

Supporting Information

Using the Green Solvent Dimethyl Sulfoxide to Replace Traditional Solvents Partly and Fabricating PVC/PVC-g-PEGMA Blended Ultrafiltration Membranes with High Permeability and Rejection

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CONTENT

Figure S1. Morphologies and performances of the two membranes: (a) the surface morphology of membrane fabricated by evaporation for 30 s (20 K $\times$), (b) the surface morphology of membrane fabricated without evaporation (20 K $\times$), (c) the water contact angles of the two membranes, (d) the fluxes performance of the two membranes and (e) the antifouling properties and SA rejection rates of the two membranes

Table S1. Statistics of the Pore Size Properties and Thicknesses of the Two Membranes

Table S2. Solubility Parameter Component Group Contributions Method (Hoftyzer – Van Krevelen)

Table S3. Addition of the Group Contributions of PEGMA

Table S4. δ_d , δ_p and δ_h Values and Densities of Some Solvents

Table S5. Comparison on the Pure Water Flux, SA Rejection and FRR Ratio of the Membranes in this Study and from Literatures

The number of pages: 18 (S1-S18)

The number of tables: 5 (Table S1-Table S5)

The number of figures: 1 (Figure S1)

The Performances of PVC/PVC-g-PEGMA Membranes Using the Solvent NMP/THF/DMSO=4/3/3 With 0 s or 30 s Evaporation Time. We fabricated PVC/PVC-g-PEGMA membranes with the solvent NMP/THF/DMSO=4/3/3 and the evaporation time was 0 s, and compared with the membrane fabricated by evaporation for 30 s (M5). The results are shown in **Figure S1**. Based on the surface SEM images, the statistics of the pore size properties and membrane thicknesses are in **Table S1**.

From **Figure S1** and **Table S1**, we known that the hydrophilicities of the two membrane were almost the same. However, the membrane without evaporation had much larger pore size, leading to lower sodium alginate (SA) rejection rates (93.0 % for membrane of 30 s evaporation and 73.9 % of membrane without evaporation). Although larger pore size, the pure water flux of membrane without evaporation was much lower than membrane with 30 s evaporation time, which can be explained by larger thickness and large resistance. The four indexes: FRR (flux recovery ratio), DR_r (reversible flux decline ratio), DR_{ir} (irreversible flux decline ratio) and DR_t (total flux decline ratio) showed that the membrane without evaporation had a little better antifouling property.

In order to fabricate membrane with high pure water flux and high SA rejection, we chose to add the step of 30 s evaporation time before immersing.

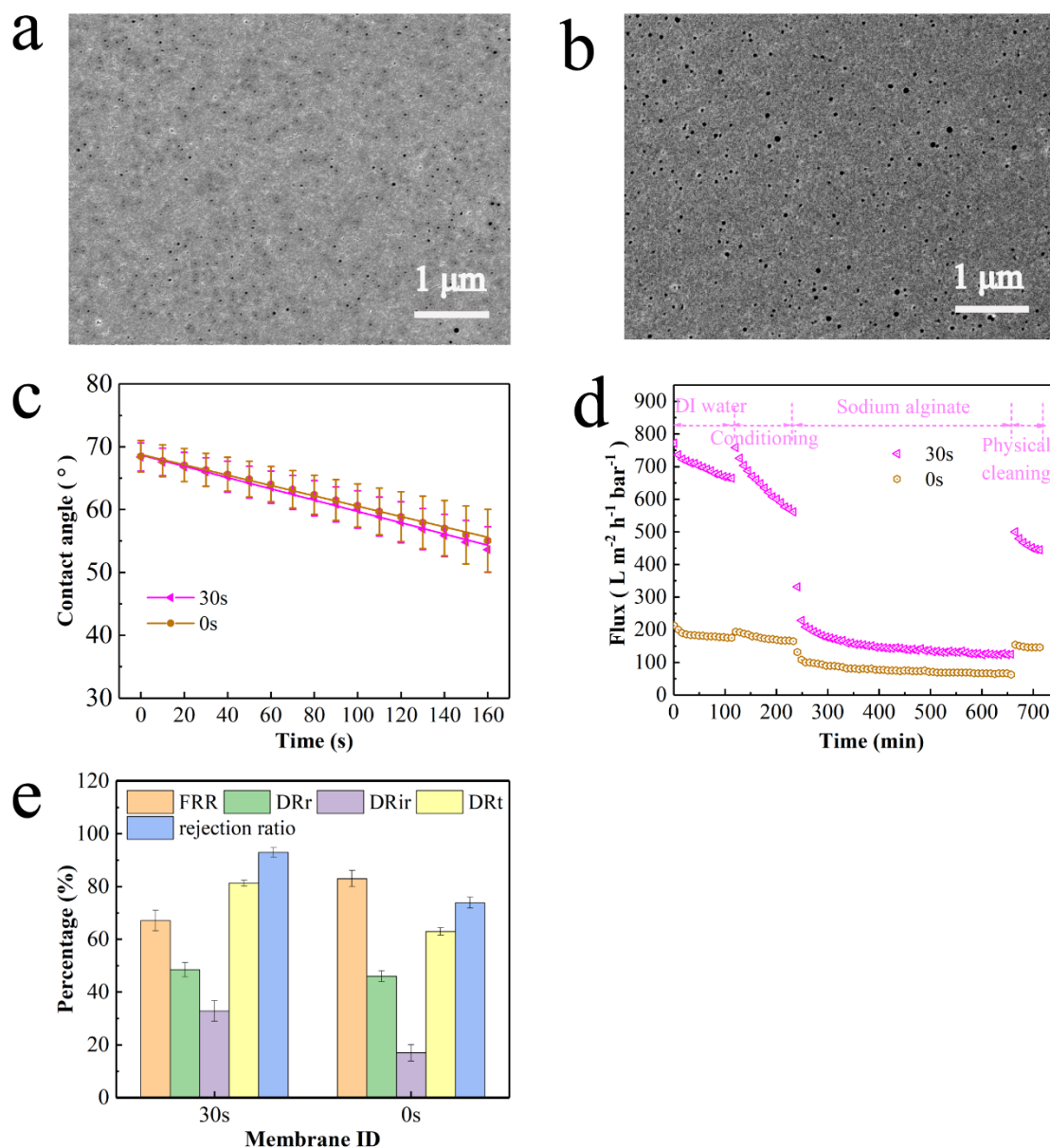


Figure S1. Morphologies and performances of the two membranes: (a) the surface morphology of membrane fabricated by evaporation for 30 s (20 K $\times$), (b) the surface morphology of membrane fabricated without evaporation (20 K $\times$), (c) the water contact angles of the two membranes, (d) the fluxes performance of the two membranes and (e) the antifouling properties and SA rejection rates of the two membranes

Table S1. Statistics of the Pore Size Properties and Thicknesses of the Two Membranes

Evaporation time	D _{average} (nm)	D _{max} (nm)	Pore density (m ⁻²)	Surface porosity (%)	Thickness (mm)
30 s	24	28	1.2×10 ¹³	0.84	0.108
0 s	34	39	1.0×10 ¹³	1.03	0.142

Group Contribution Method to Calculate the Solubility Parameter of PEGMA. Based on Hansen solubility parameters theory, the total solubility parameter δ_t is divided into 3 parts: δ_d , δ_p and δ_h , which quantitatively represent the dispersion parameter(δ_d), the polar parameter (δ_p) and the hydrogen bonding parameter (δ_h).¹

$$\delta_t^2 = \delta_d^2 + \delta_p^2 + \delta_h^2 \quad (1)$$

δ_d , δ_p and δ_h can be calculated by group contribution method using the following equations:

2

$$\delta_d = \frac{\sum F_{di}}{V} \quad (2)$$

$$\delta_p = \frac{\sqrt{\sum F_{pi}^2}}{V} \quad (3)$$

$$\delta_h = \frac{\sqrt{\sum E_{hi}}}{V} \quad (4)$$

The values of F_{di} , F_{pi} and E_{hi} of each structural group of PEGMA are listed in **Table S2**. The calculation procedures can figure to **Table S3** and equation (5), (6), (7) (8) and (9).

Table S2. Solubility Parameter Component Group Contributions Method (Hoftyzer – Van Krevelen) ²

Structural group	F _{di} $(\text{MJ/m}^3)^{1/2} \cdot \text{mol}^{-1}$	F _{pi} $(\text{MJ/m}^3)^{1/2} \cdot \text{mol}^{-1}$	E _{hi} (J/mol).
-CH ₃	420	0	0
-COO-	390	490	7000
-O-	100	400	3000
-CH ₂ -	270	0	0
CH ₂ =	400	0	0
-C- 	70	0	0

Table S3. Addition of the Group Contributions of PEGMA

Structural group	F _{di}	F _{pi} ²	E _{hi}
2 -CH ₃	840	0	0
-COO-	390	240100	7000
9 -O-	900	1440000	27000
18 -CH ₂ -	4860	0	0
CH ₂ =	400	0	0
-C- 	70	0	0
Adding	7460	1680100	34000

The molar volume V is calculated via the following equation.

$$V = \frac{M_n}{\rho} = \frac{500 \text{ g/mol}}{1.08 \text{ g/mL}} = 462.9630 (\text{cm}^3/\text{mol}) \quad (5)$$

According to Eq (2), (3) and (4):

$$\delta_d = \frac{\sum F_{di}}{V} = 7460 / 462.9630 = 16.1176 (\text{MJ}/\text{m}^3)^{1/2} \quad (6)$$

$$\delta_p = \frac{\sqrt{\sum F_{pi}^2}}{V} = \sqrt{1680100} / 462.9630 = 2.7998 (\text{MJ}/\text{m}^3)^{1/2} \quad (7)$$

$$\delta_h = \frac{\sqrt{\sum E_{hi}}}{V} = \sqrt{34000} / 462.9630 = 8.5697 (\text{MJ}/\text{m}^3)^{1/2} \quad (8)$$

According to Eq (1):

$$\delta_t = \sqrt{\delta_d^2 + \delta_p^2 + \delta_h^2} = \sqrt{16.1176^2 + 2.7998^2 + 8.5697^2} = 18.47 (\text{MJ}/\text{m}^3)^{1/2} \quad (9)$$

So, the total solubility parameter δ_t for PEGMA is $18.47 (\text{MJ}/\text{m}^3)^{1/2}$.

Calculation of Hansen Solubility Parameters of Mixed Solvents. The δ_d , δ_p and δ_h values and densities of NMP, DMSO, DMAc and THF are list in **Table S4**.

Table S4. The δ_d , δ_p and δ_h Values and Densities of Some Solvents

	δ_d	δ_p	δ_h	density
	$(\text{MJ}/\text{m}^3)^{1/2}$	$(\text{MJ}/\text{m}^3)^{1/2}$	$(\text{MJ}/\text{m}^3)^{1/2}$	(g/cm^3)
NMP	18.0	12.3	7.2	1.028
DMSO	18.4	16.4	10.2	1.096
DMAc	16.8	11.5	10.2	0.937
THF	16.8	5.7	8.0	0.889

δ_d , δ_p and δ_h values of binary mixed solvents can be calculated in the following way. ¹

First, calculate the volume fraction of NMP, DMSO according to the mass fraction using equation (10).

$$(\text{Vol.Fraction})_1 = \frac{\left(\frac{W_t.\text{Fraction}}{\text{Density}} \right)_1}{\left(\frac{W_t.\text{Fraction}}{\text{Density}} \right)_1 + \left(\frac{W_t.\text{Fraction}}{\text{Density}} \right)_2} \quad (10)$$

Second, calculate the values of δ_d , δ_p and δ_h of mixed solvents, following equation (11).

$$\begin{aligned} \delta_d &= (\text{Vol.Fraction})_1 \times \delta_{d1} + (\text{Vol.Fraction})_2 \times \delta_{d2} \\ \delta_p &= (\text{Vol.Fraction})_1 \times \delta_{p1} + (\text{Vol.Fraction})_2 \times \delta_{p2} \\ \delta_h &= (\text{Vol.Fraction})_1 \times \delta_{h1} + (\text{Vol.Fraction})_2 \times \delta_{h2} \end{aligned} \quad (11)$$

For solvent NMP/DMSO=7/3:

$$\begin{aligned} (\text{Vol.Fraction})_{\text{NMP}} &= \frac{\left(\frac{W_t.\text{Fraction}}{\text{Density}} \right)_{\text{NMP}}}{\left(\frac{W_t.\text{Fraction}}{\text{Density}} \right)_{\text{NMP}} + \left(\frac{W_t.\text{Fraction}}{\text{Density}} \right)_{\text{DMSO}}} = \frac{\frac{0.7}{1.028}}{\frac{0.7}{1.028} + \frac{0.3}{1.096}} = 0.713 \\ (\text{Vol.Fraction})_{\text{DMSO}} &= \frac{\left(\frac{W_t.\text{Fraction}}{\text{Density}} \right)_{\text{DMSO}}}{\left(\frac{W_t.\text{Fraction}}{\text{Density}} \right)_{\text{NMP}} + \left(\frac{W_t.\text{Fraction}}{\text{Density}} \right)_{\text{DMSO}}} = \frac{\frac{0.3}{1.096}}{\frac{0.7}{1.028} + \frac{0.3}{1.096}} = 0.287 \end{aligned}$$

$$\begin{aligned} \delta_d &= (\text{Vol.Fraction})_{\text{NMP}} \times \delta_{d1} + (\text{Vol.Fraction})_{\text{DMSO}} \times \delta_{d2} \\ &= 0.713 \times 18 + 0.287 \times 18.4 \\ &= 18.11 \left(\text{MJ}/\text{cm}^3 \right)^{1/2} \\ \delta_p &= (\text{Vol.Fraction})_{\text{NMP}} \times \delta_{p1} + (\text{Vol.Fraction})_{\text{DMSO}} \times \delta_{p2} \\ &= 0.713 \times 12.3 + 0.287 \times 16.4 \\ &= 13.48 \left(\text{MJ}/\text{cm}^3 \right)^{1/2} \end{aligned}$$

$$\begin{aligned}
\delta_h &= (\text{Vol.Fraction})_{\text{NMP}} \times \delta_{h1} + (\text{Vol.Fraction})_{\text{DMSO}} \times \delta_{h2} \\
&= 0.713 \times 7.2 + 0.287 \times 10.2 \\
&= 8.06 \left(\text{MJ}/\text{cm}^3 \right)^{1/2}
\end{aligned}$$

δ_d , δ_p and δ_h values of ternary mixed solvents NMP/THF/DMSO=4/3/3 and NMP/DMAc/DMSO=4/3/3 can be calculated in the same way. For mixed solvents NMP/THF/DMSO=4/3/3:

$$\begin{aligned}
(\text{Vol.Fraction})_{\text{NMP}} &= \frac{\left(\frac{\text{W}_t.\text{Fraction}}{\text{Density}} \right)_{\text{NMP}}}{\left(\frac{\text{W}_t.\text{Fraction}}{\text{Density}} \right)_{\text{NMP}} + \left(\frac{\text{W}_t.\text{Fraction}}{\text{Density}} \right)_{\text{THF}} + \left(\frac{\text{W}_t.\text{Fraction}}{\text{Density}} \right)_{\text{DMSO}}} \\
&= \frac{\frac{0.4}{1.028}}{\frac{0.4}{1.028} + \frac{0.3}{0.889} + \frac{0.3}{1.096}} \\
&= 0.389
\end{aligned}$$

$$\begin{aligned}
(\text{Vol.Fraction})_{\text{THF}} &= \frac{\left(\frac{\text{W}_t.\text{Fraction}}{\text{Density}} \right)_{\text{THF}}}{\left(\frac{\text{W}_t.\text{Fraction}}{\text{Density}} \right)_{\text{NMP}} + \left(\frac{\text{W}_t.\text{Fraction}}{\text{Density}} \right)_{\text{THF}} + \left(\frac{\text{W}_t.\text{Fraction}}{\text{Density}} \right)_{\text{DMSO}}} \\
&= \frac{\frac{0.3}{0.889}}{\frac{0.4}{1.028} + \frac{0.3}{0.889} + \frac{0.3}{1.096}} \\
&= 0.337
\end{aligned}$$

$$\begin{aligned}
(\text{Vol.Fraction})_{\text{DMSO}} &= \frac{\left(\frac{\text{W}_t.\text{Fraction}}{\text{Density}} \right)_{\text{THF}}}{\left(\frac{\text{W}_t.\text{Fraction}}{\text{Density}} \right)_{\text{NMP}} + \left(\frac{\text{W}_t.\text{Fraction}}{\text{Density}} \right)_{\text{THF}} + \left(\frac{\text{W}_t.\text{Fraction}}{\text{Density}} \right)_{\text{DMSO}}} \\
&= \frac{\frac{0.3}{1.096}}{\frac{0.4}{1.028} + \frac{0.3}{0.889} + \frac{0.3}{1.096}} \\
&= 0.274
\end{aligned}$$

$$\begin{aligned}
\delta_d &= (\text{Vol.Fraction})_{\text{NMP}} \times \delta_{d1} + (\text{Vol.Fraction})_{\text{THF}} \times \delta_{d2} + (\text{Vol.Fraction})_{\text{DMSO}} \times \delta_{d3} \\
&= 0.389 \times 18 + 0.337 \times 16.8 + 0.287 \times 18.4
\end{aligned}$$

$$= 17.70 \left(\text{MJ}/\text{cm}^3 \right)^{1/2}$$

$$\begin{aligned}
\delta_p &= (\text{Vol.Fraction})_{\text{NMP}} \times \delta_{p1} + (\text{Vol.Fraction})_{\text{THF}} \times \delta_{p2} + (\text{Vol.Fraction})_{\text{DMSO}} \times \delta_{p3} \\
&= 0.389 \times 12.3 + 0.337 \times 5.7 + 0.287 \times 16.4
\end{aligned}$$

$$= 11.20 \left(\text{MJ}/\text{cm}^3 \right)^{1/2}$$

$$\begin{aligned}
\delta_h &= (\text{Vol.Fraction})_{\text{NMP}} \times \delta_{h1} + (\text{Vol.Fraction})_{\text{THF}} \times \delta_{h2} + (\text{Vol.Fraction})_{\text{DMSO}} \times \delta_{h3} \\
&= 0.389 \times 7.2 + 0.337 \times 8.0 + 0.287 \times 10.2
\end{aligned}$$

$$= 8.29 \left(\text{MJ}/\text{cm}^3 \right)^{1/2}$$

For mixed solvents NMP/DMAc/DMSO=4/3/3:

$$\begin{aligned}
(\text{Vol.Fraction})_{\text{NMP}} &= \frac{\left(\frac{\text{W}_t.\text{Fraction}}{\text{Density}} \right)_{\text{NMP}}}{\left(\frac{\text{W}_t.\text{Fraction}}{\text{Density}} \right)_{\text{NMP}} + \left(\frac{\text{W}_t.\text{Fraction}}{\text{Density}} \right)_{\text{DMAc}} + \left(\frac{\text{W}_t.\text{Fraction}}{\text{Density}} \right)_{\text{DMSO}}} \\
&= \frac{\frac{0.4}{1.028}}{\frac{0.4}{1.028} + \frac{0.3}{0.937} + \frac{0.3}{1.096}} \\
&= 0.396
\end{aligned}$$

$$\begin{aligned}
(\text{Vol.Fraction})_{\text{DMAc}} &= \frac{\left(\frac{\text{W}_t.\text{Fraction}}{\text{Density}} \right)_{\text{DMAc}}}{\left(\frac{\text{W}_t.\text{Fraction}}{\text{Density}} \right)_{\text{NMP}} + \left(\frac{\text{W}_t.\text{Fraction}}{\text{Density}} \right)_{\text{DMAc}} + \left(\frac{\text{W}_t.\text{Fraction}}{\text{Density}} \right)_{\text{DMSO}}} \\
&= \frac{\frac{0.3}{0.937}}{\frac{0.4}{1.028} + \frac{0.3}{0.937} + \frac{0.3}{1.096}} \\
&= 0.326
\end{aligned}$$

$$\begin{aligned}
(\text{Vol.Fraction})_{\text{DMSO}} &= \frac{\left(\frac{\text{W}_t.\text{Fraction}}{\text{Density}} \right)_{\text{DMSO}}}{\left(\frac{\text{W}_t.\text{Fraction}}{\text{Density}} \right)_{\text{NMP}} + \left(\frac{\text{W}_t.\text{Fraction}}{\text{Density}} \right)_{\text{DMAc}} + \left(\frac{\text{W}_t.\text{Fraction}}{\text{Density}} \right)_{\text{DMSO}}} \\
&= \frac{\frac{0.3}{1.096}}{\frac{0.4}{1.028} + \frac{0.3}{0.937} + \frac{0.3}{1.096}} \\
&= 0.278
\end{aligned}$$

$$\begin{aligned}
\delta_d &= (\text{Vol.Fraction})_{\text{NMP}} \times \delta_{d1} + (\text{Vol.Fraction})_{\text{DMAc}} \times \delta_{d2} + (\text{Vol.Fraction})_{\text{DMSO}} \times \delta_{d3} \\
&= 0.396 \times 18 + 0.326 \times 16.8 + 0.278 \times 18.4 \\
&= 17.72 \left(\text{MJ}/\text{cm}^3 \right)^{1/2}
\end{aligned}$$

$$\begin{aligned}
\delta_d &= (\text{Vol.Fraction})_{\text{NMP}} \times \delta_{d1} + (\text{Vol.Fraction})_{\text{DMAc}} \times \delta_{d2} + (\text{Vol.Fraction})_{\text{DMSO}} \times \delta_{d3} \\
&= 0.396 \times 12.3 + 0.326 \times 11.5 + 0.278 \times 16.4 \\
&= 13.18 \left(\text{MJ}/\text{cm}^3 \right)^{1/2}
\end{aligned}$$

$$\begin{aligned}
\delta_d &= (\text{Vol.Fraction})_{\text{NMP}} \times \delta_{d1} + (\text{Vol.Fraction})_{\text{DMAc}} \times \delta_{d2} + (\text{Vol.Fraction})_{\text{DMSO}} \times \delta_{d3} \\
&= 0.396 \times 7.2 + 0.326 \times 10.2 + 0.278 \times 10.2 \\
&= 9.01 \left(\text{MJ}/\text{cm}^3 \right)^{1/2}
\end{aligned}$$

Comparison on the Membrane Performance and Production Costs.

Table S5. Comparison on the Pure Water Flux, SA Rejection and FRR Ratio of the Membranes in this Study and from Literature

Author	Polymer	Additive	Solvent	Pure water flux (L /m ² h bar)	SA rejection (%)	FRR (%)
This study	PVC	PVC-g-PEGMA	NMP, DMAc, DMSO	892±64	94.7±1.3	81.88±0.39
Fan et al ³	PVC	PVF ^a	DMAc	216±4	NA [*]	NA
Fang et al ⁴	PVC	MPC-PPGMA ^b	DMAc	100±17	89.0	100
Fang et al ⁵	PVC	PMMA-g-PEG ^c	DMAc	550	NA	NA
Liu et al ⁶	PVC	PEG ^d , F 127 ^e	DMAc	224	51.0	75
Wu et al ⁷	PVC	PVC-g-PEGMA	DMAc	659±57	93.7±1.1	80.12±4.92
Zhang et al ⁸	PVC	F 127 ^e	NMP	1405	92.3	88.00
Behboudi et al ⁹	PVC	PC ^f	NMP	504	NA	NA
Xu et al ¹⁰	PVC	PEG ^d	DMAc	201	NA	NA
Roy et al ¹¹	PVC	PEG ^d	DMAc	224	NA	NA
Yu et al ¹²	PVC	PVP ^g , nano-SiO ₂	DMAc	233	NA	NA
Zhou et al ¹³	PVC	PVC-co-PEGMA	DMAc	185	NA	NA
Zhu et al ¹⁴	PVC	F 127 ^e , TETA ^h	DMAc	260±3	NA	NA
Ahmad et al ¹⁵	PVC	bentonite-5	DMAc	467	NA	NA
Mishra et al ¹⁶	PVC	halloysite nanotubes	DMAc	52	NA	NA
Jhaveri et al ¹⁷	PVC	PVP ^g , GO-TiO ₂	DMAc	98	NA	NA

Fang et al ¹⁸	PVC	PVC-g-PMAA ⁱ	DMAc	87	NA	NA
Jiang et al ¹⁹	PVC	PES-g-PEGMA ^j	DMAc	110	NA	NA
Zhou et al ²⁰	PVC-co-PEGMA	-	DMAc	1310	NA	NA
Liu et al ²¹	PVC, CPVC ^k	F 127 ^e	DMAc	209±2	NA	NA
Wang et al ²²	PVC-PAN-PSS ^l	PEG ^d 200	DMSO	114±6	91.0±1.0	75.00
Arthanareeswaran et al ²³	PES	-	DMSO	12±1	NA	NA
Abdullah et al ³⁵	PES	-	DMSO	1.2	NA	NA
Meringolo et al ²⁴	PVDF	-	DMSO	12100	NA	NA
Chang et al ²⁵	PVDF	-	TEP	21800	NA	NA
Cui et al ²⁶	PVDF	-	ATBC	1161±34	NA	NA
Cui et al ²⁷	PVDF	-	TEGDA	2343	NA	NA
Jung et al ²⁸⁻³⁰	PVDF	-	PolarClean	988	NA	NA
Fadhil et al ³¹	PVDF-HFP ^m	-	TEP	16100	NA	NA
Marino et al ³²	PES	PEG ^d , PVP ^g	PolarClean	490	NA	NA

Note: *NA means not available.

PVF^a: polyvinyl formal

MPC-PPGMA^b: methacryloyloxyethylphosphorylcholine-co-poly(propylene glycol) methacrylate

PMMA-g-PEG^c: poly(methyl methacrylate-graft-poly(ethylene glycol) methacrylate)

PEG^d: poly(ethylene glycol)

F 127^e: Pluronic F 127

PC^f: polycarbonate

PVP^g: Poly(ethylene glycol)

TETA^h: triethylenetetramine

PVC-g-PMAAⁱ: poly (vinyl chloride)-graft-poly(methacrylic acid)

PES-g-PEGMA^j: poly(ether sulfone)-graft-poly(ether glycol) methyl ether methacrylate

CPVC^k: chlorinated polyvinyl chloride

PVC-PAN-PSS^l: poly(vinyl chloride-co-acrylonitrile-co-sodium 4-styrenesulfonate)

PVDF-HFP^m: poly(vinylidene fluoride-hexafluoropropylene)

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