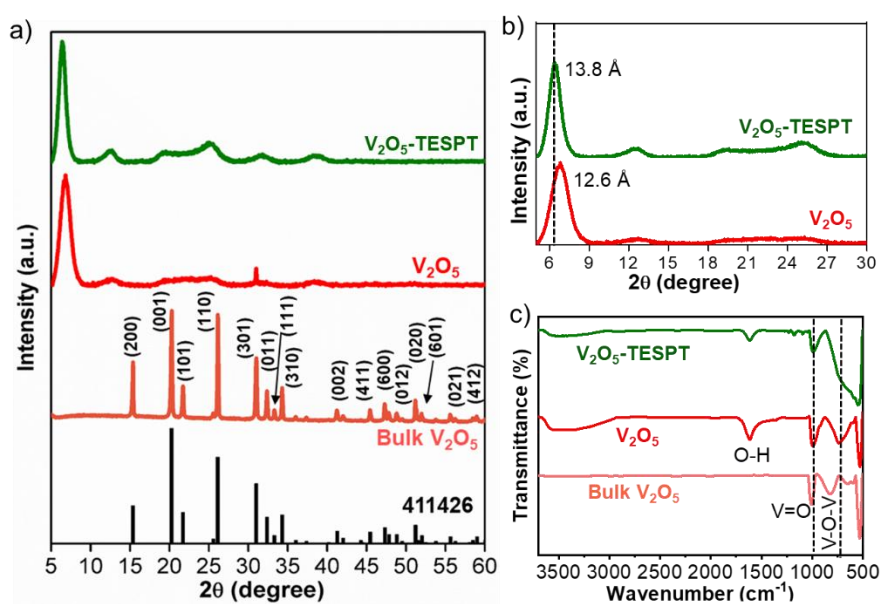


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168 Fig. S1. (a) XRD patterns and (b) FTIR spectra of CoFe_2O_4 , $\text{CoFe}_2\text{O}_4 @ \text{SiO}_2$ (CS) and SiO_2

169 (c) SEM image and corresponding (d) elemental mapping of CS.

170 5. Characterization of V_2O_5 -TESPT



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Fig. S2. (a) & (b) XRD patterns and c) FTIR spectra of bulk V_2O_5 , V_2O_5 (V_2O_5 nanosheets) and V_2O_5 –TESPT.

Table S1. Components used in composite membrane fabrication.

Sl. No.	Abbreviation	V_2O_5 –TESPT (mg)	CS (mg)	P (ml)
1	P(1)(0.45)	-	-	1
2	BV-P(1-1)(0.45)	1	-	1
3	BV-CS-P(1-5-1)(0.2)	1	5	1
4	BV-CS-P(1-2-1)(0.2)	1	2	1
5	BV-CS-P(1-5-1)(0.45)	1	5	1
6	BV-CS-P(1-2-1)(0.45)	1	2	1
7	BV-CS-P(1-5-1)(0.8)	1	5	1

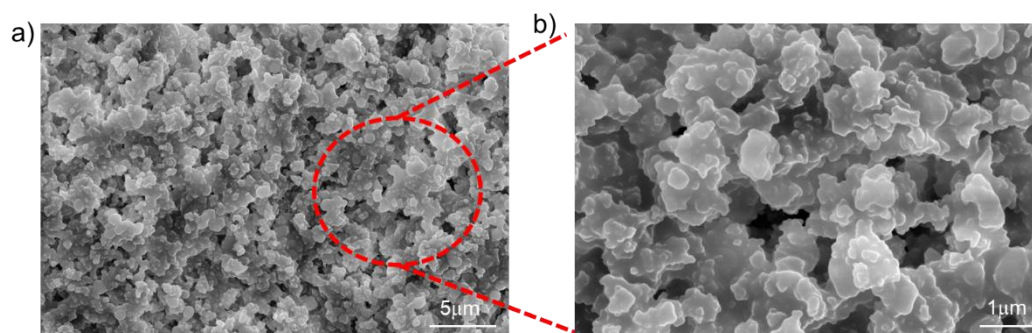


Fig. S3. (a) & (b) SEM images of BV-CS-P(1-5-1)(0.45) at different magnifications.

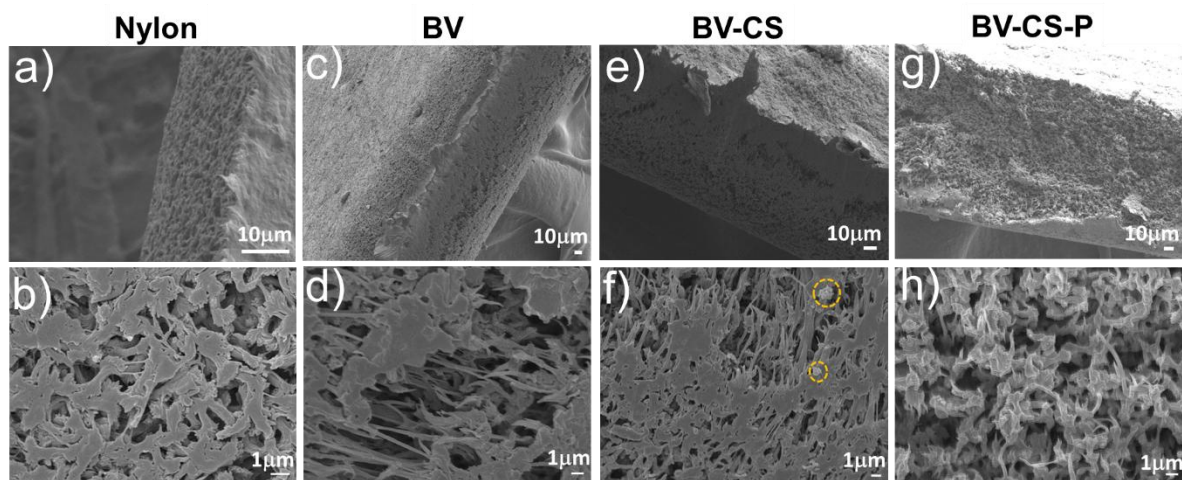


Fig. S4. Cross-sectional SEM images of membranes at low (top row) and high (bottom row) magnifications.

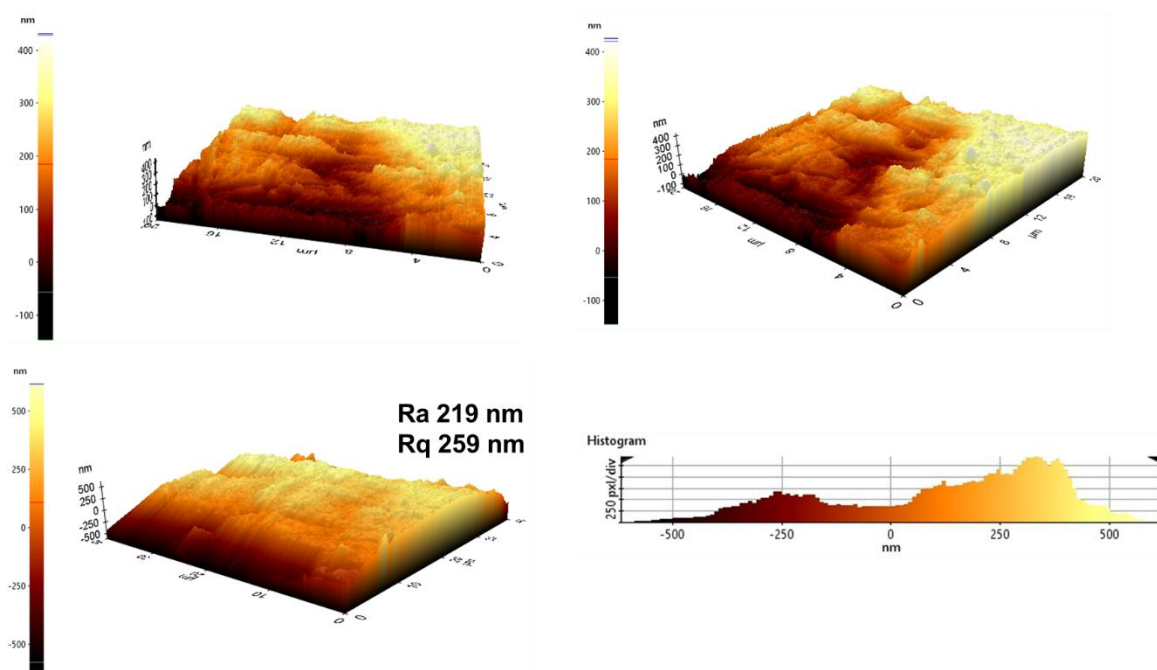


Fig. S5. AFM images, height profile and roughness parameters of BV-CS-P(1-5-1)(0.45).

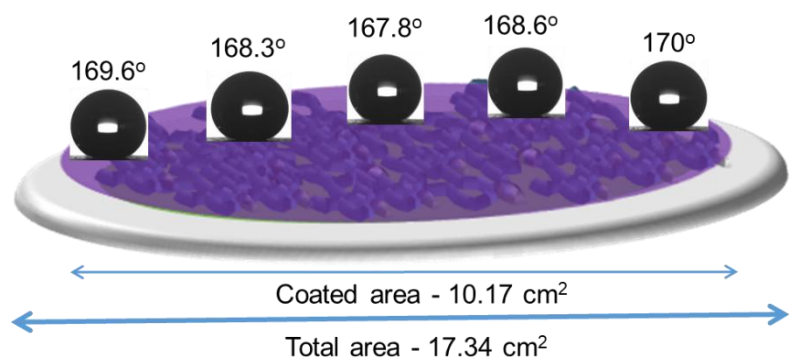


Fig. S6. Water contact angle (WCA) taken across the BV-CS-P(1-5-1)(0.45).

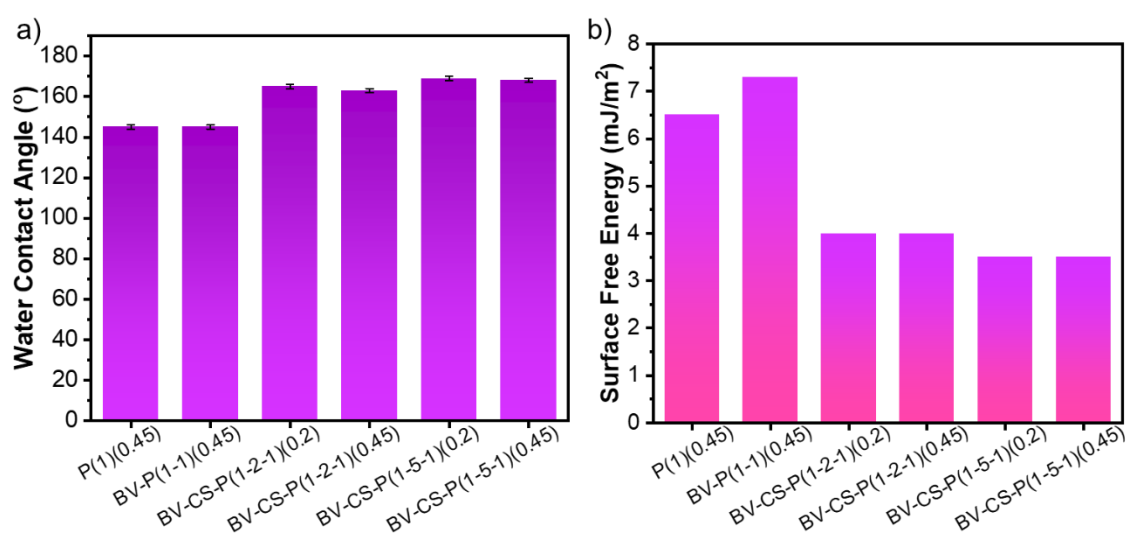


Fig. S7. (a) WCA and (b) corresponding surface free energy of composite membranes.

Table S2. Solvent contact angles and surface free energy parameters of various composite membranes.

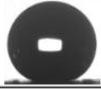
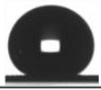
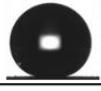
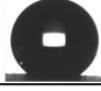
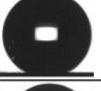
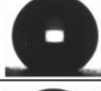
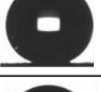
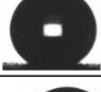
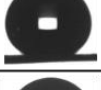
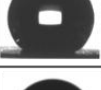
Solvents Membranes	Water	Ethylene glycol	n-hexadecane	S.F.E (mJ/m²)
BV-CS-P (1:5:1)(0.45)	168±1° 	152° 	10° 	3.5
BV-CS-P(1:5:1)(0.2)	169±1° 	153° 	10° 	3.5
BV-CS-P (1:2:1)(0.45)	163±1° 	147° 	10° 	4
BV-CS-P (1:2:1)(0.2)	165±1° 	145° 	12° 	4
BV-P(1-1)(0.45)	145±1° 	125° 	14° 	7.3
P(1)(0.45)	145±1° 	132° 	13° 	6.5
BV-CS-P (1:5:1)(0.45) backside	145±2° 	61° 	16° 	18.4
BV(1)(0.45)	76±3° 	47° 	17° 	34
Bare nylon (0.45)	31±1° 	29° 	12° 	201.8

Table S3. Advancing (θ_A), receding (θ_R) contact angle and contact angle hysteresis (θ_{CAH}) of the BV-CS-P(1-5-1)(0.45) taken during the stability test in hot saline water.

Time (hr)	θ_A (°)	θ_R (°)	θ_{CAH} (°)
0	168.8	168.8	0
24	168.8	168.2	0.6
48	167	166.2	0.8
72	164	163.2	1.2
96	164.6	161.6	3
120	166.7	158.4	8.3
144	163.8	151.3	12.5

6. TG Analysis

The effect of PDMS coating on the thermal stability of the BV-CS-P was investigated using thermogravimetric analysis (TGA). Thermal stability has always been the key to long-term stable use of MD membranes. As shown in Fig. S8, three weight loss were observed. The first weight loss at 35 °C to 300 °C (9.15% for BV-CS and 3.64 % for BV-CS-P was mainly owing to the elimination of the water molecule adsorbed on the membrane. The inhibition of water desorption from the PDMS-coated membrane could be ascribed to a high water barrier property of the hydrophobic PDMS coating. The degradation observed at 350 °C to 650 °C can be ascribed to the pyrolysis of the functional groups of the polymer. For instance, the weight loss of PDMS non-coated BV-CS membrane is 88.4 % whereas the PDMS coated membrane can be estimated as 75.6%. This suggests that the interactions between the silica coated cobalt ferrite and the PDMS functionalities effectively promote thermal stability. The last stage (0.11 % for BV-CS and 19.11 % for BV-CS-P) was the decomposition of polymer backbones.

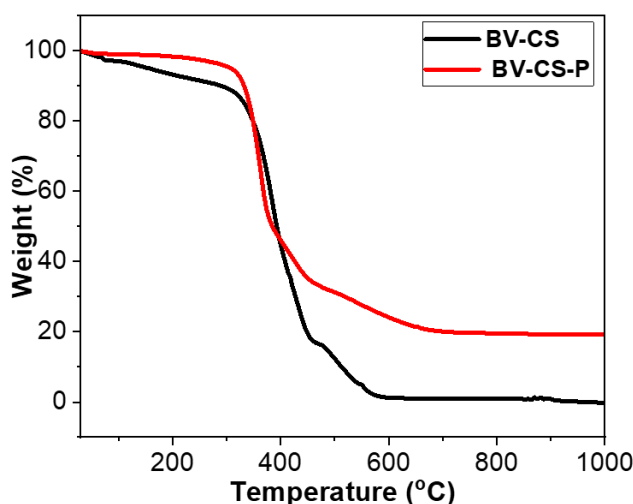


Fig. S8. TG analysis of the composite membrane conducted before and after PDMS modification.

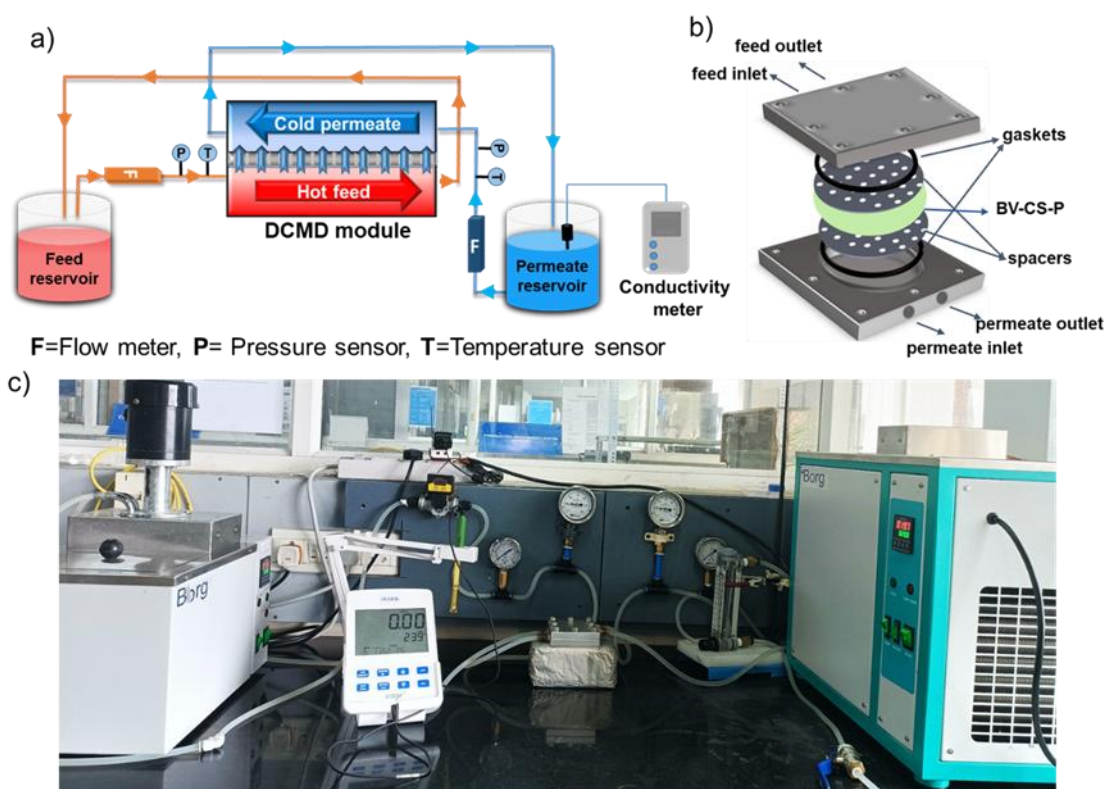


Fig. S9. (a) Schematic representation of the Direct Contact Membrane Distillation (DCMD) setup, (b) 3D diagram illustrating the layout of the DCMD module and (c) photograph of DCMD testing setup.

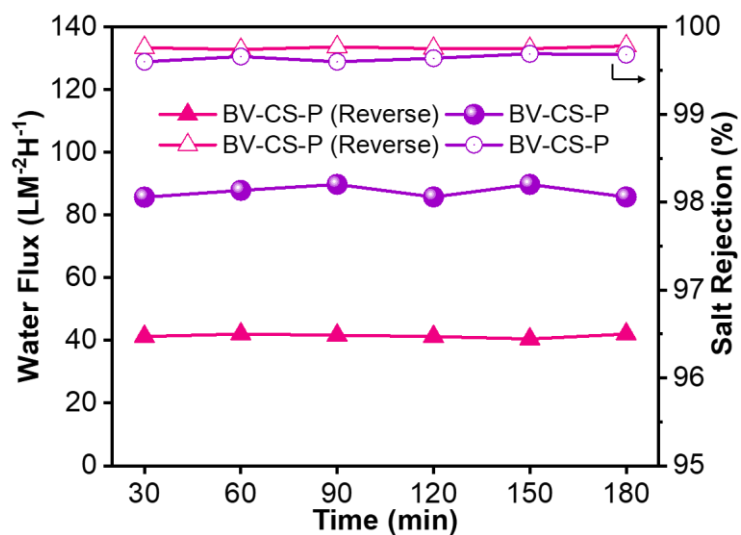
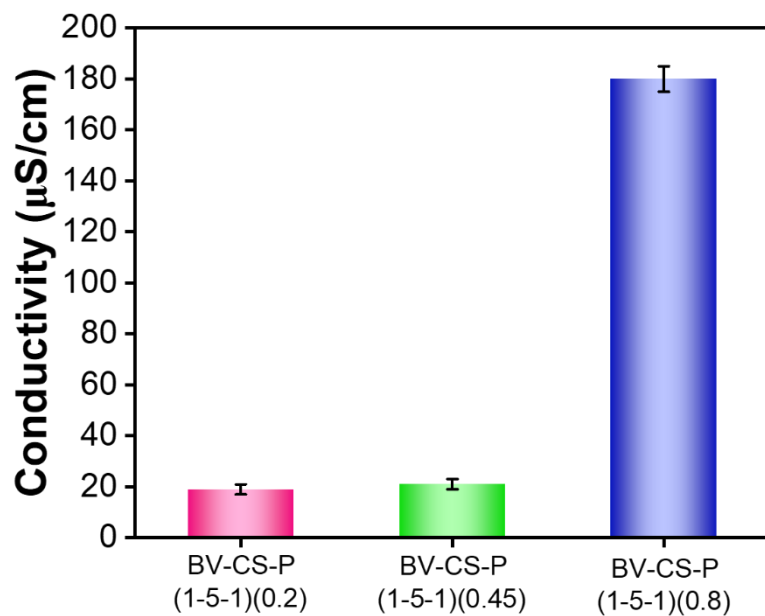
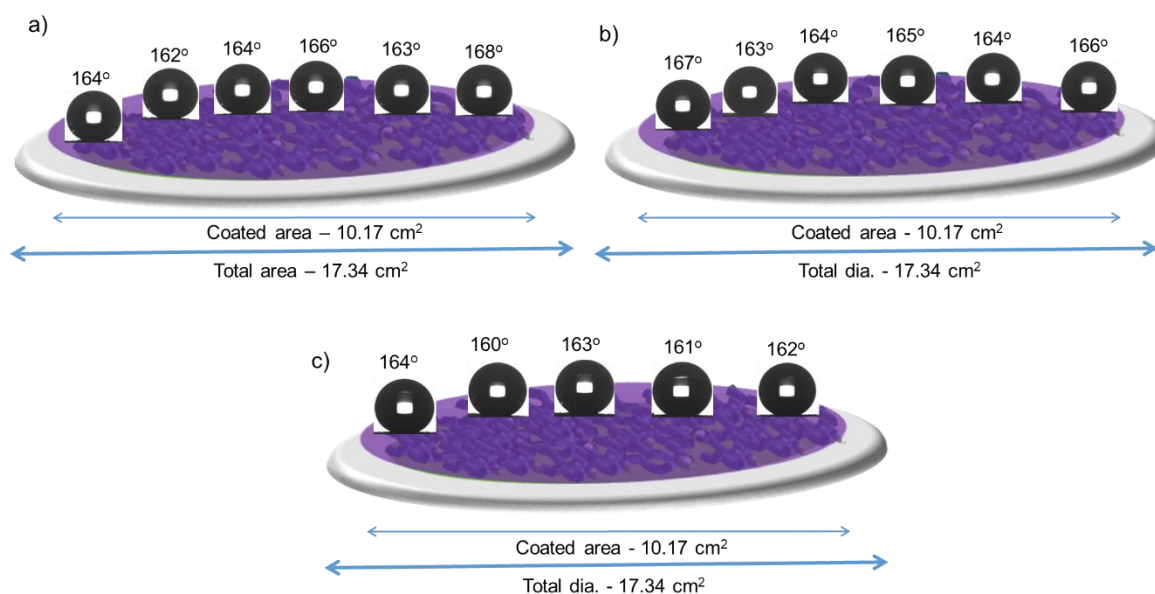


Fig. S10. Comparison of water flux (closed symbols) and salt rejection (open symbols) of BV-CS-P in the standard and reverse orientation configurations.



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228 Fig. S11. Comparison of the permeate conductivity with respect to substrate pore size.



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230 Fig. S12. WCA across the BV-CS-P(1-5-1)(0.45) after treatment with (a) 3.5 g/L, (b) 7 g/L

231 NaCl and c) sea water as feed solution.

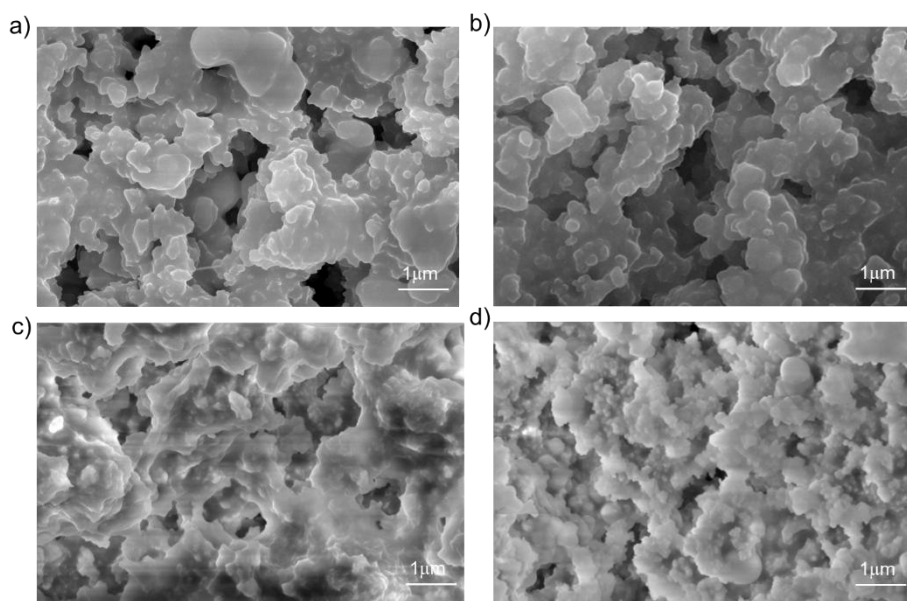


Fig. S13. SEM images of untreated and recovered BV-CS-P(1-5-1)(0.45) after treatment with various salt concentrations. (a) Untreated, (b) 0.58 g/L, (c) 3.5 g/L, (d) 7 g/L NaCl as feed solution.

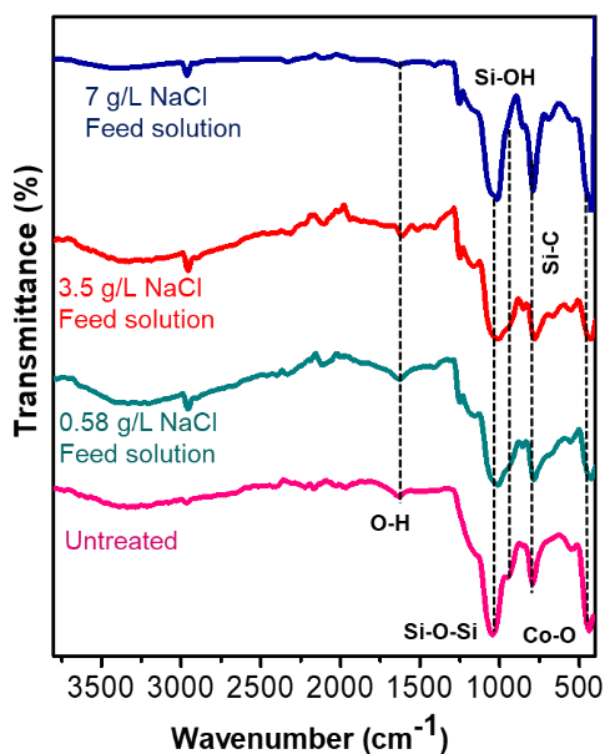


Fig. S14. FTIR spectra of recovered BV-CS-P(1-5-1)(0.45) after treatment with various salt concentrations as feed solution with respect to the untreated membrane.

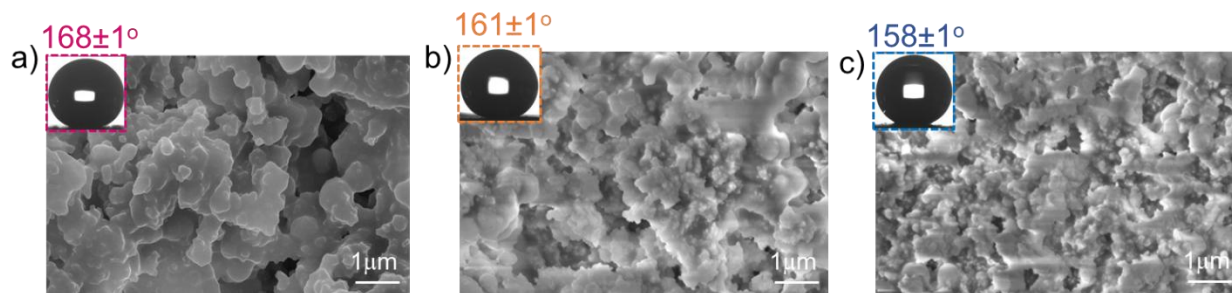


Fig. S15. SEM image and corresponding WCA of untreated and recovered BV-CS-P(1-5-1)(0.45) after treatment with foulants. (a) Untreated, (b) rust, and (c) SDS with NaCl as feed solution.

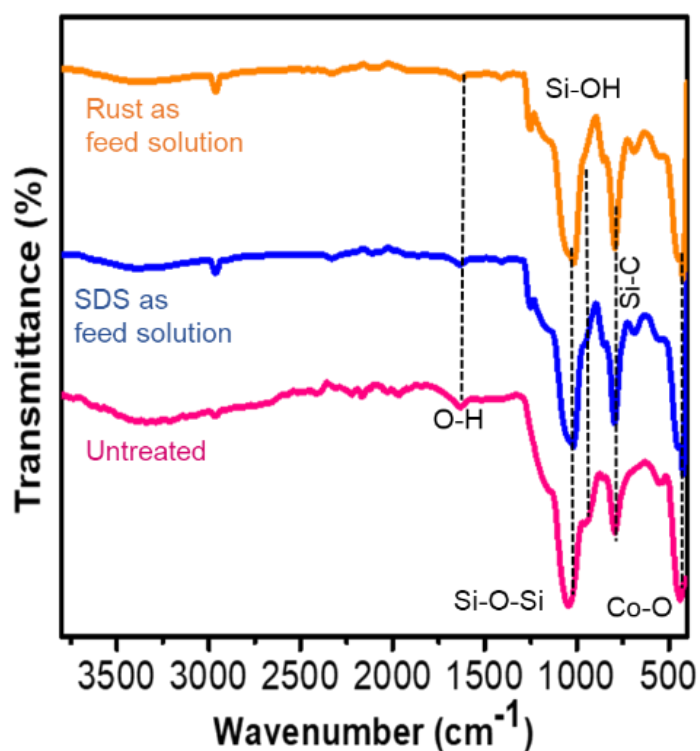


Fig. S16. FTIR spectra of recovered BV-CS-P(1-5-1)(0.45) with respect to the untreated membrane after treatment with SDS and rust with NaCl as feed solution.

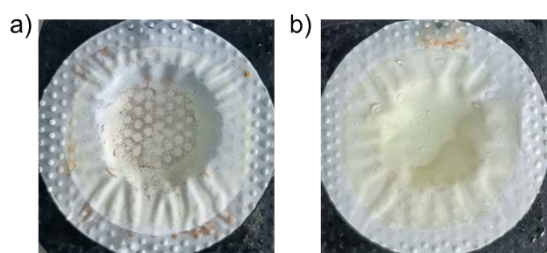


Fig. S17. Digital images of the BV-CS-P(1-5-1)(0.45) before and after water flushing, following membrane distillation of a saline solution containing rust $[\text{Fe}(\text{OH})_3]$.

Table S4. Comparison of the membrane distillation performance of BV-CS-P with the state-of-the-art hydrophobic membranes using PDMS as the polymer and surface modifier.

Sl. No.	Fabrication process	WCA	MD Configuration	Operational Parameters	Performance		Ref.
					Flux (LMH)	Rejection	
1	Electrospinning of PAN followed by heat-pressing at 140°C : PAN Electrospinning of PH (PVDF-HFP) over PAN: PH-PAN Electrospraying PS/PDMS over PH-PAN: PH-PS/PDMS	148°	DCMD	3.5 wt% NaCl, 70°C , 10°C Area- 22.7 cm^2 Flow rate- 0.55 L/min	27	$\sim 100\%$	⁶
2	Electrospinning of PVDF: PVDF Polymerization of aniline over PVDF for 30 min: PANi@PVDF Immersion of PANi@PVDF in 17-FAS, PDMS for 12 h : F/Si-PANi@PVDF	155°	DCMD	3.5 wt% 45°C . Area - 16 cm^2	28	99%	⁷

3	Electrospinning of PVDF: PVDF Electrospraying of premixed PVDF, PVDF/ PDMS and SiO₂ solution over PVDF: PDMS-3	170°	DCMD	3.5wt % NaCl, 60 °C, 20 °C Area- 28 cm ² Flow rate-0.5 L/min	28	99.9%	⁸
4	Electrospinning of PVDF: PVDF ENM Immersion of PVDF ENM in PDMS, triethoxyvinylsilane, and ditin butyl dilaurate solution followed by heating at 80°C for 12 h : Composite ENM	148°	VMD	3.5 wt% NaCl, 80 °C, Area-19.63 cm ² Flow rate-130 mL/min	35	99%	⁹
5	Dip-coating of PES substrate (0.2 µm) in APTES followed by heating at 60°C for 5 min. Immersion in silica NPs (5–15 nm) solution, and dried at 60°C for 1 h. Filtration of PDMS and FAS, dried at 60°C for 7 h: PDMS-FAS/SiNP	131°	DCMD	1 M NaCl 60 °C, 20 °C Flow rate-600 mL/s 300 mL/s Area-8.6 × 3.6 cm ²	29	99.9%	¹⁰
8	PVDF-PDMS membrane using NIPS method. Dip-coating in Tollens' reagent followed by air drying for 12 h: Ag/PVDF-PDMS Membrane activated with Pd and reduced using hydrochloric acid:	85°	VMD	3 wt% NaCl, 80 °C Flow rate-80 L/h	5.5	99.9%	¹¹

	Ag/a-PVDF-PDMS						
7	Immersion of PVDF substrate (0.22 μ m) in catechol and PEI solution, then drying at 45°C overnight: PCPA/PVDF In-situ growth of Ag nanoparticle over PCPA/PVDF in silver nitrate and PVP followed by drying at 45°C: Ag/PCPA/PVDF Immersion of Ag/PCPA/PVDF in PFDT at 30°C for 24 h then drying at 45°C : F-Ag/PCPA/PVDF Treating F-Ag/PCPA/PVDF with PDMS followed by heating at 120°C for 2 h : PF-Ag/PCPA/PVDF	151°	DCMD	3.5 wt% NaCl 60°C and 20°C Flow rate-0.9 L/min Area- 63.59 cm²	17.6	99.9%	¹²
8	Immersion of electrospun polyimide in dopamine and PEI solution: PDA/PEI Deposition of SiO₂ NPs (30 nm) over PDA/PEI: PDA/PEI-SiO₂ Soaking PDA/PEI-SiO₂ in 17-FAS and PDMS precursor followed by heating at 120°C: RSHO	158°	DCMD	20 wt% NaCl, 55°C and 20°C Flow rate-100 mL/min Area-9 cm²	25.4	100%	¹³

9	Immersion of PVDF substrate (0.45 μm) in PDMS and cured at 70°C for 12 h: PDMS-PVDF Polymerization of dopamine over PDMS-PVDF: PDA-PVDF Coating of PDA-PVDF with PEI: PEI-PVDF Filtration of MXene over PEI-PVDF followed by drying at 70°C for 3 h: MXene-PVDF	130°	DCMD	0.4 mM SDS in 1.5 wt% NaCl, 100 ppm soybean oil in 1.5 wt% NaCl 80 °C and 12 °C Area-15.9 cm^2	9.3, 9.1	99.9%	¹⁴
10	Filtration of V_2O_5-TESPT over nylon substrate (0.45μm): BV Roughness creation by depositing $\text{CoFe}_2\text{O}_4@\text{SiO}_2$ NPs (CS) over BV: BV-CS PDMS (P) coating on BV-CS followed by drying at 120°C for 2 h: BV-CS-P	168°	DCMD	3.5 g/L NaCl 70°C and 20°C Sea water (~45,000 $\mu\text{S}/\text{cm}$) 35 mg/L SDS in 3.5 g/L NaCl 100 mg/L rust in 3.5 g/L NaCl Flow rate-93 mL/min Area-4.9 cm^2	87 82 90 83	99.4-99.8 %	Our work

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Table S5. Comparison of the membrane distillation performance of BV-CS-P with the state-of-the-art hydrophobic membranes made via vacuum filtration route.

Sl. No.	Fabrication Process	WCA	MD configuration	Operational Parameter	Performance		Ref.
					Flux (LMH)	Rejection	
1	Electrospinning of PVDF followed by hot pressing: PVDF Filtration of PP solution over PVDF: PP/PVDF-60	156°	DCMD	3.5 wt% NaCl 80°C and 20°C 0.6 L/min 30 cm ²	135.3	99.9%	¹⁵
2	Polymerization of dopamine over PTFE substrate (0.3 µm): P-PTFE Filtration of Fe ₃ O ₄ NPs over P-PTFE: Fe/P-PTFE Polymerization of dopamine over Fe/P-PTFE: P/Fe/P-PTFE	25°	VMD	6.98 wt.% 65°C 120 L/h 0.012 m ²	18.3	99.9%	¹⁶
3	Electrospinning of PVDF followed by hot pressing: PVDF Filtration of PP solution over PVDF: PP/PVDF-58	157°	DCMD	2 wt% NaCl 60°C 0.6 L/min 12cm ²	53.9	99.9%	¹⁷
4	Filtration of PDA NPs over PVDF substrate (0.38µm): PDA-PVDF Dip coating of PDA-PVDF in PDMS followed by heating at 120°C for 2 h:	162°	VMD	3.5 wt% NaCl 75°C 12 cm ² 10 L/h	19	99.9%	¹⁸

	PDMS-PDA-PVDF						
5	Dip-coating of PES substrate (0.2 μm) in APTES followed by heating at 60°C for 5 min. Immersion in silica NPs (5–15 nm) solution and drying at 60°C. Filtration of PDMS and FAS, dried at 60°C for 7 h: PDMS-FAS/SiNP	131°	DCMD	1 M NaCl 60°C and 20°C 600 mL/s 300 mL/s 8.6 $\times$ 3.6 cm^2	29	99.9%	¹⁰
6	Reduction of GO to rGO microspheres. Filtration of rGO microspheres (2 μm) and PTFE nanorods (100–200 nm) mixed with Nafion over nylon substrate (0.45 μm), followed by drying at 60°C for 4 h: Wrinkled rGO microsphere-PTFE	142°	VMD	3.5 wt% NaCl 50°C 30 L/min 12.57 cm^2	35.7	99.7%	¹⁹
7	Filtration of GO with silica NPs (400–500 nm) over PAN substrate followed by grafting of HDTMS and heating at 80°C for 4 h: hGOM4	120°	VMD	3.5 wt% NaCl 60°C 120 L/hr	13.5	99.9%	²⁰
8	Filtration of GO substrate (1μm): GOM Air spraying of TMOS and	150°	AGMD	hydrogen isotopic water separation	0.036	-	²¹

	FTMS-modified silica over GOM. Spray coating of 17-FTMS and MTMOS followed by drying at 70°C for 2 h: 3L-40F						
9	Filtration of V ₂ O ₅ -TESPT over nylon substrate (0.45µm): BV Roughness creation by depositing CoFe ₂ O ₄ @SiO ₂ NPs (CS) over BV: BV-CS PDMS (P) coating on BV-CS followed by drying at 120°C for 2 h: BV-CS-P	168°	DCMD	3.5 g/L NaCl 70°C, and 20°C Sea water (~ 45,000 µS/cm) 35 mg/L SDS in 3.5 g/L NaCl 100 mg/L rust in 3.5 g/L NaCl Flow rate-93 mL/min Area-4.9 cm ²	87 82 90 83	99.4-99.8%	Our work

7. References

- Y. Kitazaki, T. Hata, *J. Adhes.*, 1972, **4**, 123-132.
- K.-Y. Law, H. Zhao, Determination of Solid Surface Tension by Contact Angle, in: K.-Y. Law, H. Zhao (Eds.), *Surface Wetting: Characterization, Contact Angle, and Fundamentals*, Springer International Publishing, 2016pp. 135-148.
- S. Leaper, E. O. Avendaño Cáceres, J. M. Luque-Alled, S. H. Cartmell, P. Gorgojo, *ACS Appl. Polym. Mater.*, 2021, **3**, 1854-1865.
- S. Lee, B. G. Choi, D. Choi, H. S. Park, *J. Membr. Sci.*, 2014, **451**, 40-45.
- G. Pharr, W. Oliver, *J. Mater. Res.*, 1992, **7**, 1564-1583.
- S. Li, L. Li, J. Zhong, R. Ma, X. Xu, H. Wu, Y. Yu, *Desalination*, 2022, **539**, 115950.
- L. Zhong, Z. Zhu, Y. Han, Q. Wang, D. Liu, F. Cui, B. Li, W. Wang, *Environ. Sci. Nano*, 2019, **6**, 2553-2564.
- Y. Liao, G. Zheng, J. J. Huang, M. Tian, R. Wang, *J. Membr. Sci.*, 2020, **601**, 117962.

277 9 L. Jiao, K. Yan, J. Wang, S. Lin, G. Li, F. Bi, L. Zhang, *Desalination*, 2020, **476**,
278 114210.

279 10 A. A. Khan, M. I. Siyal, C.-K. Lee, C. Park, J.-O. Kim, *Sep. Purif. Technol.*, 2019, **210**,
280 20-28.

281 11 D. Yue, Y. Wang, H. Zhang, D. Sun, B. Li, X. Ye, W. Fang, M. Liu, *J. Membr. Sci.*,
282 2021, **638**, 119718.

283 12 Z. Wei, Y. Jin, J. Li, L. Jia, Y. Ma, M. Chen, *Desalination*, 2022, **529**, 115649.

284 13 Z. Zhu, L. Zhong, T. Horseman, Z. Liu, G. Zeng, Z. Li, S. Lin, W. Wang, *J. Membr.*
285 *Sci.*, 2021, **620**, 118768.

286 14 X. Yan, C. Yang, C. Ma, H. Tao, S. Cheng, L. Chen, G. Wang, X. Lin, C. Yao,
287 *Chemosphere*, 2022, **307**, 136114.

288 15 L. Deng, P. Li, K. Liu, X. Wang, B. S. Hsiao, *J. Mater. Chem. A*, 2019, **7**, 11282-11297.

289 16 W. Zhang, S. Yu, P. Li, X. Ji, R. Ning, P. Li, *Sep. Purif. Technol.*, 2023, **313**, 123465.

290 17 L. Deng, K. Liu, P. Li, D. Sun, S. Ding, X. Wang, B. S. Hsiao, *J. Membr. Sci.*, 2020,
291 **598**, 117813.

292 18 C. Li, W. Liu, J. Mao, L. Hu, Y. Yun, B. Li, *Sep. Purif. Technol.*, 2023, **304**, 122182.

293 19 C. Lai, H. Wang, Z. Qu, S. Cheng, H. Fan, H. Meng, *Ind. Eng. Chem. Res.*, 2024, **63**,
294 12574-12581.

295 20 Y. Mao, Q. Huang, B. Meng, K. Zhou, G. Liu, A. Gugliuzza, E. Drioli, W. Jin, *J.*
296 *Membr. Sci.*, 2020, **611**, 118364.

297 21 M. Wen, M. Chen, K. Chen, P.-L. Li, C. Lv, X. Zhang, Y. Yao, W. Yang, G. Huang,
298 G.-K. Ren, S.-J. Deng, Y.-K. Liu, Z. Zheng, C.-G. Xu, D.-L. Luo, *J. Membr. Sci.*, 2021,
299 **626**, 119136.

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