Electronic Supplementary Information

A compact and scalable fabrication method for robust thin film composite membranes[†]

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Fig. S1 The surface SEM images and pore size distribution of (a) PET non-woven, (b) TR-NFM, and (c) PE separator.



Fig. S2 The SEM images of developed thin film composite membranes on the PE separator which were coated with (a) 1.0 ml (0.28 Lm^{-2}), (b) 1.5 ml (0.42 Lm^{-2}), and (c) 2.0 ml (0.56 Lm^{-2}) of loading volume at fixed membrane area (0.0036 m^{2}).



Fig. S3 The surface SEM images of the fabricated S-TFC-TR membranes at 0.56 L m⁻² loading volumes with different concentration of sulfonated poly(arylene ether sulfone) solution: (a) 4 wt%, (b) 6 wt%, and (c) 8 wt%.



Fig. S4 (a) Schematic diagram of sprayed layer thickness according to loading volumes at different spray gun-to-collector distances (15 cm and 20 cm, respectively). The surface SEM images of the fabricated S-TFC-TR membranes fabricated at 20 cm of spray gun-to-collector distance with (b) 1.11 Lm^{-2} and (c) 1.25 Lm^{-2} of loading volumes, respectively.





Fig. S5 Digital photographs of developed membranes on (a) PE separator and (b) TR-NFM before and after the thermal crosslinking.



Fig. S6 (a) A cross-flow and (b) alternative high-temperature apparatuses for OSN test.



Fig. S7 UV spectra of feed and permeate solutions which were measured after 40 times dilution: (a) 1 g L⁻¹ Chrysoidine G (C.S.G.)/DMF solution and (b) 2 g L⁻¹ Dispersed Red (D.R.) and Brilliant Blue (B.B.)/DMF solution. Digital photograph of feed and permeate solutions before and after 40 times dilution of (c) 1 g L⁻¹ C.S.G./DMF solution and (d) 2 g L⁻¹ D.R. & B.B./DMF solution, respectively.



Fig. S8 Illustration for investigating a potential of mass production using the electrospinning machine which attached the spray gun instead of a syringe. The moving speed of traverse and machine direction was designed to overlap 1/3 radius of the sprayed area each other.

| | Unit area (m ²) | Polymer formula | Polymer usage (kg m ⁻²) | Carbon footprint from polymer (kg m ⁻²) | Solvent 1 formula | Solvent 1 usage (kg m ⁻²) | Solvent 2 formula | Solvent 2 usage (kg m ⁻²) | Carbon footprint from solvent (kg m ⁻²) | Water bath usage (L) | Carbon footprint from distillation (kg m ⁻²) ^a |
|------------------------------------|-----------------------------------|------------------------------------|---|---|---|---|---|--|---|--|---|
| PI conventional fabrication method | 0.06 | $C_{24}H_{16}N_2O_5$ | 0.11 | 3.38 | C5H9NO | 0.29 | C_4H_8O | 0.10 | 10.66 | 20 | 0.05 |
| PEEK green fabrication method | 0.06 | $C_{21}H_{18}O_3$ | 0.06 | 2.09 | CH ₄ O ₃ S | 0.33 | H_2SO_4 | 0.11 | 1.82 | 15 | 0.04 |
| Proposed fabrication method | 0.0585 | $C_{26}H_{22}\!O_{6.4}S_{1.8}_{b}$ | 0.02 | 0.56 | (CH ₃) ₂ SO | 0.29 | - | - | 3.93 | - | - |
| | Unit area (m ²) | Crosslinker formula | Crosslinker usage (kg m ⁻²) | Solvent 3 formula | Solvent usage (kg m ⁻²) | Conditioner formula | Conditioner usage (kg m ⁻²) | Solvent 4 formula | Solvent 4 usage (kg m ⁻²) | Carbon footprint from coagulation bath (kg m ⁻²) | Total carbon footprint (kg m ⁻²) |
| PI conventional fabrication method | 0.06 | $C_{6}H_{16}N_{2}$ | 0.8 | C_3H_8O | 1.2 | $C_{18}H_{38}O_{10}$ | 1.2 | C_3H_8O | 0.8 | 102.25 | 116.29 |
| PEEK green fabrication method | 0.06 | - | - | - | - | - | - | - | - | 0.04 | 3.95 |
| Proposed fabrication method | 0.0585 | - | - | - | - | - | - | - | - | - | 4.49 |

Table S1 Information for calculating the carbon footprint from distillation and each fabrication process.

^a Carbon footprint of distillation for water bath was calculated on the based of MSF plant. Liu *et al* reported that MSF plant can process 5,032,133 m³ of water per day and it generates 13667.27 t of CO_2 per day.¹ Therefore, we assumed that the carbon footprint of distillation for water bath is 0.0025 kg L⁻¹.

^b Average polymer formula was calculated from 40 % disulfonated poly(aryelene ether sulfone) and 60% poly(arylene ether sulfone).

Carbon foot print of distillation for DMF was calculated in order to compare with that of membrane fabrication processes from Szekely's work.² As followed previous work, DMF is the 10^{th} largest system in OSN field. Annually, DMF was generated 2.79×10^{6} kg and 2×10^{6} kg of CO₂ was also generated for 70 % solvent recovery. Based on this relationship, the profile of CO₂ emission in distillation was drawn in **Fig. 11b**. To compare each system intuitively, the CO₂ emission was simply calculated following below assumptions: (a) CO₂ emission from distillation was considered from heating and evaporation to condensing for 70 % solvent recovery, (b) bench-scale membranes after use will be disposed by incineration, and (c) contaminated waste from each fabrication process is distilled.

References

- 1. J. Liu, S. Chen, H. Wang and X. Chen, *Energy Procedia*, 2015, **75**, 2483-2494.
- 2. G. Szekely, M. F. Jimenez-Solomon, P. Marchetti, J. F. Kim and A. G. Livingston, *Green Chem.*, 2014, **16**, 4440-4473.