

Enhanced hollow fiber membrane performance via semi-dynamic layer-by-layer polyelectrolyte inner surface deposition for nanofiltration and forward osmosis applications

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Table 1. Characteristics of PES hollow fiber substrate

| Dimension | | | Porosity | MWCO Outer/inner surface | | Mean pore size | Standard deviation | Pure water permeability | Tensile modulus | Stress at break | Strain at break |
|-----------|------------|-------------------|----------|--------------------------------|----|-------------------|-----------------------|----------------------------|--------------------|--------------------|--------------------|
| OD (μm) | ID (μm) | Thickness (μm) | ε (%) | (kDa) | | D* (nm) | σ | (LMH/bar) | (MPa) | (MPa) | % |
| 1480 | 1080 | 200 | 84 | 39 | 56 | 10.9 | 1.04 | 350 | 70.4 | 3.64 | 66 |

Table 2. Polyelectrolyte structure and functional groups

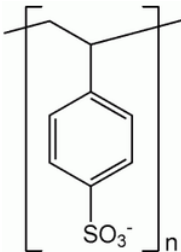
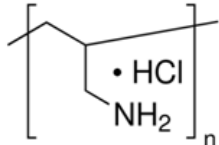
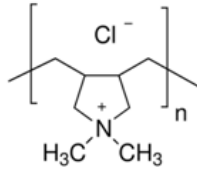
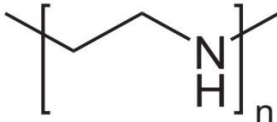
| Name | Molecular Structure | Functional Groups |
|---|---|---|
| Poly(styrene sulfonate) PSS |  | SO_3^- |
| Poly(allylamine hydrochloride) PAH |  | NH_3^+ |
| Poly(diallyl-dimethylammonium chloride) PDADMAC |  | NR_4^+ |
| Poly(ethyleneimine) PEI |  | NH_3^+ NRH^{2+} NR_2H^+ |

Table 3. Neutral solutes rejection performance

| Solute | Molecular weight (Da) | Stokes radius (nm) | LBL-2O | LBL-2I |
|-----------|--------------------------|-----------------------|-----------------------------------|-----------------------------------|
| | | | Solute rejection (%) ⁺ | Solute rejection (%) ⁺ |
| Glucose | 180 | 0.36 | 38.2 | 69.8 |
| Sucrose | 342 | 0.46 | 67.2 | 98.1 |
| Raffinose | 504 | 0.54 | 74.4 | 98.3 |

⁺Tested at 1 bar using 200 ppm neutral solute solutions

Table 4. Comparison of various NF membranes

| Membranes | Salt water permeability (LMH/bar) | Salt rejection (%) | MgCl ₂ concentration (ppm) | Operation pressure (bar) | Reference |
|----------------|-----------------------------------|--------------------|---------------------------------------|--------------------------|----------------------|
| LBL-2I | 9.8 | 98.2 | 1000 | 2 | Present work |
| | 8.2 | 97.4 | 4000 | 4.8 | Present work |
| LBL-6O | 12 | 94 | 1000 | 2 | [16] |
| UTC-20 | 9.7 | 98 | 1500 | 10 | [28] |
| NF-270 | 4 | 53 | 4000 | 4.8 | [10] |
| LBL flat sheet | 4 | 93 | 4000 | 4.8 | [10] |
| PDMAEMA/Psf | 1 | 98 | 1000 | 8 | [29] |
| PEI/TMC IP | 9.75 | 80 | 1000 | 4 | [30] |
| PDMCHEAs/Psf | 3.2 | 94.3 | 1000 | 6 | [31] |

Table 5. Separation performance of mixed salt solutions

| Feed solution TDS (ppm) | Ion composition (ppm) | | | Membrane ^a | SWP ^b (LMH/bar) | Ionic rejection (%) | | |
|----------------------------|-----------------------|------------------|-----------------|-----------------------|-------------------------------|---------------------|------------------|-----------------|
| | Mg ²⁺ | Ca ²⁺ | Na ⁺ | | | Mg ²⁺ | Ca ²⁺ | Na ⁺ |
| 3000 | 142 | 266 | 562 | od-LBL | 11 | 91 | 87 | 11 |
| | | | | id-LBL | 8.1 | 99.5 | 99 | 13.7 |
| 5000 | 218 | 420 | 894 | od-LBL | 7.2 | 72.5 | 68.8 | -7.1 |
| | | | | id-LBL (2 bar) | 4.8 | 96.2 | 92.1 | 7.6 |
| | | | | id-LBL (4.8 bar) | 7.4 | 99.6 | 97 | 16.4 |

^a Deposited with (PSS with 0.5 M NaCl/PAH with 2.5 M NaCl) x 2

^b Tested at 2 bar without special indication

Table 6. Comparison of various LBL FO membranes

| Sample | Water flux (L/m ² .h) | Salt flux/water flux(g/L) | Orientation | Reference |
|----------------------------|----------------------------------|---------------------------|--------------|--------------|
| LBL-2I | 73 | 0.06 | AL-facing-DS | Present work |
| LBL-6O | 40.5 | 0.2 | AL-facing-DS | [23] |
| 6#LBL FO | 22 ^a | 0.3 ^a | AL-facing-DS | [33] |
| (PAA-PSS/PAH) ₃ | 28 | 0.07 | AL-facing-DS | [34] |
| xLbL3 | 60 ^a | 0.1 ^a | AL-facing-DS | [35] |
| (PAH/PSS) _{3XX} | 21 | 0.06 | AL-facing-DS | [36] |
| LBL-2I | 21.5 | 0.03 | AL-facing-FW | Present work |
| LBL-6O | 18.4 | 0.11 | AL-facing-FW | [23] |
| 6#LBL FO | 16 ^a | 0.57 ^a | AL-facing-FW | [33] |
| xLbL3 | 30 ^a | 0.3 ^a | AL-facing-FW | [35] |
| (PAH/PSS) _{3XX} | 14.5 | 0.08 | AL-facing-FW | [36] |

Feed solution: DI water; draw solution:0.5M MgCl₂ solution

^a Estimated from published figures



Fig. 1. Schematic drawing of layer-by-layer deposition on hollow fiber inner surface.

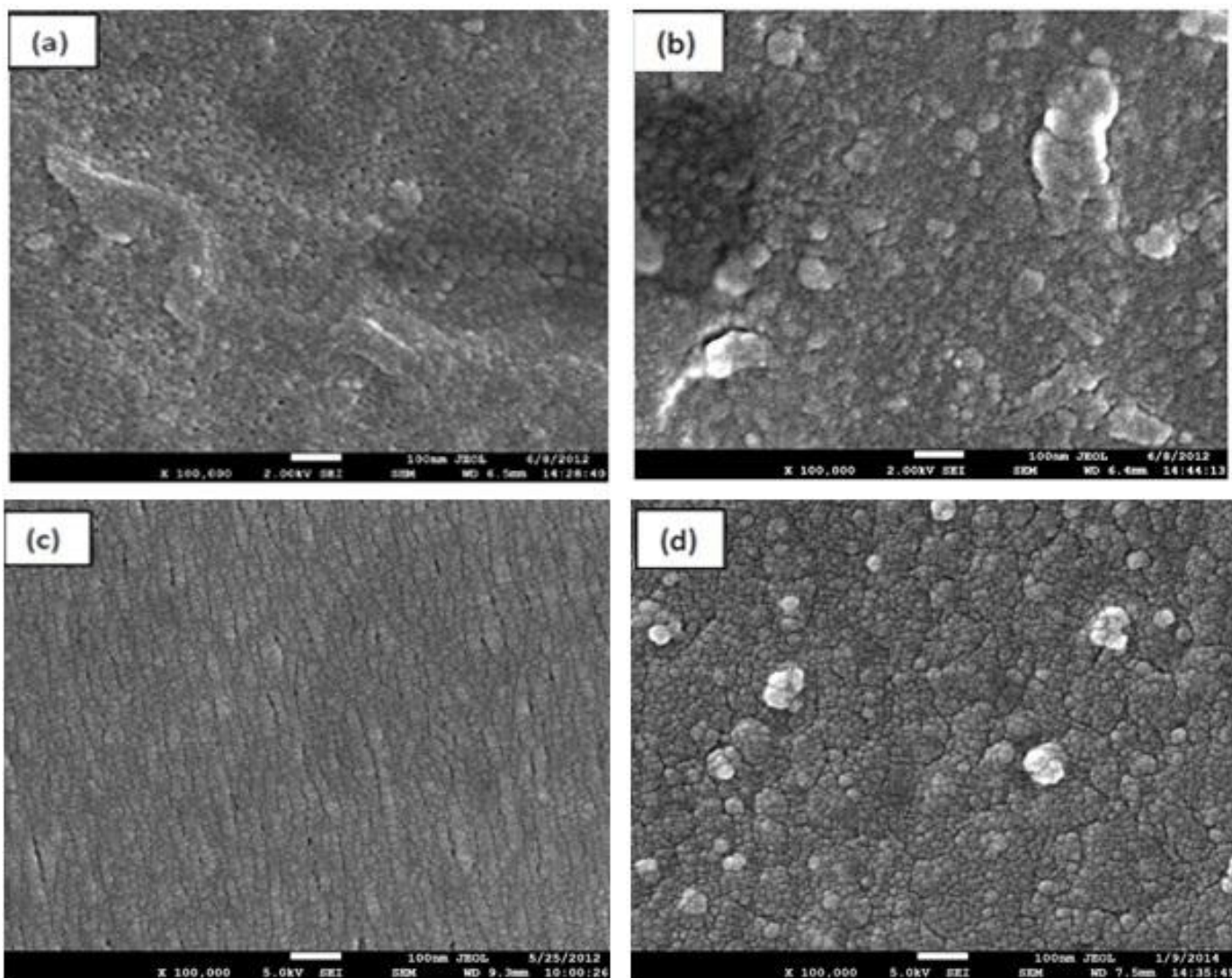


Fig.2. FESEM images of

- (a) substrate outer surface; (b) od-LBL membrane outer surface; (c) substrate inner surface;
 (d) id-LBL membrane inner surface at magnification of 100k.
 (Deposited with (PSS with 0.5 M NaCl/PAH with 2.5 M NaCl) x 2)

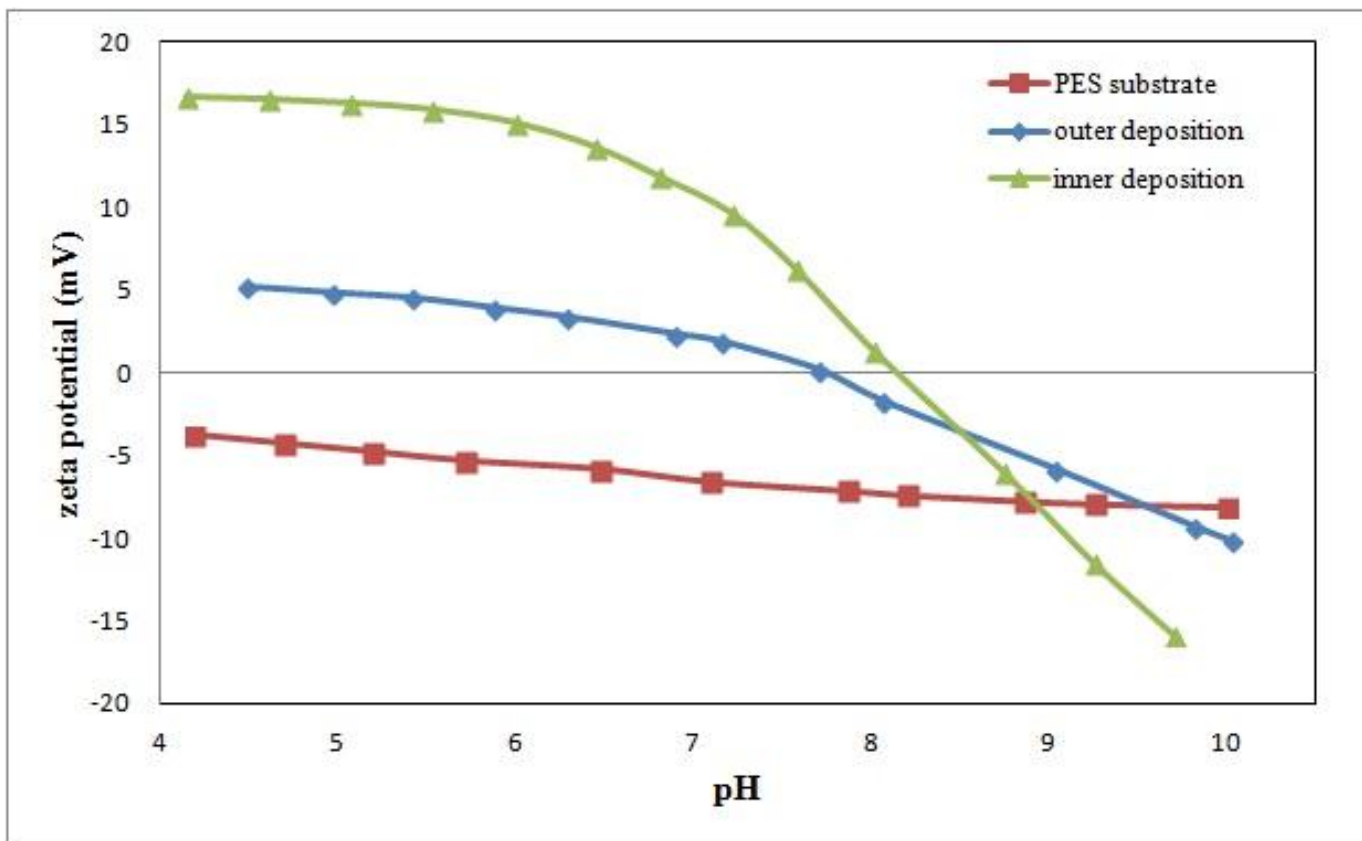


Fig. 3. Zeta potential of PES substrate and LBL membranes (Deposited with (PSS with 0.5 M NaCl/PAH with 2.5 M NaCl) x 2).

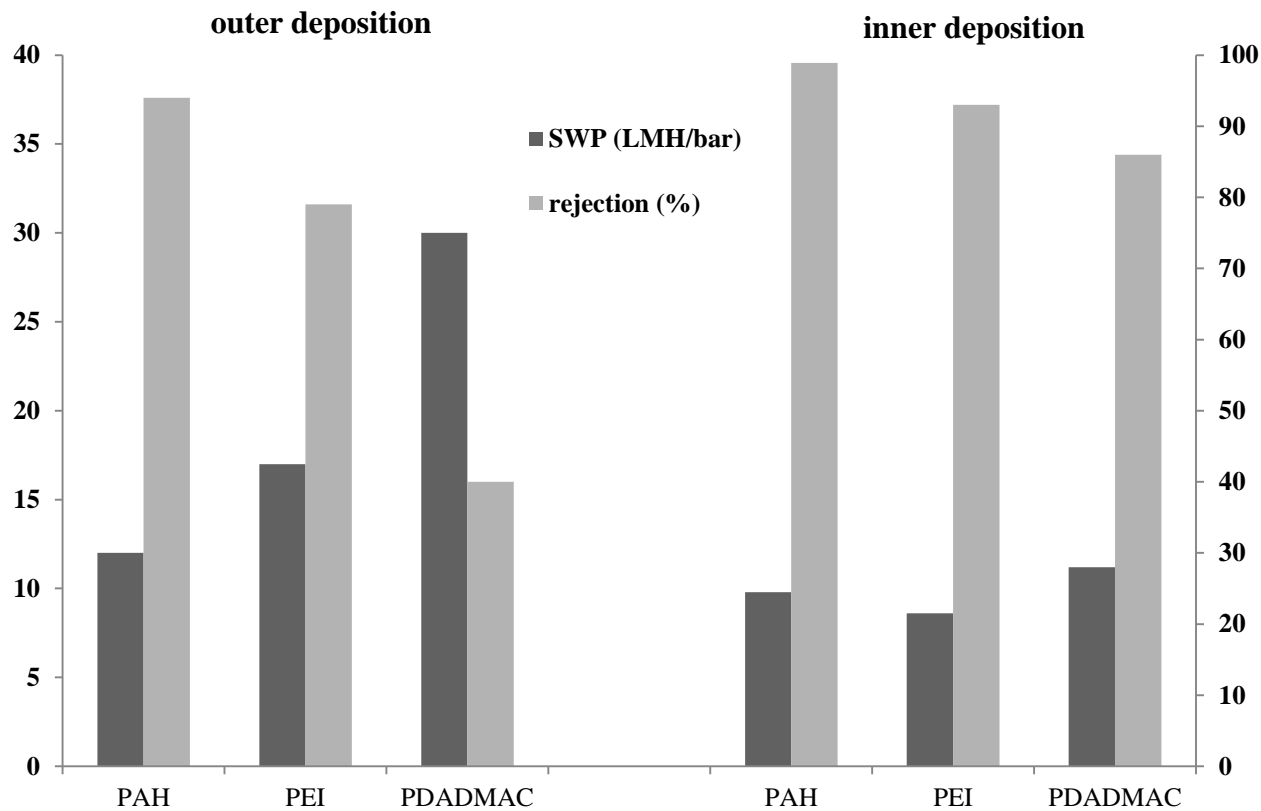


Fig. 4. Nanofiltration performance of LBL membranes using different polycations.
(Deposited with (PSS with 0.5 M NaCl/Poly-cations with 2.5 M NaCl) x 2;
tested at 2 bar using 1000 ppm MgCl_2 solution)

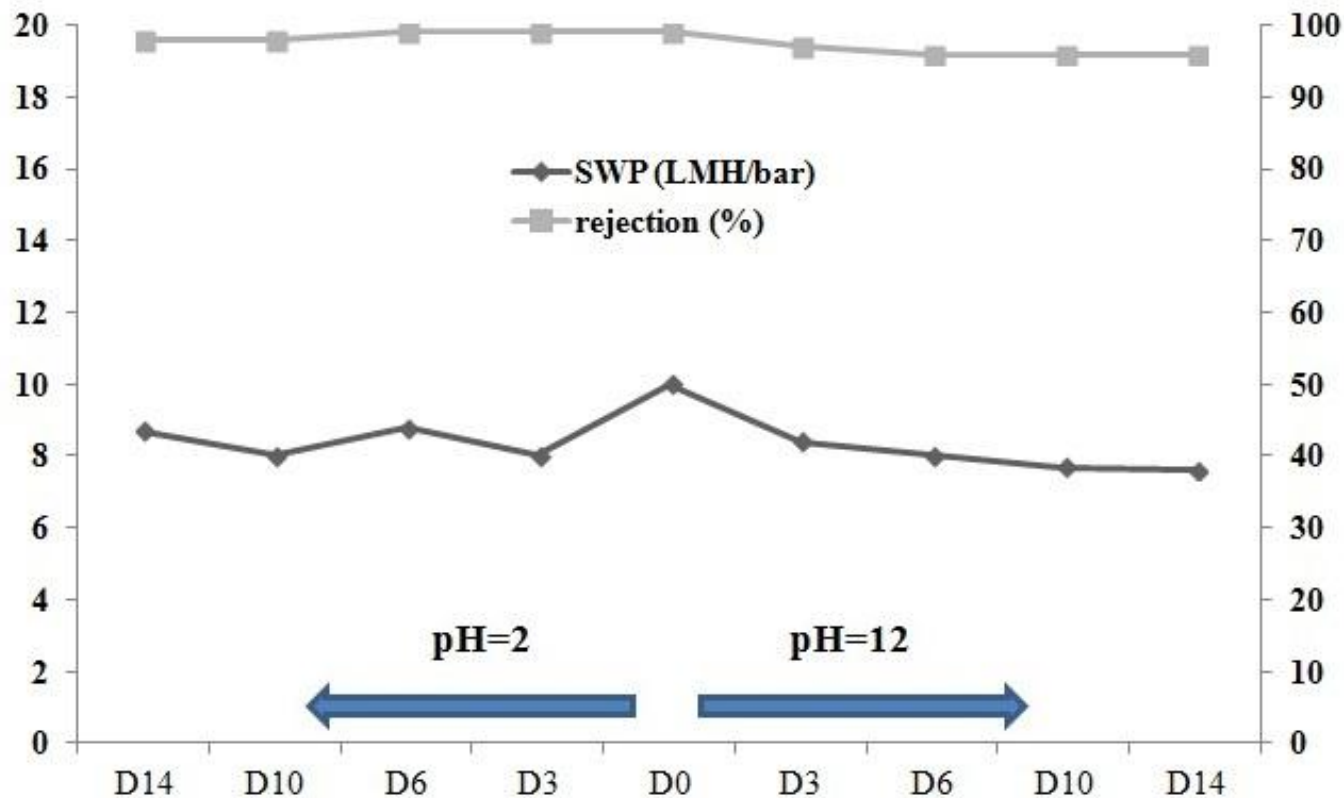


Fig. 5. 14-day acid/alkaline stability tests of inner deposited LBL membranes (Deposited with (PSS with 0.5 M NaCl/PAH with 2.5 M NaCl) x 2).

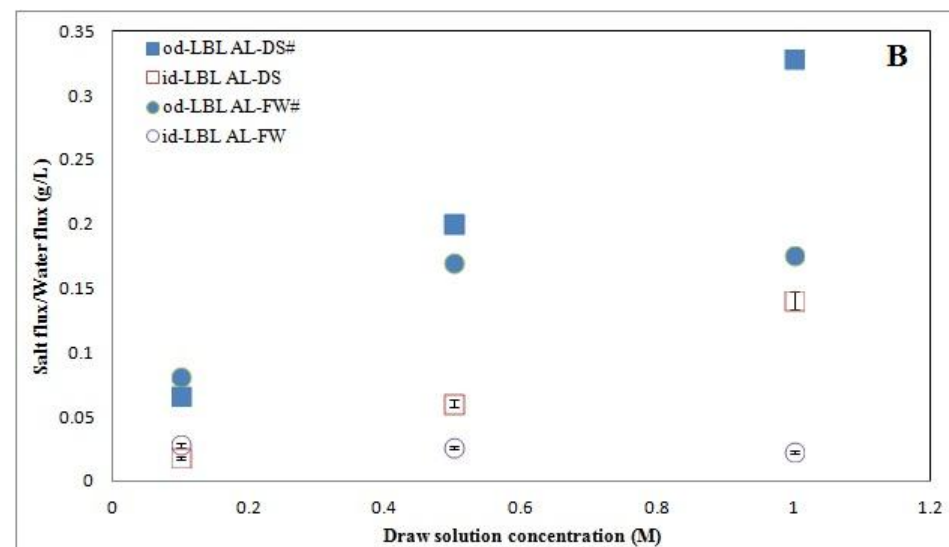
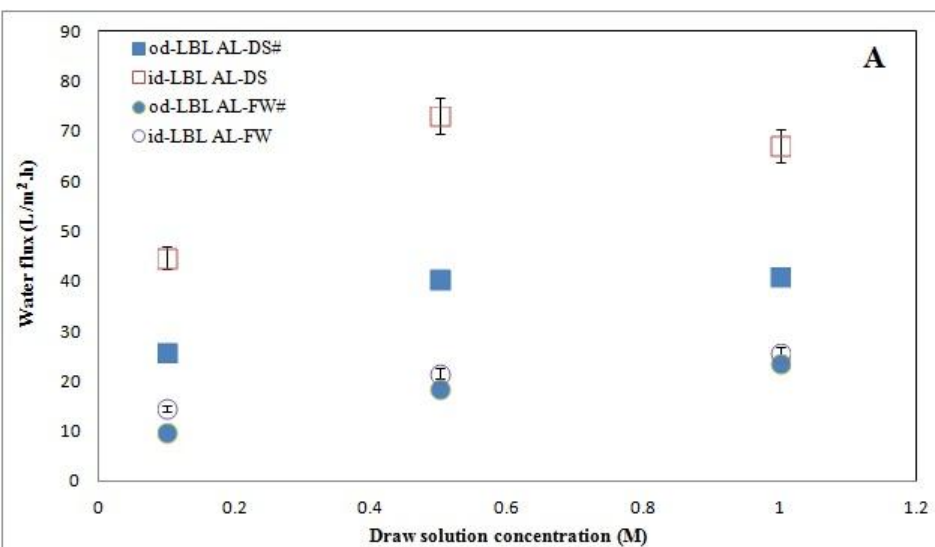


Fig. 6. FO performance of LBL membranes using up to 1 M MgCl_2 draw solutions
 (A) Water flux; (B) Salt flux.
 (od-LBL membranes deposited with $(\text{PSS}/\text{PAH}) \times 6$ [23]; id-LBL membranes deposited with $(\text{PSS}/\text{PAH}) \times 2$)