Electronic Supplementary Material (ESI) for Green Chemistry.
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Electronic Supplementary Information
for
Reporting the unreported: the reliability and comparability of the literature on organic solvent nanofiltration

Hai Anh Le Phuong, [a,b] Christopher F Blanford [b,c]* and Gyorgy Szekely [a,d]*

[a] Department of Chemical Engineering & Analytical Science, School of Engineering, The University of Manchester, The Mill, Sackville Street, Manchester, M13 9PL, United Kingdom

[b] Department of Materials, School of Natural Sciences, University of Manchester, Oxford Road, Manchester, M13 9PL, United Kingdom

[c] Manchester Institute of Biotechnology, University of Manchester, 131 Princess Street, Manchester, M1 7DN, United Kingdom

[d] Advanced Membranes and Porous Materials Center, Physical Science and Engineering Division (PSE), King Abdullah University of Science and Technology (KAUST), Thuwal 23955-6900, Saudi Arabia

* Corresponding authors: Tel.: +441613068915, email: christopher.blanford@manchester.ac.uk; Tel.: +966128082769, email: gyorgy.szekely@kaust.edu.sa, gyorgy.szekely@manchester.ac.uk

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List of definitions

Process parameters:

Flux (*J*) is defined as the volume of liquid (V) that transports through the membrane per unit area (A) and per unit time (t). It is most commonly expressed in units of L m^{-2} h^{-1}.

$$J = \frac{V}{At} \quad \textbf{(Eq. S1)}$$

Permeance (*P*) can be defined as the flux normalized by the transmembrane pressure (p). It is most commonly expressed in units of L m^{-2} h^{-1} bar^{-1}.

$$P = \frac{J}{p} = \frac{V}{At} \quad \textbf{(Eq. S2)}$$

Rejection (*R*) is calculated as the function of the ratio of solute concentration in the permeate (C_P) and retentate (C_R). It is expressed as a percentage or decimal.

$$R = 100\% \left(1 - \frac{C_\mathrm{P}}{C_\mathrm{R}}\right) \quad \textbf{(Eq. S3)}$$

Molecular weight cut-off (MWCO) is defined as the solute molecular weight at which 90% of it is rejected by the membrane.

Flow rate of feed (F_feed) is the sum of the permeate (F_P) and retentate flow rate (F_R). The flow rate of feed therefore is only applicable for cross-flow configurations where a retentate flow can be defined.

$$F_\mathrm{feed} = F_\mathrm{P} + F_\mathrm{R} \quad \textbf{(Eq. S4)}$$

Green metrics:

E-factor (E) is calculated as the mass ratio of waste generated (m_{waste}) and the desired product ($m_{product}$).

$$E = \frac{m_\mathrm{waste}}{m_\mathrm{product}} = \frac{m_\mathrm{total} - m_\mathrm{product}}{m_\mathrm{product}} \quad \textbf{(Eq. S5)}$$

Process mass intensity (PMI) is calculated as the mass ratio of all materials used (m_{total}) and the desired product ($m_{product}$).

$$PMI = \frac{m_\mathrm{total}}{m_\mathrm{product}} = E + 1 \quad \textbf{(Eq. S6)}$$

Atom efficiency (AE) is calculated as the molecular weight ratio of the desired product ($M_{product}$) and starting materials ($M_{reactants}$)

$$AE = \frac{M_\mathrm{product}}{M_\mathrm{reactan\,ts}} \quad \textbf{(Eq. S7)}$$

Experimental

Literature data mining

OSN research papers published between 2015 and 2019 with at least 3 citations were selected for analysis. For the literature datamining the information below were gathered from all papers:

- Filtration conditions: e.g., solute-solvent system, concentration, membrane area, pressure, temperature, cross-flow velocity/stirring speed, feed flow rate, system volume, steady-state operation, configuration type, membrane area, experimental time
- Employed materials characterization techniques
- Membrane performance data: e.g., rejection, highest permeance
- Evidence of replication: standard deviation and the source of standard deviation (i.e., originated from deviation in the performance of the same membrane or originated from membrane replicates)
- Green considerations: e.g., solvents, solute used, polymer used for membranes, number of pre- and post-treatment, number of steps required for fabrication, green metric analysis
- Focus of the paper: membrane development, process or fundamental work

The list of OSN publications were acquired by using the Web of Science search engine (All Databases) with TOPIC=("organic solvent nanofiltration" OR "solvent resistant nanofiltration" OR "organophilic nanofiltration") as key words.

Analysis of green literature

Green-focused, OSN research papers published between 2014 and 2019 were selected for analysis. The list of OSN publications were acquired by using the Web of Science search term TOPIC=(("organic solvent nanofiltration" OR "solvent resistant nanofiltration" OR "organophilic nanofiltration") AND ("green" OR "sustainable" OR "efficient" OR "environment" OR "recycle" OR "recover" OR "renewable")).
For the literature datamining the information below were gathered from all papers:

- Reason for improved sustainability: use of green solvents, chemicals, use of renewable polymer, valorization of renewable feedstock, reduced energy consumption, lower carbon footprint, waste minimization through reduced steps, solvent, reagent, catalyst recovery
- Provided quantitative information on sustainability: E-factor, energy, carbon footprint calculations
- Focus of the paper: membrane development, process or fundamental work

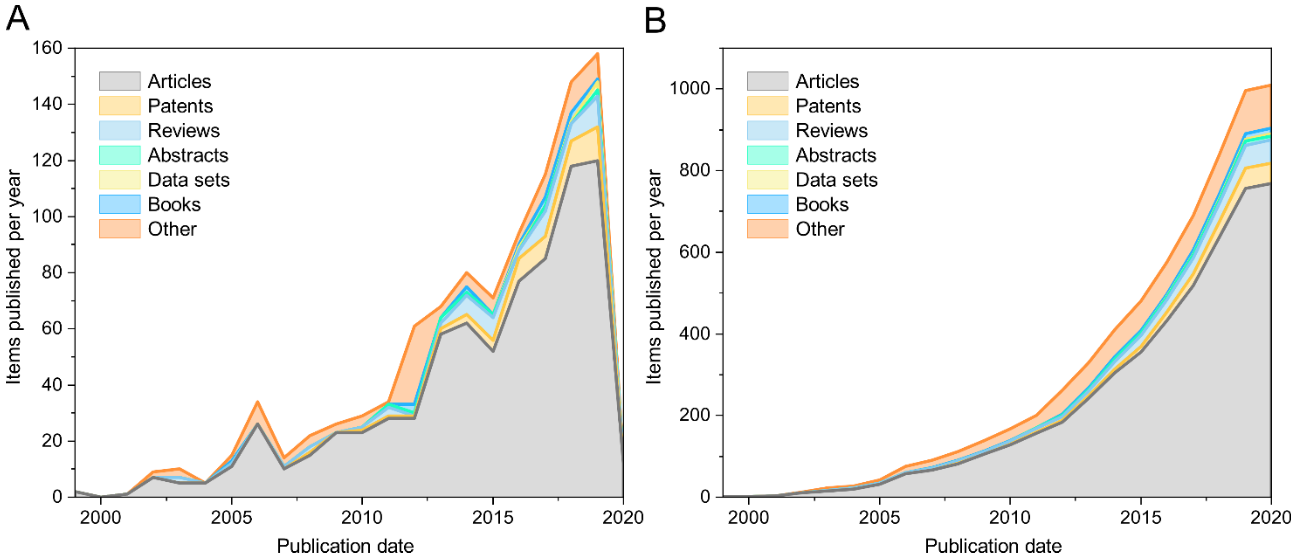

Fig. S1. Analysis of organic solvent nanofiltration publications. Publications by type in a given calendar year (A). Cumulative publications by type at the end of a calendar year (B). Document types for "Other" include art and literature, biographies, case reports, clinical trials, corrections, data papers, data studies, editorials, letters, meetings, news, reference materials, reports, retracted publications, thesis dissertations and unspecified publications. Data source: Web of Knowledge (All Databases). Search term: TOPIC=("organic solvent nanofiltration" OR "solvent resistant nanofiltration" OR "organophilic nanofiltration") "Early Access" included with "Papers". Search date: 2020-02-14.

Survey questions and answers

1. **What is your current position?**
   ☐ Doctoral student
   ☐ Other student
   ☐ Postdoctoral researcher
   ☐ Industrial researcher
   ☐ Other industrial role
   ☐ Faculty/Academic
   ☐ Other (please specify): ____

2. **What are the main issues with published papers on membrane filtration?** Select as many options as you want.
   ☐ I don't see a problem with the membrane literature in general
   ☐ Insufficient filtration tests performed on the membrane (e.g., single solvent, single solute, no pressure variation, etc.)
   ☐ Insufficient characterization of membranes
   ☐ No/unclear evidence of replication of datasets / membrane performance
   ☐ Insufficient experimental descriptions to reproduce the work
   ☐ Insufficient information on obtaining the data
   ☐ Inability to compare literature data (e.g., data obtained under different experimental conditions and therefore the membrane performance in the literature cannot be directly compared)
   ☐ Lack of long-term stability tests (at least few days)
   ☐ Other issues (please specify): ____

3. **Which parameters should be presented in papers on membranes to ensure reproducibility and comparability of the performance tests?** Select as many options as you want.
   ☐ Operating pressure
   ☐ Temperature
   ☐ Feed flow rate
   ☐ Materials characterization
   ☐ Cross-flow velocity
   ☐ Membrane area
   ☐ Solvent type
   ☐ Solute type
   ☐ Solute concentration
   ☐ Experimental time
   ☐ Configuration type
   ☐ Configuration dimensions
   ☐ Other process parameter (please specify): ____

4. **What is the minimum materials characterization to report?** Select as many options as you want.
   ☐ FTIR
   ☐ SEM
   ☐ AFM
   ☐ Other characterization technique (please specify): ______________________________________

5. **What is the minimum membrane performance characterization to report?** Select as many options as you want.
   ☐ Membrane chemical stability test (e.g., solvent resistance)
   ☐ Rejection of a single solute
   ☐ Rejection of multiple solutes
   ☐ MWCO using a series of oligomers (PS, PEG, etc)
   ☐ Flux decline
   ☐ Flux or permeance of a single solvent
   ☐ Flux or permeance of multiple solvents
   ☐ Flux or permeance of solutions
   ☐ Other metrics (please specify): ____

6. **Which membrane configuration do you usually use for testing the membrane performance?**
   ☐ Dead-end
   ☐ Cross-flow
   ☐ Other configuration (please specify): ____

7. **If you selected dead-end configuration, why do you prefer using dead-end configuration over cross-flow?** Select as many options as you want.
   ☐ The full set-up is commercially available
   ☐ Cheap
   ☐ Other reasons (please specify): ____
   ☐ N/A

8. **If you selected cross-flow configuration, why do you prefer using cross-flow configuration over dead-end?** Select as many options as you want.
   ☐ Due to decreased concentration polarisation
   ☐ Can be used to assess continuous long-term performance (over several days)
   ☐ Other reasons (please specify): ____
   ☐ N/A

9. **Do you use in-house built or commercially available set-up?**
   ☐ In-house built
   ☐ Commercially available

10. **How do you select the solutes for performance test?**
    ☐ Literature-based selection
    ☐ Application-oriented selection

11. **If there were a guideline available on the report of experimental data to avoid missing parameters (e.g., pressure, temperature etc.), would it be beneficial to the OSN community?**
    ☐ Yes
    ☐ No

12. **Please write any additional recommendations on how to improve the quality of the data in membrane-focused publications.**

Survey results

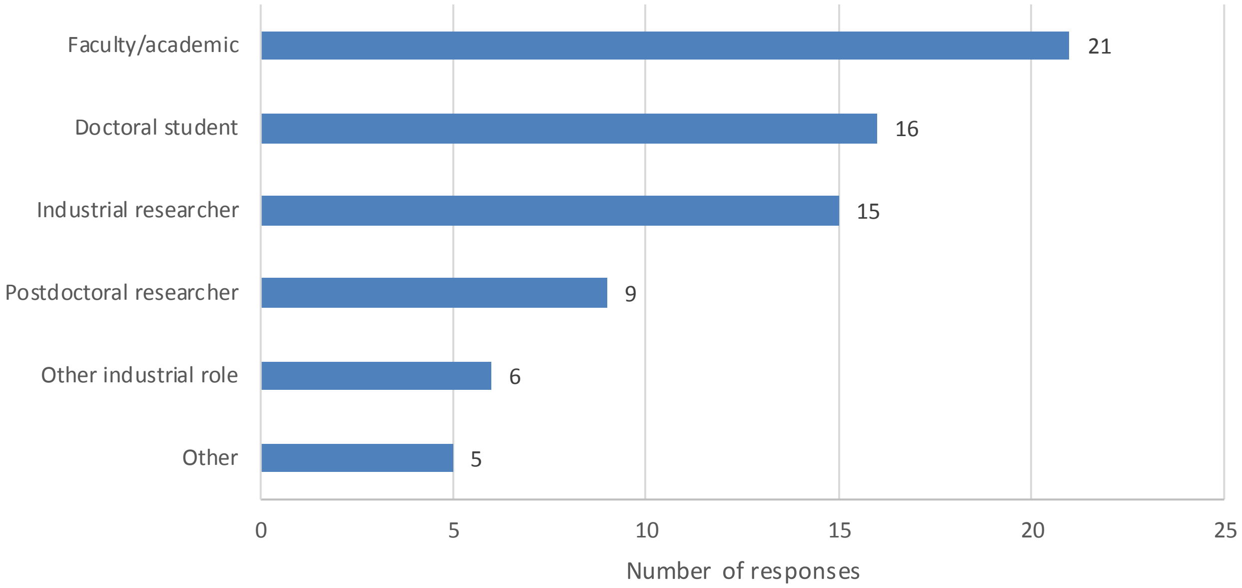

Fig. S2. Employment demographics of survey participants displayed as number of respondents per category (survey question 1). The total number of survey participants was 70.

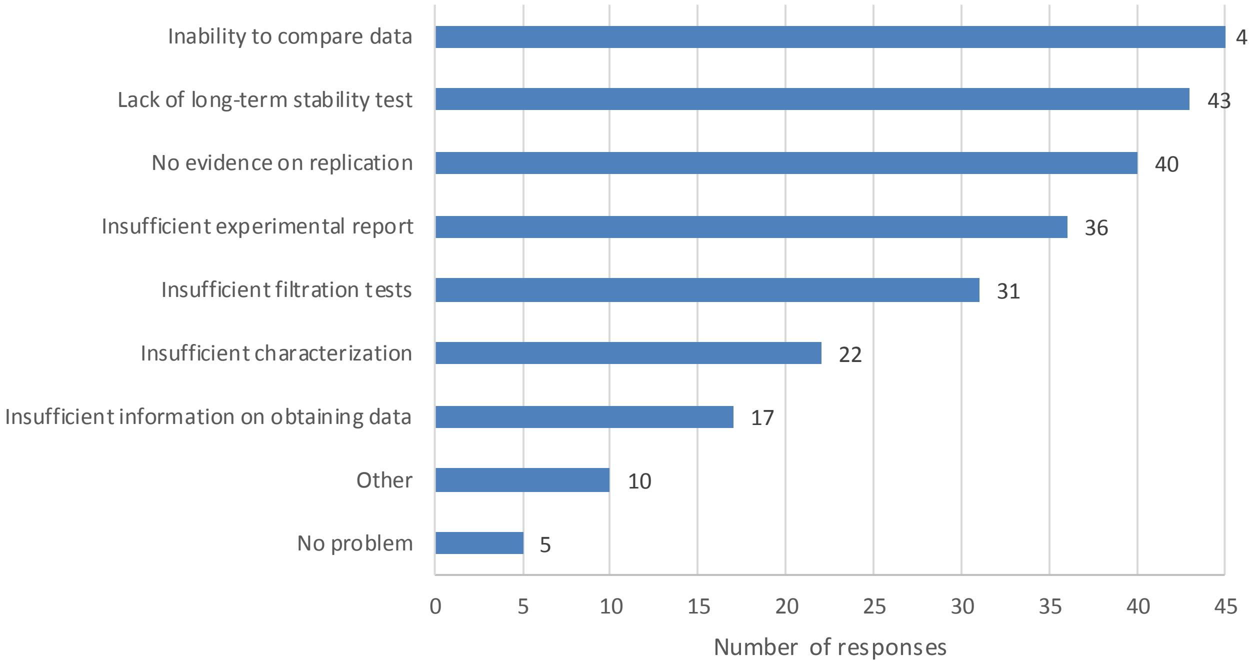

Fig. S3. Main issues within the OSN literature (survey question 2) ranked based on the number of responses (horizontal axis). The number of survey participants was 70. Each participant could select multiple responses.

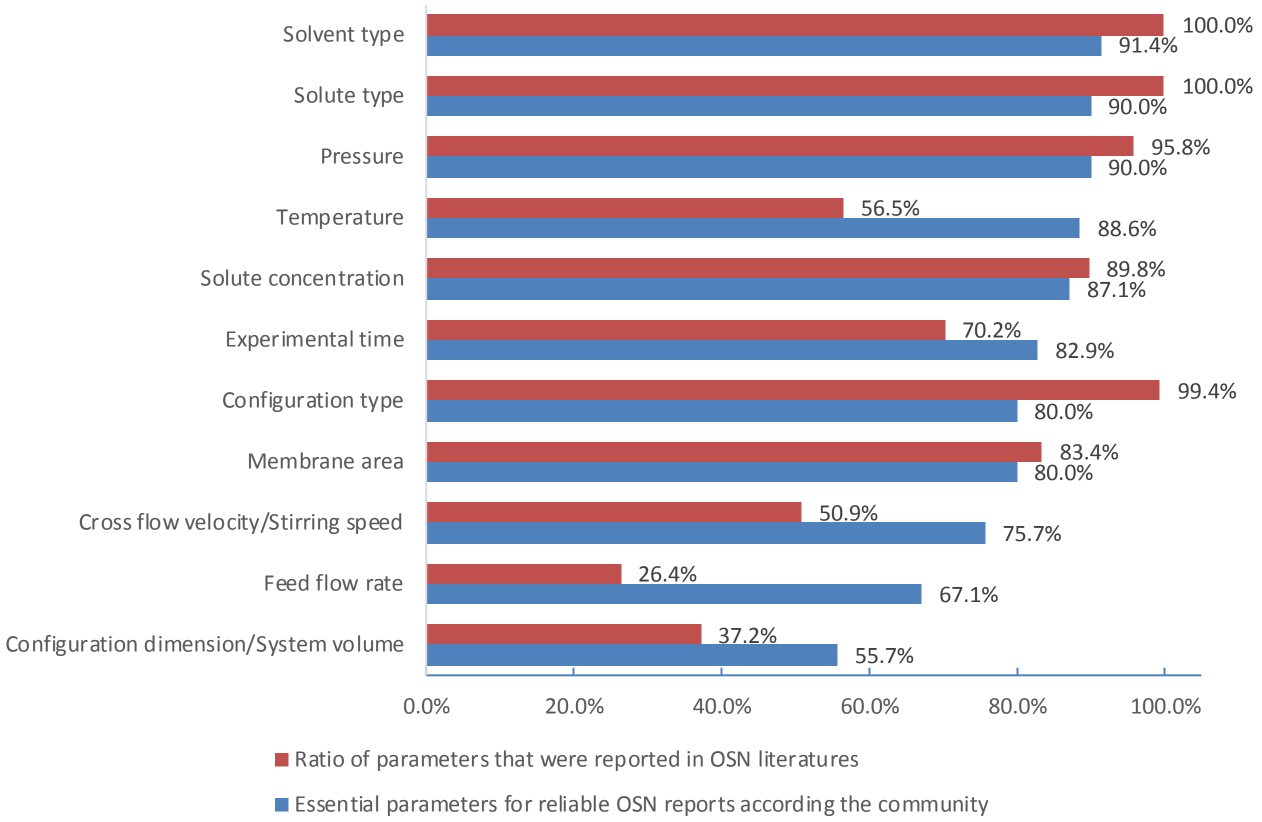

Fig. S4. Statistical breakdown of essential parameters for reproducible OSN reports based on the responses of survey participants to survey question 3 (blue) and percentages of parameters that were reported in the OSN literature (red).

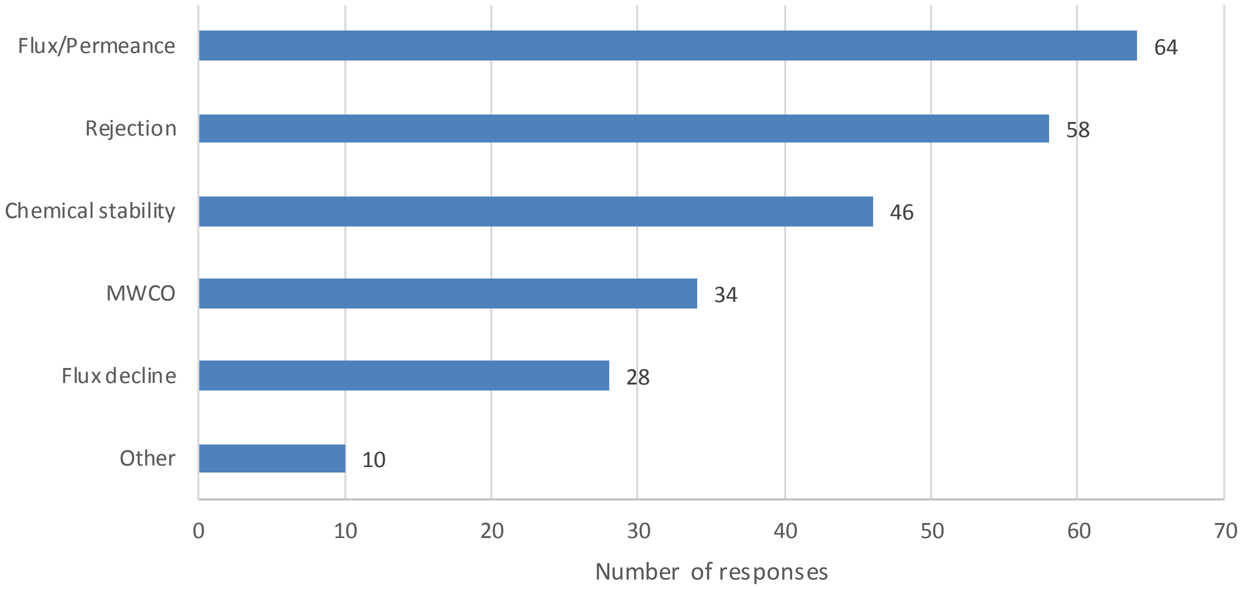

Fig. S5. Essential parameters for the characterization of membrane performance that is ranked based on the number of responses to survey question 4 (horizontal axis). The number of responses for flux/permeance includes responses that selected either flux/permeance for a single solvent, or for multiple solvents or for solutions. The number of responses for rejection includes responses that selected either rejection for a single solute or for multiple solutes. The number of survey participant was 70. Participants could select more than one response.

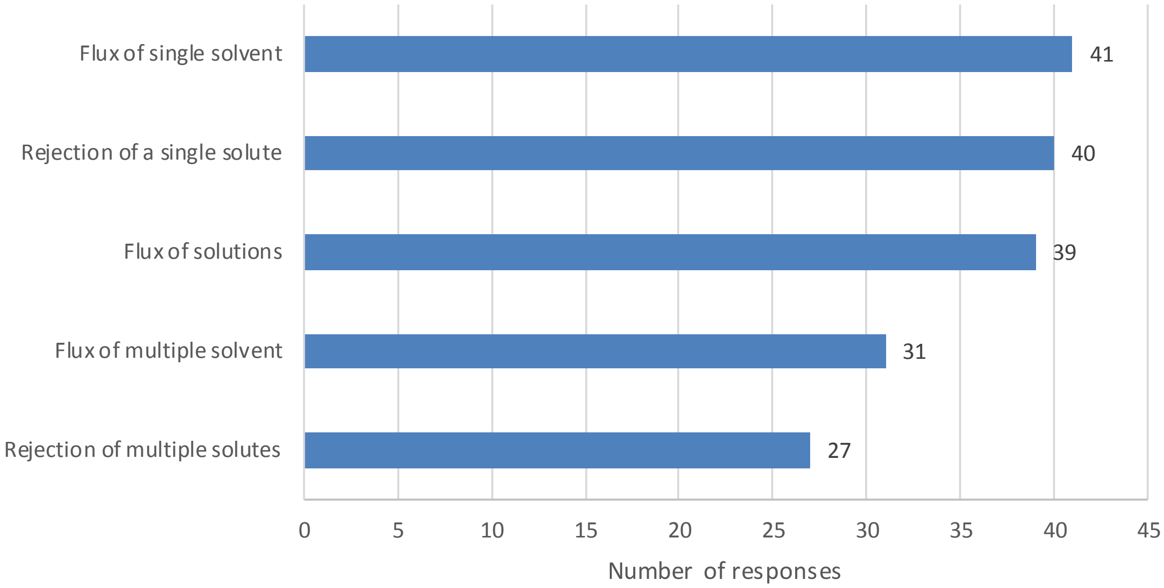

Fig. S6. Survey participants' responses to question 5 on minimum reporting requirements for membrane performance data based on the source of flux/permeance and rejection. Parameters were ranked based on the number of responses (horizontal axis). The number of survey participant was 70. Participants could select more than one response.

Results of literature analysis

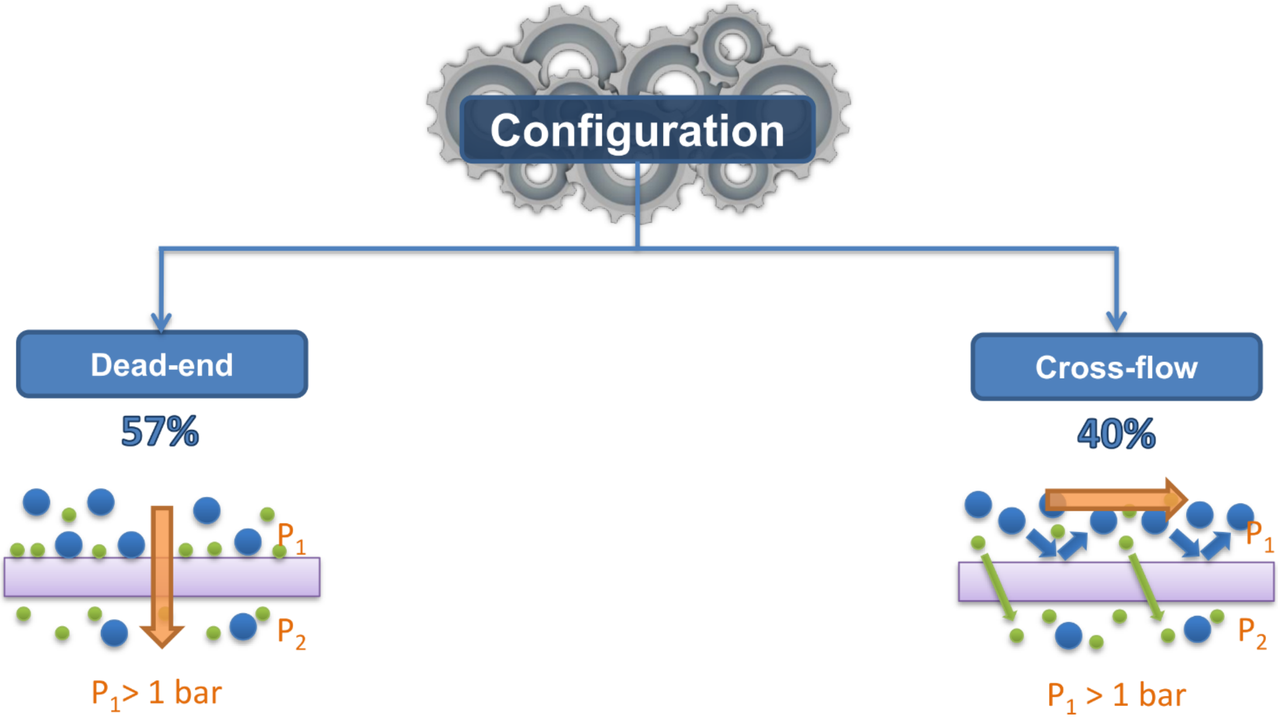

Fig. S7. Ratio of configurations commonly used within publications. 3% accounts for other configurations such as vacuum filtration, liquid-liquid phase separator.

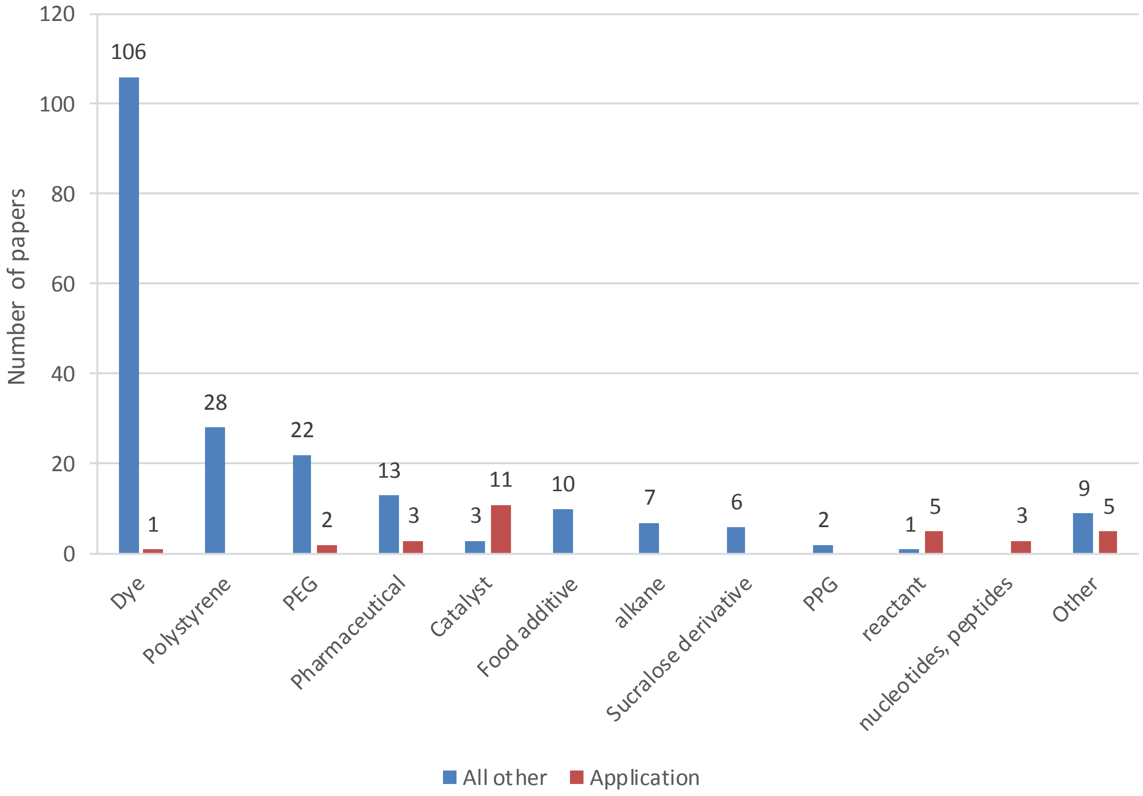

Fig. S8. Different types of solute used for membrane screening or for a case study within the OSN literature. Solutes were ranked based on the number of papers using them. The total number of solutes were 237.

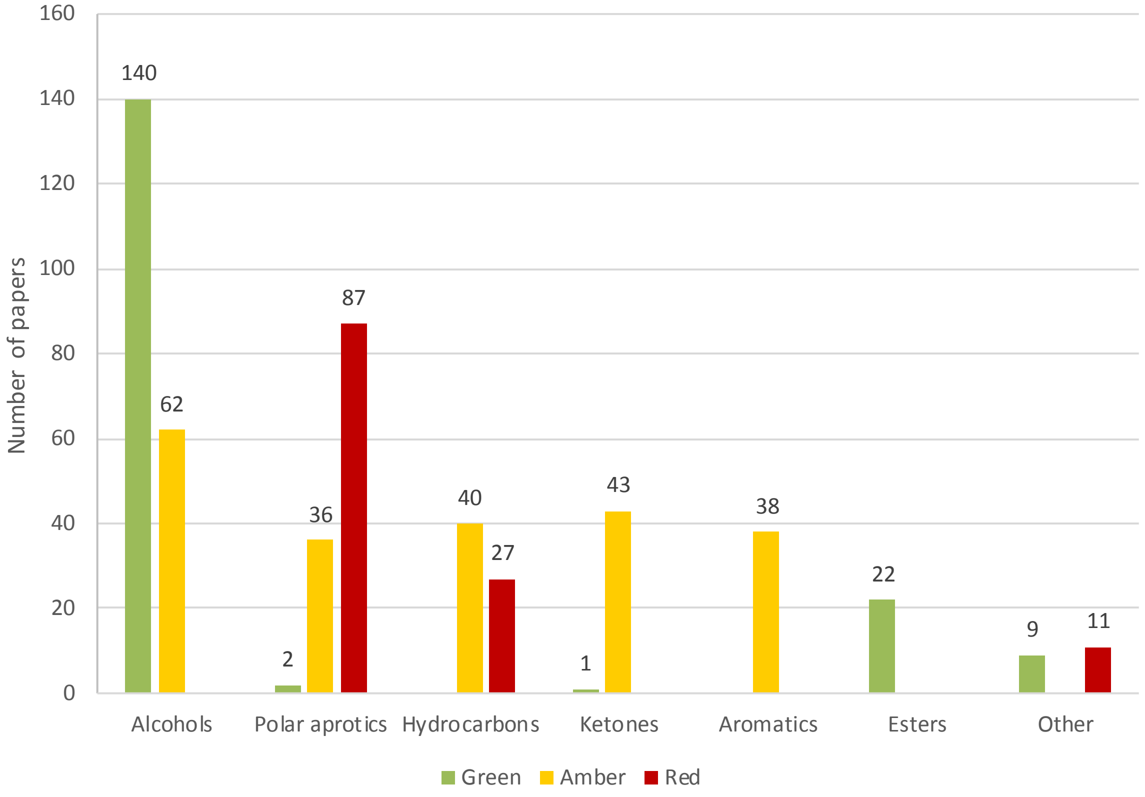

Fig. S9. Different types of green, amber and red solvents used within the OSN literature ranked based on the number of papers that employed the solvent. The categorization of solvents was carried out based on the GSK solvent selection guide.[1] The total number of solvents tested in the surveyed papers was 518.

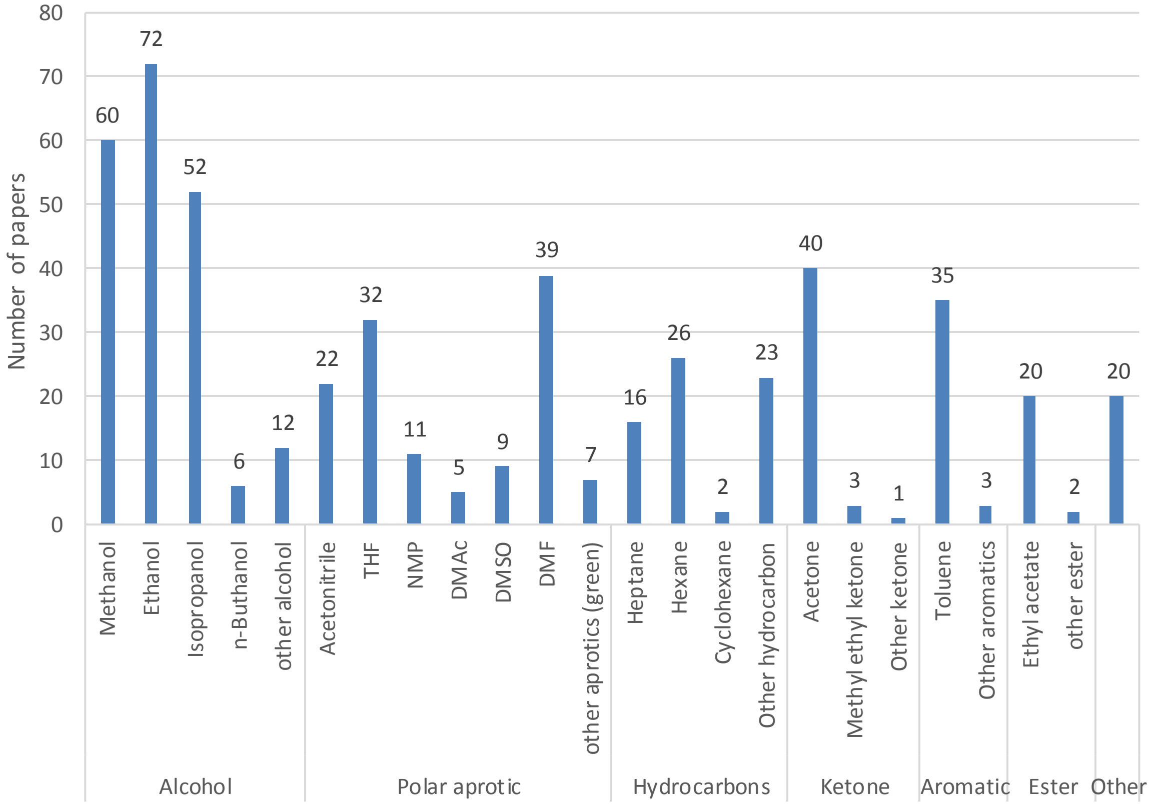

Fig. S10. Number of different solvents used within the OSN literature. The total number of solvents tested in the surveyed papers was 518.

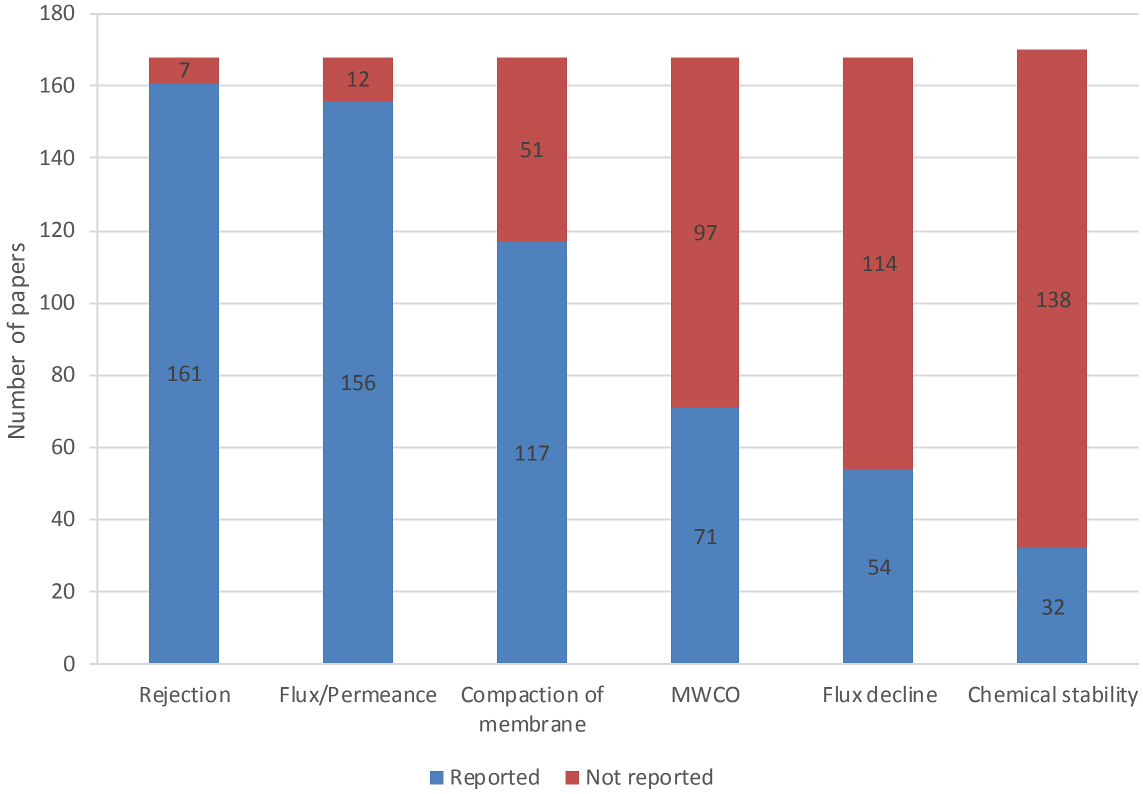

Fig. S11. Membrane performance parameters that were reported (blue) and were not (red) reported in OSN papers. Parameters are ranked based on the number of papers (vertical axis) that reported the information.

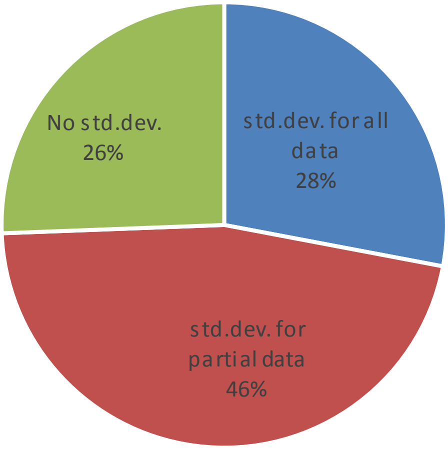

Fig. S12. Ratio of OSN papers that reported standard deviation (std.dev.) for all, for partial membrane data and papers in which std.dev. was not provided. Number of analyzed papers was 168. The remaining papers (9) did not perform filtration tests, therefore were not analyzed.

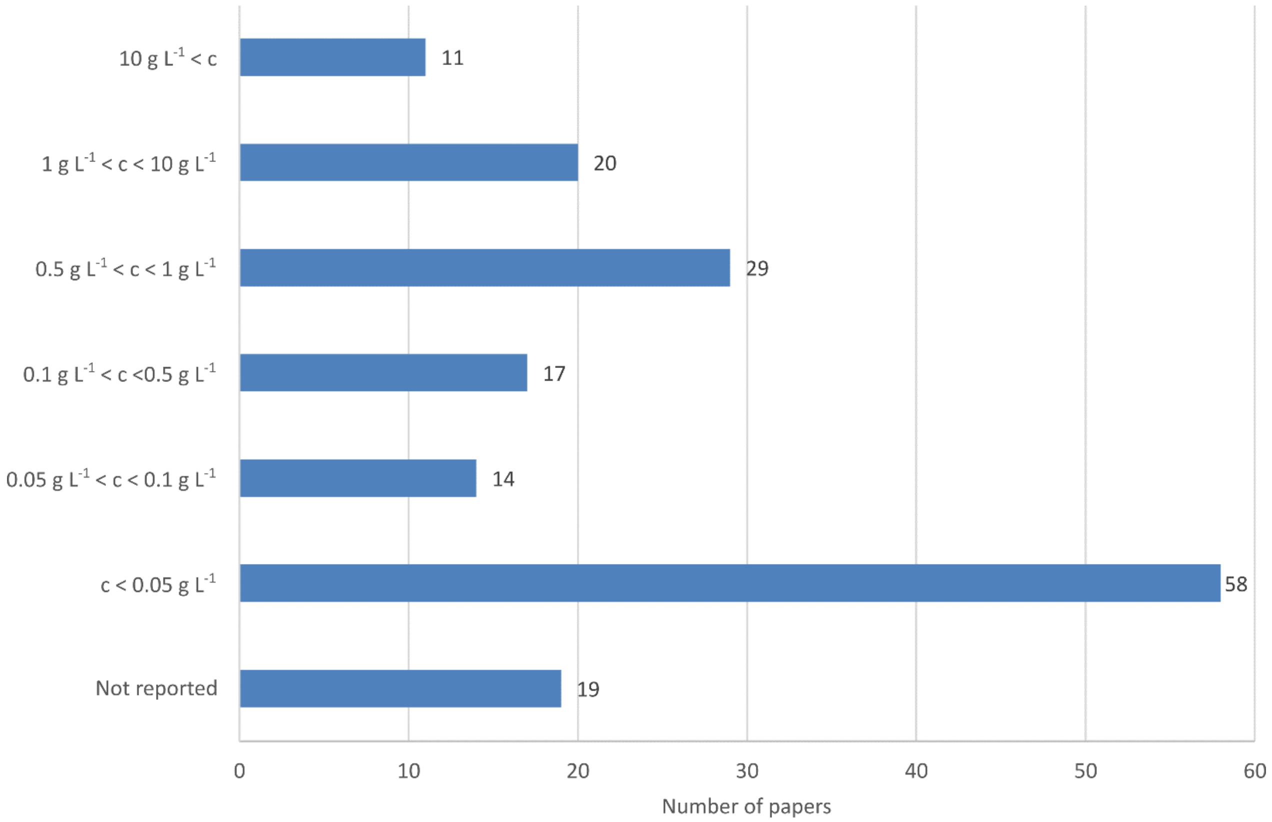

Fig. S13. Number of OSN papers (horizontal axis) divided based on the employed concentration. Total number of papers were 168. The other papers (9) did not perform filtration tests, therefore were not analyzed.

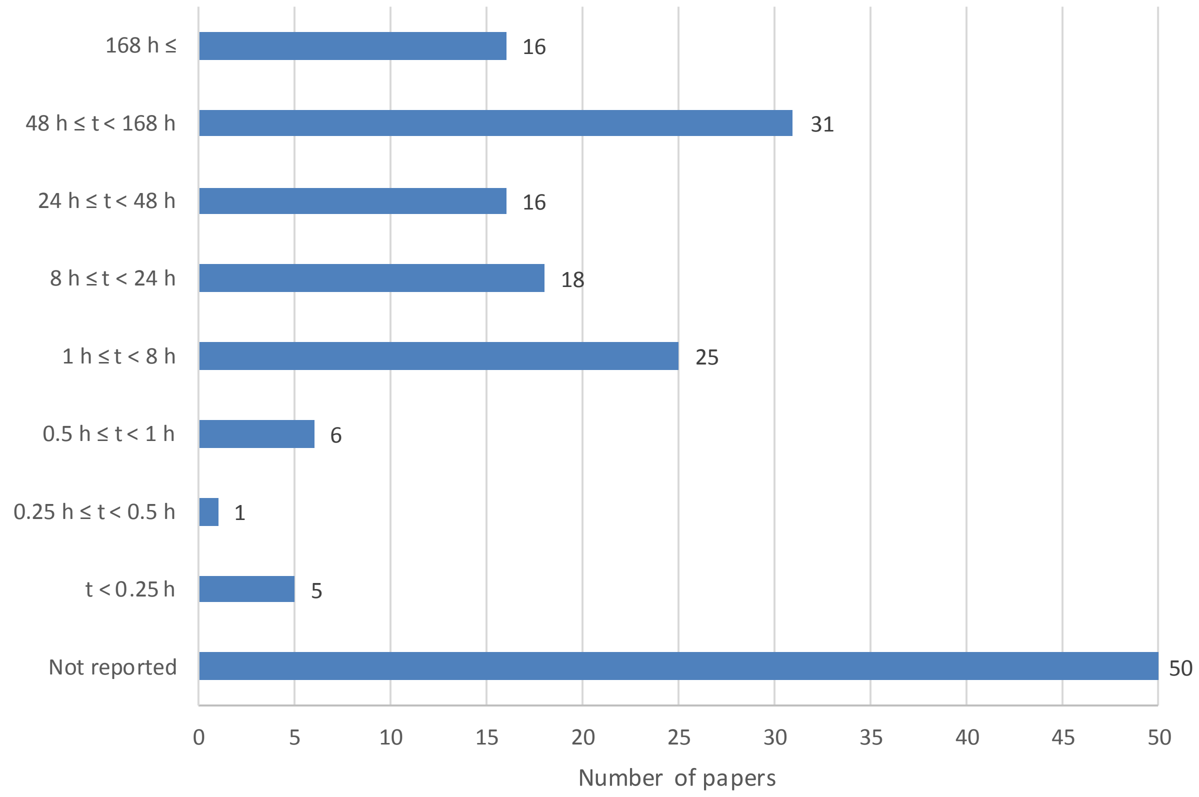

Fig. S14. Number of OSN papers (horizontal axis) divided based on the employed experimental time. Total number of papers were 168. The other papers (9) did not perform filtration tests, therefore were not analyzed.

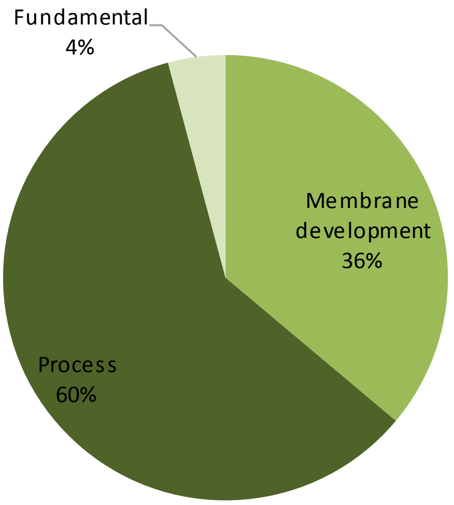

Fig. S15.The ratio of different types of papers, namely, membrane development, process and fundamental among green-focused OSN papers. Results were obtained from analyzing 72 green-focused papers.

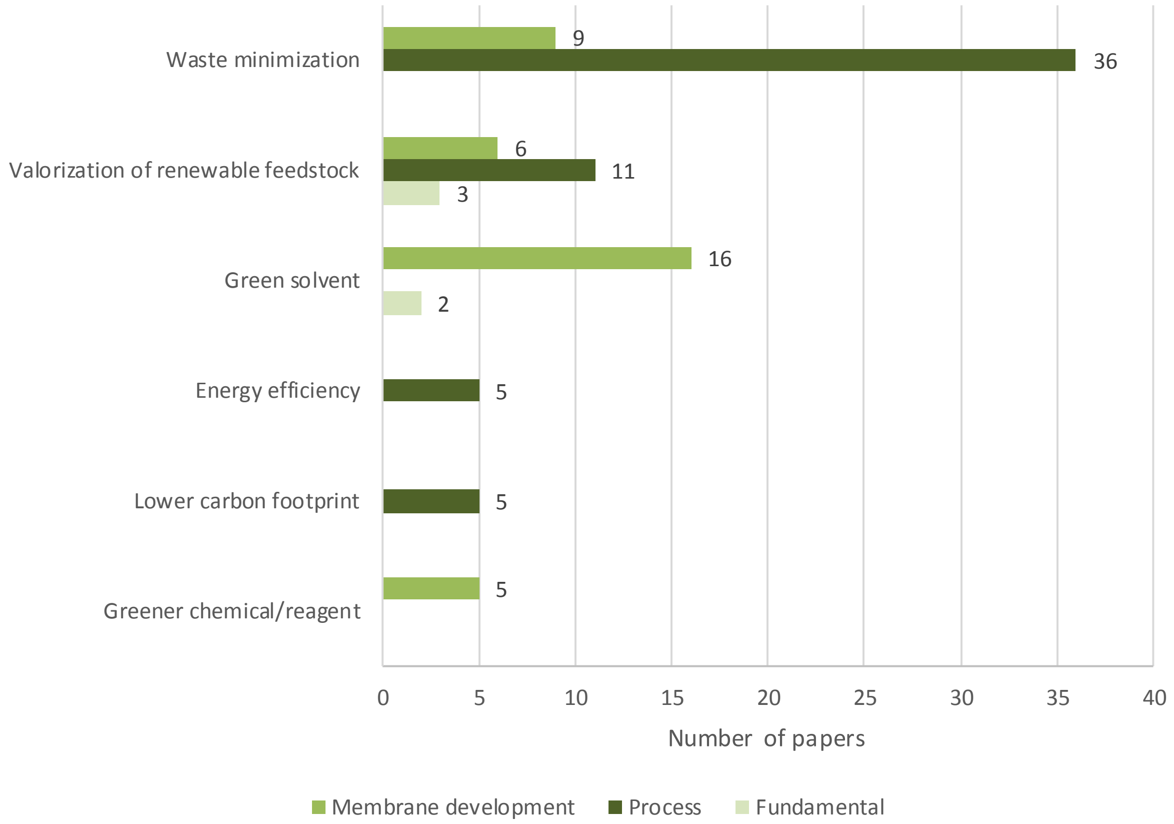

Fig. S16. Strategies to improve the sustainability of membrane fabrication and processes ranked based on the number of green-focused papers (horizontal axis) that have employed that method. Results were obtained from analyzing 72 green-focused papers. A paper can appear in more than one category.

Table S1. Summary of proposed standard protocol for testing membrane performance.

Category	**Livingston et al.**[2]	**Wessling et al.**[3]
Solutes	• Any series of oligomers • Three similar sized solutes with different physicochemical characteristics (not specified)	• Series of *n*-alkanes: $C_{10}H_{22}$ to $C_{36}H_{74}$ • Sucrose octaacetate • Series of polystyrene oligomers, poly(methyl methacrylate)
Solvents	One solvent (not specified) of each type: • non-polar • polar protic • mild polar aprotic • strong polar aprotic	• ethanol • isopropanol • butanone • ethyl acetate • toluene • *n*-heptane
Comparable results for different systems and analytical methods	Not tested	Yes

Details of materials characterization methods for OSN membranes

The goals of characterization of new or modified membrane materials are to allow synthesis, structure and performance to be interrelated; to assess membranes' suitability for their operating environments; and to ensure that the materials characteristics are uniform. Almost all materials testing is done under conditions that do not reflect how membranes are used: in solvent and under pressure. Nonetheless, certain techniques are essential to carry out when reporting research on new membrane materials.

Visible-light analyses readily allow the surface macrostructures to be examined in a wet state. They can be used to study matrix components with lateral dimensions larger than ~1 µm (e.g., flakes of graphene oxide). With appropriate refractive index matching or multiphoton imaging, some information can be obtained from the membranes' interior.[4] Scanning confocal microscopy allows for three-dimensional reconstruction of a porous network.[5] However, the cross-sectional area of the pores responsible for molecular separation are ~100 × below the spatial resolution. The related technique of white-light interferometry allows nanometer-scale axial resolution, but still has diffraction-limited lateral resolution of ~1 µm.

Electron microscopy offers higher spatial resolution. Conventional scanning electron microscopy (SEM) and transmission electron microscopy (TEM) require samples to be dry and have no outgassing components. Desolvation changes a membrane's structure:[6] a 10 nm pore in a polymer membrane will experience ~40 MPa of Laplace pressure when a membrane is dried. Despite this shrinkage, SEM is almost always used to determine membrane thickness and microstructure from cross-sections/fracture surfaces. SEM, coupled with X-ray energy dispersive spectroscopy (EDS), has been used to measure the distribution of components in MMMs.[6] The <10 nm pores thought to be responsible for size selectivity cannot be imaged by SEM either because of pore collapse, electron probe broadening or sample preparation (e.g., metal coating).

TEM requires electron-transparent specimens (<~100 nm thick), so membranes must be sectioned[7] or separated from supports,[8] but has been used to analyze the microporous region of OSN membranes.[6-8]

Environmental SEM (ESEM) circumvents some of these problems by using differential pumping that allows the sample being imaged to be imaged in ~1 kPa gas pressure. This is comparable to the vapor pressure of some organic solvents, so the membrane can be imaged wet and dried in situ. The spatial resolution of ESEM under these conditions is lower than that of conventional SEM but is still finer than visible-light microscopy and approaches the size of the membranes' mesopores. ESEMs typically image in water vapor so the microscope's gas handling system needs to be adapted to allow non-aqueous liquids. This technique has not been widely adopted by the membrane community.[4, 6]

Atomic force microscopy (AFM) provides images of membrane surfaces with a lateral resolution comparable to ESEM and sub-nanometer height resolution. AFM can be run in liquid, but no papers to date report using liquid cell AFM for membrane analysis. Pores cannot be imaged with this technique because the shape of the AFM probe's tip is convoluted with the surface topology and because the tip attracts a water meniscus running under ambient humidity. AFM can be combined with IR spectroscopy to allow superresolution mapping of the surface groups on membranes.[9, 10]

AFM's greatest value to membrane studies is that it provides a necessary correction to contact angle methods for determining surface wettability. Drop-shape analysis is frequently applied to study the surface energy and wettability of membranes, which influence the flow of solvent through them. In cases where a solvent wets a rough membrane surface rather than trapping air (Wenzel-type behavior), contact angles will suggest that hydrophilic surfaces are more hydrophilic than an untextured surface is. That is, the rough hydrophilic surface will have a lower contact angle than a flat surface made of the same material. (Conversely, rough hydrophobic surfaces will have higher contact angles than their flat counterparts.) AFM and white-light interferometry allow the roughness ratio, r, and thus the "true" contact angle and surface energy to be calculated:

$$r = \frac{\text{actual surface area}}{\text{projected surface area}} = S_{dr} + 1 \qquad \textbf{(Eq. S8)}$$

$$\cos \vartheta_{rough} = r \cos \vartheta_{flat} \qquad \textbf{(Eq. S9)}$$

where S_{dr} is the developed interfacial area ratio (an area roughness parameter determined by white-light interferometry), ϑ_{rough} is the contact angle measured on the textured surface and ϑ_{flat} is the contact angle measured on a flat surface. The effect of roughness on wettability and thus surface energy never appears to be accounted for although this would affect any modelling of flow through the membrane.

The amount and type of spectroscopic characterization of OSN membranes depends on the membrane type. For single-component polymer or ceramic membranes, vibrational spectroscopy (e.g., infrared absorption and Raman scattering) is useful for checking that the chemical functionality of a membrane matches expectations. Infrared spectra of solid films are often acquired using attenuated total reflectance (ATR), in which the evanescent light wave propagating inside the probe tip analyses the top 1–10 µm of the sample. Raman scattering is limited by absorption of the incident laser light. The surface sensitivity of these two techniques makes them essential when analyzing MMMs and TFC membranes. These techniques can generate maps of chemical functionality and thus homogeneity of the membrane surface. Raman analysis may be impossible in membranes that fluoresce.

X-ray photoelectron spectroscopy (XPS), also known as ESCA, gives the atomic composition of the top few nanometers of a surface held in ultrahigh vacuum. The binding energies associated with a particular element shift depending on how they're bonded, so some chemical information can be deduced. Surface contamination makes quantitative analysis of carbon in organic membranes nearly impossible with special handling or variable-angle analysis. The technique is well suited to detecting the presence of additions in MMMs and to the composition of the top layer of TFC membranes, providing they do not change significantly on drying (e.g., cracking and exposing fracture surfaces to the analysis). Deconvoluting the C 1s peak of membranes incorporating sp^2-rich carbon nanomaterials like nanotubes or graphene need to consider the asymmetry from plasmon absorption of those materials. The concentration of carbon bonded to other atoms such as oxygen will be overestimated if an asymmetric sp^2 component is not included.

The dimensional stability of polymer membranes needs to be measured through swelling tests in a range of process solvents. These solvents should include those that vary in polarity, are protic and aprotic, and aliphatic or aromatic. The swelling is likely to be anisotropic, so both the change in width and thickness need to be measured. Surface analyses of TFC membranes and MMMs need to be run after swelling tests. The dry mass of a polymer-based membrane before and after soaking should be checked to assess the mass of leachable components. The crosslink density of the membrane can be calculated from the Flory-Rehner Eq..[11]

Thermogravimetric analysis (TGA) and differential thermal analysis (DTA, the change in mass with temperature) are most useful for membranes made from new polymers, TFC membranes and MMMs. T_{d10}, the point at which 10% of the initial membrane mass (excluding solvent loss) is lost on heating, is a proxy for maximum service temperature, especially in heavily crosslinked membranes. For membranes containing a mixture of materials, distinct steps in the mass loss curve provide a measurement of the fraction of each component.

The glass transition temperature, T_g, of polymer-based membranes with few or no crosslinks sets a lower service temperature than T_{d10}. Above T_g, membrane polymers become rubbery and have lower dimensional stability. T_g is most commonly measured by differential scanning calorimetry (DSC). DSC has been used to approximate pore-size distributions in microporous membranes.[12] In thermoporometry, solvated porous materials are snap frozen, then the temperature of the material is increased from temperatures well below the solvent's standard melting point and endothermic events indicative of melting solvent are recorded as a function of temperature.[13] The change in freezing point in cylindrical pores is inversely proportional to pore diameter, following the Gibbs–Thomson Eq.. Comparable measurements of the temperature-dependence of the solid–liquid transition can be carried out by NMR.[14]

Mechanical testing measures membrane materials' resistance to the fluid flow forces in OSN. Tensile testing is generally run on dry samples and membranes are not typically subjected to tension, so is not relevant to membrane material performance. Instead, membranes' properties under shear and compression are more relevant. Bend tests measure elastic modulus, can be run on wet membranes, and measure how much a membrane will distort when compressed against a support. These measurements can be carried out by nanoindentation on membranes with a uniform cross-section (i.e., *not* ISA or TFC membranes).

The shear stress on a membrane surface is proportional to a fluid's dynamic viscosity and its flow rate. In a typical cross-flow setup, this is predicted to be <1 Pa for laminar flow. For membranes with sub-micrometer active layers, dynamic mechanical analysis (DMA) should be used on wet membranes to determine the strain to failure. DMA on TFC membranes may reveal separate failure strain values for the surface layer and the support. Post-mortem microscopic analysis is essential.

Essential materials characterization list

Table S2. Essential materials characterization needed for membranes for OSN according to membrane type. Abbreviations: ISA = integrally skinned asymmetric (membrane), MMM = mixed-matrix membrane, TFC = thin-film composite (membrane), inorg = inorganic/ceramic (membrane). Key: ++ = essential, + = desirable, o = optional, — = unnecessary.

Technique	Information provided and caveats	Examples	ISA	MMM	TFC	inorg
Visible-light ("optical") microscopy, differential interference contrast (DIC) and confocal microscopy	Surface structure, <1 µm lateral resolution, wide field of view, can be done wet, inclusions may be visible, no nanoscale roughness or porosity, some depth information possible (especially with multiphoton) at ~1 µm axial resolution with confocal	[4, 5, 15]	+	++	++	+
Scanning electron microscopy (SEM)	Surface analysis, membrane thickness, dry, nanometric resolution	common	++	++	++	+
Environmental SEM	Solvated, <100 nm resolution, surface analysis	[6]	o	o	+	—
(Scanning) transmission electron microscopy (STEM/TEM)	Dry, nanometer resolution, requires <0.1 µm samples, can give pore distribution	[6-8, 16]	o	o	o	—
Atomic force microscopy (AFM)	Basal surface topology/roughness, wet or dry, comparable spatial resolution to ESEM	common [a]	+	++	++	+
Liquid contact angle	Surface wettability, must be corrected for surface roughness	common [b]	++	++	++	++
Vibrational spectroscopy (IR/Raman)	Chemical functionality, fouling, surface sensitive (1–10 µm) with ATR	[15, 17, 18]	o	++	++	o
X-ray diffraction (XRD)	Membrane crystallinity, matrix size and aggregation	[16, 18, 19]	+	+	+	+
X-ray photoelectron spectroscopy (XPS)	Atomic composition, some bonding information, top ~10 nm	[20, 21, 22]	o	+	+	o
Positron annihilation lifetime spectroscopy (PALS)	Free-volume measurements, dry	[16, 23, 24]	o	o	o	+
Solvent swelling/stability	Solvent compatibility, dimensional stability, leachable fraction	[25, 26, 27]	++	++	++	—
Thermogravimetric analysis (TGA) and differential thermal analysis (DTA)	Service temperature, mass fraction of membrane components	[15, 27, 28]	+	++	++	—
Differential scanning calorimetry (DSC)	Glass transition temperature, pore size distribution	[28, 29, 30]	+	+	+	—
Dynamic mechanical analysis (DMA)	Shear stability of membrane surface	[31]	++	+	++	—
Tensile testing	Elastic modulus, tensile strain to failure, toughness in tension, dry	[31, 32, 33]	—	—	—	—
Nanoindentation	Elastic modulus, flexural modulus, uniform cross-section	[15, 28, 34]	o	+	o	+
Bend testing	Elastic (flexural) modulus, distortion on compression, can be wet	unknown	+	++	++	o
Nitrogen porosimetry ("BET")	Pore volume, pore size distribution, dry	[17, 19]	–	o	o	+

[a] Has been combined with infrared spectroscopy for superresolution mapping.[9,10] [b] Effects of surface roughness typically not considered.[35, 36]

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