

## Rapid water transport through controllable, ultrathin polyamide nanofilms for high-performance nanofiltration

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Table S1 Fabrication of the free-standing nanofilms at a free aqueous-organic interface. The nanofilms were then transferred onto a silicon wafer for AFM analyses

| Membrane ID            | PIP concentration<br>(w/v%) | TMC concentration<br>(w/v %) | Reaction<br>Time (min) |
|------------------------|-----------------------------|------------------------------|------------------------|
| Free-standing PA films | PAF_0.025%-0.05%            | 0.025                        | 0.05                   |
|                        | PAF_0.05%-0.05%             | 0.05                         | 0.05                   |
|                        | PAF_0.10%-0.05%             | 0.10                         | 0.05                   |
|                        | PAF_0.15%-0.05%             | 0.15                         | 0.05                   |
|                        | PAF_0.20%-0.05%             | 0.20                         | 0.05                   |

Table S2 Fabrication of nanofilm composite membranes (NCMs) via direct filtration of the free-standing PA nanofilms onto PDA-coated PAN substrates.

| Membrane ID               | PIP concentration<br>(w/v%) | TMC concentration<br>(w/v %) | Reaction<br>Time (min) |
|---------------------------|-----------------------------|------------------------------|------------------------|
| PDA-coated PAN based NCMs | NCM_0.025%-0.05%            | 0.025                        | 0.05                   |
|                           | NCM_0.05%-0.05%             | 0.05                         | 0.05                   |
|                           | NCM_0.10%-0.05%             | 0.10                         | 0.05                   |
|                           | NCM_0.15%-0.05%             | 0.15                         | 0.05                   |
|                           | NCM_0.20%-0.05%             | 0.20                         | 0.05                   |
|                           | NCM_0.05%-0.025%            | 0.05                         | 0.025                  |
|                           | NCM_0.05%-0.05%             | 0.05                         | 0.05                   |
|                           | NCM_0.05%-0.10%             | 0.05                         | 0.10                   |
|                           | NCM_0.05%-0.15%             | 0.05                         | 0.15                   |
|                           | NCM_0.05%-0.20%             | 0.05                         | 0.20                   |

Note: Transfer of the free-standing PA nanofilms onto pristine PAN was also carried out. These membranes are denoted as PAN\_x%-y%, where x represents PIP concentration and y represents TMC concentration.

Table S3 Comparison of water permeance and mono/divalent salt selectivity of various polyamide-based membranes reported in the literature and this work.

| Membranes        | Water permeance<br>( $\text{L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$ ) | Salt selectivity | Testing condition  | Ref.         |
|------------------|--|------------------|--|--------------|
| NCM_0.025%-0.05% | 25.1   | 80.6             | 1.5 g $\text{L}^{-1}$ , $\text{Na}_2\text{SO}_4 / \text{NaCl}$ solution, 4 bar       | This work    |
| TFN-ZPNP3        | 11.0   | 28.4             | 1.0 g $\text{L}^{-1}$ , $\text{Na}_2\text{SO}_4 / \text{NaCl}$ solution, 6 bar       | <sup>1</sup> |
| TFN-mZIF2        | 14.9   | 13.1             | 1.0 g $\text{L}^{-1}$ , $\text{Na}_2\text{SO}_4 / \text{NaCl}$ solution, 4 bar       | <sup>2</sup> |
| ZNGTFNM2         | 10.6   | 31.1             | 1.0 g $\text{L}^{-1}$ , $\text{Na}_2\text{SO}_4 / \text{NaCl}$ solution, 6 bar       | <sup>3</sup> |
| TFC2.0_5         | 14.5   | 24.1             | 1.0 g $\text{L}^{-1}$ , $\text{Na}_2\text{SO}_4 / \text{NaCl}$ solution, 4 bar       | <sup>4</sup> |
| PSS/PVA-PSF      | 8.34   | 26.1             | 0.5 g $\text{L}^{-1}$ , $\text{Na}_2\text{SO}_4 / \text{NaCl}$ solution, 5 bar       | <sup>5</sup> |
| PA/PD-PES        | 11.4   | 10.8             | 1.0 g $\text{L}^{-1}$ , $\text{Na}_2\text{SO}_4 / \text{NaCl}$ solution, 2 bar       | <sup>6</sup> |
| NF-2             | 12.1   | 9.1              | 1.0 g $\text{L}^{-1}$ , $\text{Na}_2\text{SO}_4 / \text{NaCl}$ solution, 6 bar       | <sup>7</sup> |
| NF2              | 16.4   | 25.2             | 1.0 g $\text{L}^{-1}$ , mixed $\text{Na}_2\text{SO}_4 / \text{NaCl}$ solution, 5 bar | <sup>8</sup> |
| NF4              | 17.2   | 27.9             | 0.5 g $\text{L}^{-1}$ , $\text{Na}_2\text{SO}_4 / \text{NaCl}$ solution, 5 bar       | <sup>9</sup> |

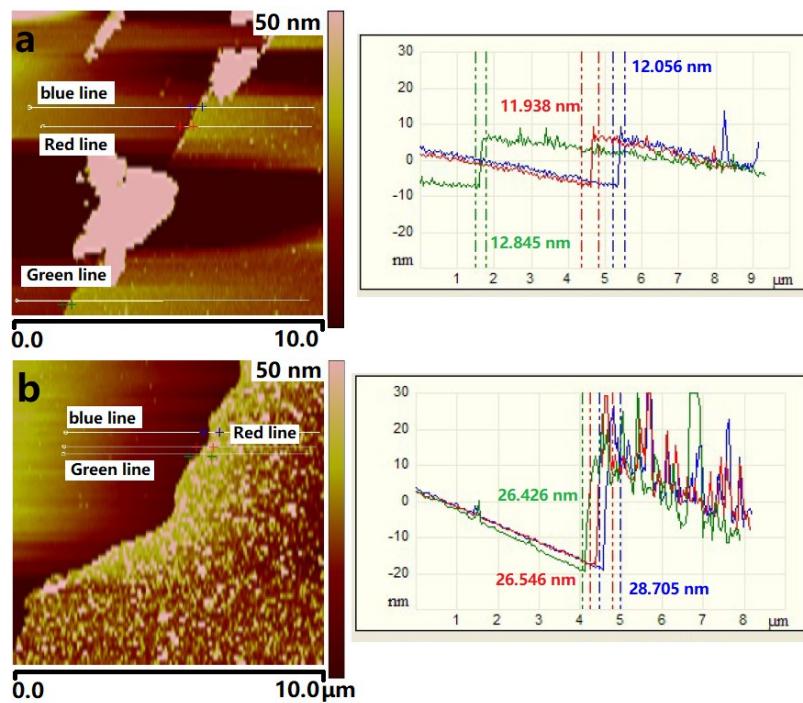


Fig. S1 AFM image and the height profile along the dotted line of the free-standing PA nanofilms transferred on a silicon substrate: (a) PAF-0.025%-0.05%, and (b) PAF-0.10%-0.05%

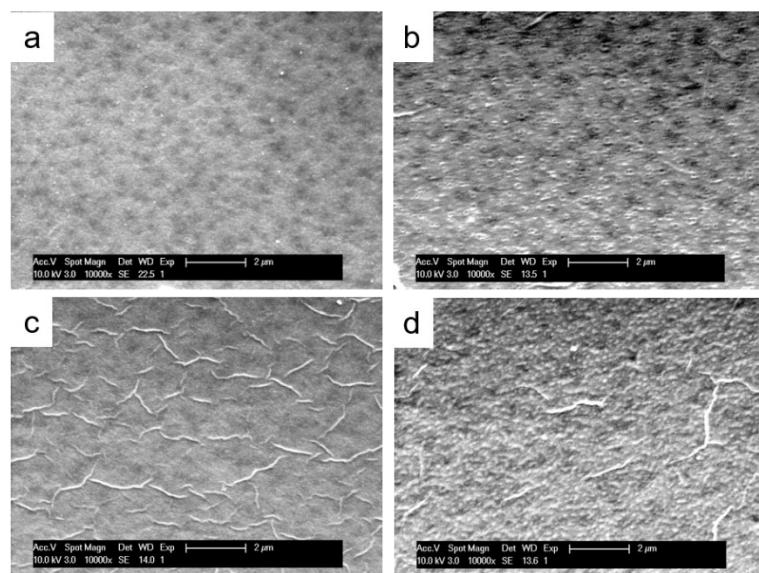


Fig. S2 SEM surface morphologies of nanofilms transferred onto unmodified PAN membranes. (a) PAN\_0.025%-0.05%, (b) PAN\_0.05%-0.05%, (c) PAN\_0.15%-0.05%, and (d) PAN\_0.20%-0.05%.

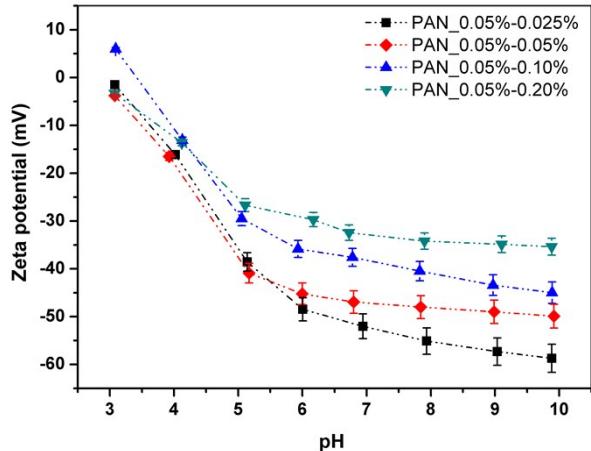


Fig. S3 The role of different TMC concentrations in zeta potentials of nanofilms that are transferred onto PAN surface.

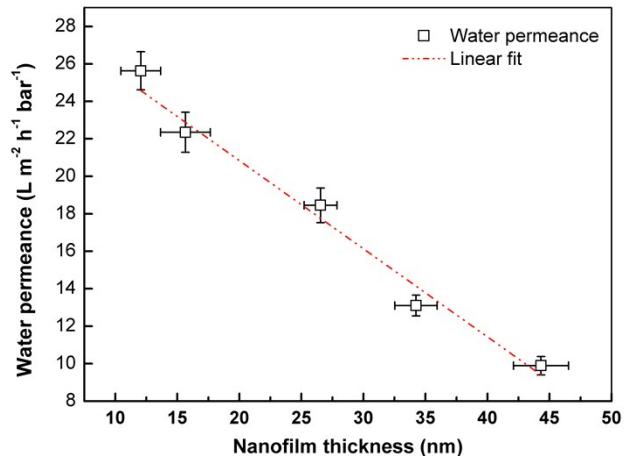


Fig. S4 Variation of water permeance of polyamide nanofilms attached onto an unmodified PAN support versus the nanofilm thickness. Nanofiltration experiments were performed in a lab-made crossflow setup at room temperature under 4 bar.

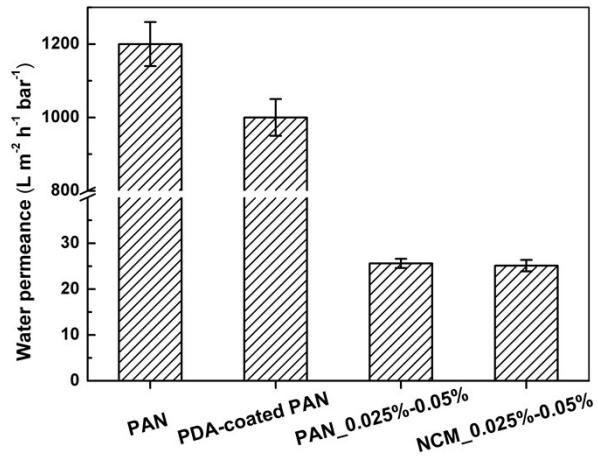


Fig. S5 Water permeance of PAN, PDA-coated PAN, PAN\_0.025%-0.05%, and NCM\_0.025%-0.05%. Tests of ultrafiltration membranes were conducted in a lab-made crossflow setup at room temperature and under 1 bar.

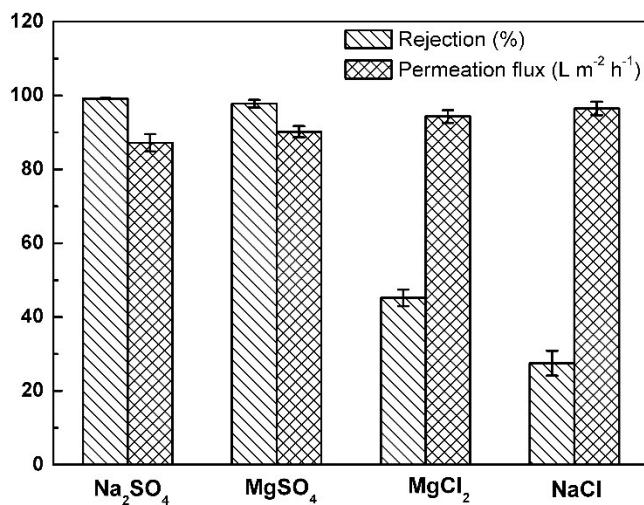


Fig. S6 Salts ( $\text{Na}_2\text{SO}_4$ ,  $\text{MgSO}_4$ ,  $\text{Na}_2\text{SO}_4$ , and  $\text{NaCl}$ ) rejection of PA nanofilms attached onto PAN (PAN\_0.025%-0.05%) via vacuum filtration. The ensuing composite membranes were directly used for NF performance measurements after simple rinse.

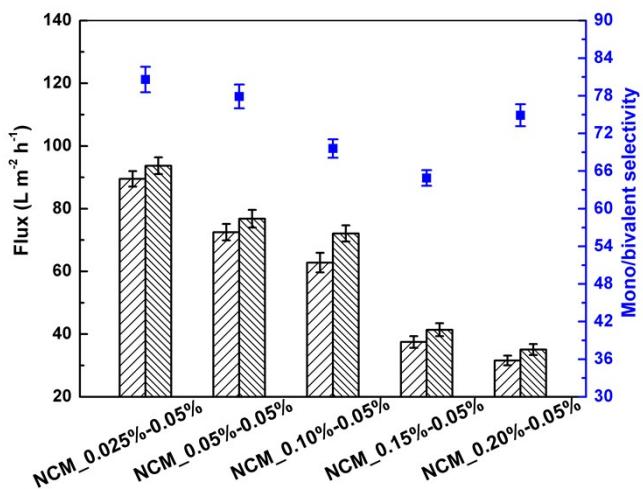


Fig. S7 Permeation flux and the calculated salt selectivity of NCMs via varying PIP monomer concentration. The used TMC concentration is 0.05 w/v%.

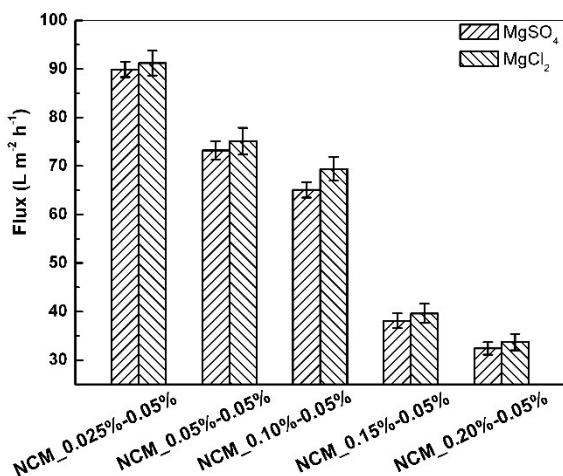


Fig. S8 Permeation flux of NCMs via varying PIP monomer concentration. The used TMC concentration is 0.05 w/v%. The inlet salt concentration is 1.5 g  $\text{L}^{-1}$ .

## References

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