

Supplementary information

Hydroxy Artificial Water Channels Enable Ultraselective Reverse Osmosis Membranes for *N*-Nitrosamine Removal in Water Reuse

Zhiwei Qiu¹, Jiansuxuan Chen¹, Chuyang Y. Tang², Ruobin Dai^{1,*}, Zhiwei Wang^{1,*}

¹ State Key Laboratory of Water Pollution Control and Green Resource Recycling, Shanghai Institute of Pollution Control and Ecological Security, School of Environmental Science and Engineering, Tongji University, Shanghai 200092, China

² Department of Civil & Environmental Engineering, National University of Singapore, Singapore 117576, Singapore

*Corresponding Authors.

Email: dairuobin@tongji.edu.cn (R. Dai);

zwwang@tongji.edu.cn (Z. Wang).

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Supplementary Methods

Supplementary Method 1. Materials and chemicals

The PA rejection layer was constructed on a polysulfone (PSf) membrane (molecular weight cut off (MWCO) = 67 kDa) from Vontron Technology Co., Ltd., Beijing, China. The pristine PSf membrane was treated twice with 20% isopropanol, with each treatment lasting for 30 min. Subsequently, the treated PSf membrane was stored in deionized (DI) water until use. Amino propanediol ($\geq 97\%$, Shanghai Macklin Biochemical Technology Co., Ltd), octyl isocyanate ($\geq 97\%$, ALFA), tetrahydrofuran ($\geq 99.8\%$, Sinopharm Chemical Reagent Co., Ltd), ethyl acetate ($\geq 99.5\%$, Sinopharm Chemical Reagent Co., Ltd), and acetonitrile ($\geq 99.7\%$, Sinopharm Chemical Reagent Co., Ltd) were used for the preparation of OH AWCs. Sodium chloride (NaCl, $\geq 99.8\%$) and ethanol ($\geq 95\%$) were purchased from Sinopharm Chemical Reagent Co., Ltd. Trimesoyl chloride (TMC, $\geq 98\%$), *m*-phenylenediamine (MPD, $\geq 99.5\%$), and *n*-hexane ($\geq 99.5\%$) were supplied by Shanghai Aladdin Bio-Chem Technology Co., Ltd. for the IP to fabricate the RO membrane. The nonporous SiO₂ (size: 1 nm), which was used to self-assemble with OUA to inhibit the formation of OH AWCs, was provided by Bohua Nano Technology Co., Ltd. A citrate-stabilized dispersed GNPs solution (5 nm, 5×10^{13} particles/mL) for investigating the water transport pathways was supplied by BBI Solutions (UK) and stored at 4°C. NDMA (74.1 Da, $\geq 99\%$), NMEA (88.1 Da, $\geq 99\%$), NPYR (100.1 Da, $\geq 99\%$), NDEA (102.1 Da, $\geq 99\%$), NPIP (114.1 Da, $\geq 99\%$), and NMOR (116.1 Da, $\geq 99\%$) were purchased from Shanghai Aladdin Bio-Chem Technology Co., Ltd. All reagents were of analytical grade and used without further purification unless otherwise specified. DI water was used in all experiments.

Supplementary Method 2. Fabrication of AWCM-0 membranes

IP on a porous supporting substrate was a well-known technique for manufacturing PA RO membranes^{1, 2}. In brief, MPD flakes were added to DI water to formulate an aqueous solution with a concentration of 2.0 wt%. TMC were introduced into *n*-hexane to prepare an organic solution with a concentration of 0.10 wt%. This is a typical concentration used for preparing PA RO membranes via IP. Firstly, pour the MPD aqueous solution onto the surface of the PSf substrate membrane fixed between two identical custom-designed stainless-steel frames³. After a 2 min wetting period, carefully remove the excess aqueous solution from the surface of the PSf membrane using a rubber roller⁴. Afterward, gently pour a 0.10 wt% TMC/*n*-hexane onto the aqueous solution soaked PSf membrane and conduct a 2 min IP reaction to form the PA RO membrane. After the reaction, the PA RO membrane (labeled AWCM-0) was washed with *n*-hexane and DI water three times. Finally, the prepared AWCM-0 membrane was stored in DI water until the testing was conducted.

Supplementary Method 3. Preparation OH AWCs

The synthesis of OH AWCs was carried out following the previously reported self-assembly method with minor modifications^{5, 6, 7}. Briefly, under sonication, amino propanediol (10 mmol) was mixed with the corresponding equimolar amount of octyl isocyanate. Subsequently, the mixture was dissolved in 10 mL of tetrahydrofuran, 5 mL of ethyl acetate, and 5 mL of acetonitrile. The reaction system was heated to 80°C for 2 h, and the resulting solution was subjected to conventional rotary evaporation under sealed conditions to obtain white powder (octyl-ureido-polyol:1-(1,3-dihydroxypropan-2-yl)-3-octylurea). After washing the products with methanol and hexane, they were separated using a rotary centrifuge, and the residual solvent was eliminated using a vacuum desiccator. Afterwards, the formed octyl-ureido-polyol self-assembles into OH AWCs through H-bonds interactions in ethanol/water environments.

Supplementary Method 4. Water permeance, NaCl rejection and water/NaCl selectivity

Water permeance (S1) was determined by continuously weighing the mass of the permeate over a specific period⁸.

$$A = \frac{\Delta m}{\rho \Delta P \Delta t A_m} \quad (S1)$$

where A represents water permeance ($L^2 \cdot h^{-1} \cdot m^{-2} \cdot bar^{-1}$), Δm is the mass of permeate collected within a certain time interval (Δt , h), A_m represents the effective membrane area (m^2), ρ represents the density of water, ΔP represents the transmembrane pressure.

According to equation (S2), the salts rejection (R) was calculated.

$$R = (1 - \frac{C_p}{C_f}) \times 100\% \quad (S2)$$

where the conductivity of the feed (C_f) and permeate (C_p) was measured using a conductivity meter (DDSJ-308F, INESA instrument). The salt permeance (B) was calculated using the following equation (S3).

$$B = (\frac{1}{R} - 1) \times A \times \Delta P \quad (S3)$$

Water/NaCl selectivity (A/B) of the membranes is calculated based on A and B .

Supplementary Method 5. MD simulation of OUA self-assembly into OH AWCs

The self-assembly of OUA into OH AWCs in water/ethanol solution was investigated through all-atom MD simulations using the Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS, version 2022; open source, available at <https://www.sandia.gov/ccr/software/lammps/>) software package⁹. The Optimized Potentials for Liquid Simulations All-Atom (OPLS-AA) force field¹⁰ was employed to describe bonded and non-bonded interactions. Initial mixed systems were constructed and pre-optimized using energy minimization with the conjugate gradient algorithm to remove unfavorable contacts prior to dynamic sampling. Accelerated sampling techniques, including temperature annealing and bias potentials, were used to efficiently explore configurational space and promote channel formation.

Equilibration and production runs were conducted in the isothermal–isobaric (NPT) ensemble at 300 K and 1 atm using the Nosé–Hoover thermostat and barostat¹¹. The integration time step was set to 1 femtosecond (fs), and each production trajectory was propagated for 1000 picoseconds (ps) to follow the complete assembly process. The main control variable monitored throughout the simulations was the characteristic bond length involved in water channel formation, allowing correlation between geometric changes and assembly kinetics. Periodic boundary conditions were applied in all directions, and long-range electrostatics were treated using the particle–particle particle–mesh (PPPM) Ewald method with a real-space cutoff of 12 Å. Van der Waals interactions were modeled with Lennard–Jones potentials and the same cutoff.

Post-processing of MD trajectories was performed using in-house Python scripts together with the MD Analysis library¹² to extract structural and dynamical descriptors of the self-assembly process. Quantities analyzed included the radius of gyration, H-bond interactions, the pore size distribution of the formed OH AWCs, and their structural diffraction peaks. This combined approach enabled direct observation of OH AWCs formation and provided mechanistic insight into the self-assembly pathway under near-ambient conditions.

Supplementary Method 6. MD simulation of the process of OH AWCs incorporation into the PA layer

MD simulations were employed to investigate the process of OH AWCs incorporation into the PA layer of RO membranes, with the molecular ratio of OH AWCs: MPD: TMC: *n*-hexane: water set to 8:100:100:1965:16145. The initial configurations of the solvent systems were constructed using PACKMOL¹³, with all molecules randomly inserted into a cubic simulation box. The OPLS-AA force field^{14, 15} was employed, consisting of bonded and nonbonded interactions, with the latter including van der Waals (vdW) and electrostatic interactions. The nonbonded interactions include vdW and electrostatic interactions, which are described by the following equations:

$$E_{LJ}(r_{ij})=4\epsilon_{ij}\left(\left(\frac{\sigma_{ij}}{r_{ij}}\right)^{12}-\left(\frac{\sigma_{ij}}{r_{ij}}\right)^6\right) \quad (S4)$$

$$E_c(r_{ij})=\frac{q_i q_j}{4\pi\epsilon_o\epsilon_r r_{ij}} \quad (S5)$$

For different types of atoms, the Lorentz–Berthelot mixing rules were adopted for vdW interactions, as defined in Equation S6. The cutoff distance for vdW and electrostatic interactions was set to 1.2 nm, and the particle mesh Ewald (PME) method was employed to calculate long-range electrostatic interactions.

$$\sigma_{ij}=\frac{1}{2}(\sigma_{ii}+\sigma_{jj}); \epsilon_{ij}=(\epsilon_{ii}*\epsilon_{jj})^{1/2} \quad (S6)$$

For this simulation, energy minimization was first performed to relax the simulation box. Then, an isothermal–isobaric (NPT) ensemble with a 1.0 fs time step was employed to optimize the simulation box, with the temperature set to 298 K and the pressure set to 1.0 atm. The temperature and pressure were maintained using the Nosé–Hoover thermostat and the Parrinello–Rahman barostat, respectively. The NPT simulation was run for 10.0 ns, which was sufficiently long to achieve a stable box size. Following the NPT simulation, a canonical (NVT) ensemble was performed for 40.0 ns to further optimize the simulation box, with the time step set to 2.0 fs. Covalent cross-linking bonds were programmatically formed between the reactive groups of TMC and MPD monomers using an in-house script that detected interatomic distances below a threshold of 5 Å and dynamically created

bonds during the initial system setup phase. A canonical (NVT) ensemble was performed for 40.0 ns to further optimize the simulation box, with the time step set to 2.0 fs. In all MD simulations, the motion of atoms was described by classical Newton's equations of motion, which were solved using the velocity-Verlet algorithm. All simulations were performed using the GROMACS software package (version 2025.1; open source, available at <https://www.gromacs.org/>)¹⁶.

Supplementary Method 7. MD simulations analyzed the self-assembly process of SiO₂ and OUA

Nonporous SiO₂ and OUA molecules were drawn using chemical software. The molecular structures were optimized using the B3LYP functional of the density functional theory (DFT) method with the 6-31G(d) basis set in Gaussian 09²². The optimized structures were then used to calculate the restrained electrostatic potential (RESP) charges of the repeating unit fragments via Multiwfn, and these charges were employed to construct the force-field parameters for the small molecules based on general amber force field (GAFF)^{17, 18, 19}. The parameters for SiO₂ were adopted from literature-reported force fields²⁰.

All simulations were performed using GROMACS software (version 2023.2; open source, available at <https://www.gromacs.org/>)²¹. The systems were parameterized with the AMBER14SB²² force field to describe their overall properties. OUA and nonporous SiO₂ were added to three simulation boxes at a molar ratio of 77:10, along with ethanol at a volume ratio of 1:1 with water. The box dimensions were set such that the distance between the box boundaries and the contents was 15 Å, and the systems were solvated with TIP3P water molecules. Energy minimization was carried out using the steepest descent algorithm (1000 kJ/mol/nm), followed by a 0.5 ns NPT equilibration. Simulation parameters were set as follows: temperature was maintained at 298 K using the V-rescale²³ thermostat, and pressure was kept at 1 bar using the C-rescale barostat²⁴. Long-range van der Waals interactions were truncated at 10 Å, and dispersion corrections were applied using EnerPres. Trajectory snapshots were obtained using visual molecular dynamics software (VMD) (version 1.9.3; open source, available at <https://www.ks.uiuc.edu/Research/vmd/>), and detailed molecular interactions were visualized with PyMOL Molecular Graphics System (version 2.5.0; open source, available at <https://github.com/schrodinger/pymol-open-source>).

Supplementary Method 8. MD simulations analyze the transport mechanism of molecules in AWCM

The structural models of the OH AWCs, HC6 AWCs, IUP AWCs, PA matrix, and AWCM were constructed through a hierarchical modeling protocol designed to closely reproduce their experimentally reported microstructures. The OH AWCs, HC6 AWCs, and IUP AWCs were first built according to their chemical formulas and three-dimensional structures reported in synthetic studies⁵, and the initial coordinates were generated in Avogadro and optimized by density functional theory calculations at the B3LYP/6-31G(d) level. Electrostatic potential-derived RESP charges were obtained from HF/6-31G(d) calculations, and the GAFF2 force field was employed to describe bonded and nonbonded interactions, with key torsional parameters refined against quantum chemical scans. A crosslinked PA network was generated via interfacial polymerization of MPD and TMC using the polymatic algorithm, yielding structural parameters consistent with experimental data. After polymerization, the network was equilibrated at 300 K and 1 atm to relax the free volume and pore structure, yielding structural features in close agreement with experimental measurements of PA membranes. AWCM models were constructed by embedding multiple optimized OH AWCs into the equilibrated PA matrix using the packmol algorithm. The composite systems were subsequently solvated with explicit TIP4P-Ew water molecules, neutralized with counterions, and equilibrated in a stepwise manner, beginning with restrained solvent relaxation and followed by extended NPT equilibration to allow relaxation of both the PA matrix and channel structures. Structural stability was confirmed by monitoring pore diameters, radial distribution functions, and free volume distributions, all of which indicated preservation of the OH AWCs geometry and realistic packing of the PA matrix. This modeling workflow ensured that the constructed OH AWCs, PA network, and AWCM closely reproduced the key microstructural features observed experimentally.

The diffusion dynamics of molecules through the OH AWCs, HC6 AWCs, IUP AWCs, PA matrix, and AWCM were investigated using a combination of molecular dynamics (MD) simulations and quantitative transport analyses. The atomic simulation environment²⁵ was employed for system setup,

trajectory analysis, and data visualization. Reactive MD simulations were performed using Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS, version 2022; open source, available at <https://www.sandia.gov/ccr/software/lammps/>) with a hybrid force field approach. The Reactive Force Field (ReaxFF)⁹ was used to capture the chemical reactivity at the water-polymer interface, while the Optimized Potentials for Liquid Simulations-All Atom (OPLS-AA) force field¹⁰ was applied to describe non-reactive interactions within the polymer backbone. MD simulations were performed using LAMMPS to model the structural evolution and transport properties of water in the OH AWCs, HC6 AWCs, IUP AWCs, PA matrix, and OH AWCs-PA hybrid system. The ReaxFF force field²⁶ was applied to accurately describe bond dissociation and formation between water molecules and both the OH AWCs and the PA matrix, while the OPLS-AA force field was used to model the non-reactive polymer backbone. The simulation box was constructed with periodic boundary conditions in all three dimensions. The system was equilibrated in the canonical (NVT) ensemble using the Nosé–Hoover thermostat at 300 K²⁷, with a time step of 1 fs and a total simulation time of 100 ns to ensure sufficient sampling of water diffusion pathways. Long-range electrostatic interactions were calculated using the Particle–Mesh Ewald (PME)²⁸ method with a real-space cutoff of 1.2 nm, while van der Waals interactions were truncated at the same distance. The steepest descent algorithm was employed for energy minimization prior to MD simulations. The diffusive behavior of water and NDMA through the OH AWCs, HC6 AWCs, IUP AWCs, PA matrix, and OH AWCs–PA hybrid system was characterized via multiple statistical and structural analyses. The number of H-bonds was calculated as a function of time to evaluate the dynamic H-bonds network during water permeation. The lifetime of H-bonds was determined by analyzing the survival probability of H-bonds over time, following the methodology introduced by Luzar and Chandler²⁹. The residence time of water molecules within the cyanide water channels was calculated by tracking the duration each molecule remained confined in the channel region. The jump frequency, defined as the number of transitions between adjacent sites, was obtained from trajectory data using mean first-passage time (MFPT) analysis³⁰. Furthermore, MD simulations were employed to analyze the permeation barriers

of water molecules, NaCl, and NDMA through OH AWCs, HC6 AWCs, IUP AWCs, and the PA matrix. The detailed computational parameters were as follows:

1. Diffusion coefficient

The diffusion coefficient (D) was calculated using the Einstein relation based on the mean-squared displacement (MSD):

$$D(x) = \frac{1}{2d} \lim_{x \rightarrow \infty} \frac{d}{dt} \left\langle |r(t) - r(0)|^2 \right\rangle_x \quad (S7)$$

where d is the dimensionality of the system, and $r(t)$ denotes the molecular trajectory as a function of time.

2. H-bonds number

The number of H-bonds at each time step in a MD trajectory is defined by geometric criteria. The donor–acceptor distance r_{DA} is less than a specified cutoff distance, typically 3.5 Å. The angle between donor-hydrogen-acceptor θ_{HDA} is smaller than a defined cutoff angle (typically 30°).

For each pair of atoms, we define a H-bonds existence function $h_{ij}(t)$ as follows:

$$h_{ij}(t) = \begin{cases} 1, & \text{if } r_{ij}(t) < r_c(t) \text{ and } \theta_{ij}(t) < \theta_c \\ 0, & \text{otherwise} \end{cases} \quad (S8)$$

Thus, the total number of H-bonds ($N_{HB}(t)$) at time t is:

$$N_{HB}(t) = \sum_{i,j} h_{ij}(t) \quad (S9)$$

3. H-bonds lifetime

The H-bonds lifetime (τ_{HB}) is defined as the average time that a H-bond remains intact. To compute this, the autocorrelation function of H-bonds existence was used:

$$C(t) = \left\langle h_{ij}(0) h_{ij}(t) \right\rangle \quad (S10)$$

where $\langle \cdot \rangle$ denotes averaging over all H-bonds pairs and different time origins.

The τ_{HB} can be computed by integrating the autocorrelation function:

$$\tau_{HB} = \int_0^\infty C(t) dt \quad (S11)$$

The integral yields the characteristic time during which H-bonds remain intact before dissociation.

4. Jump frequency

The jump frequency (f_{jump}) quantifies how often a molecule (e.g., a proton or water molecule) changes its H-bonds partner. It is calculated based on the time intervals between successive jumps. The waiting time between two jumps, denoted as τ_{jump} , is determined by identifying the moments at which a proton or molecule switches bonding partners. The jump frequency is given by the inverse of the mean waiting time:

$$f_{\text{jump}} = \frac{1}{\tau_{\text{jump}}} \quad (\text{S12})$$

where τ_{jump} denotes the average time between two consecutive jumps.

5. Mean residence time

The mean residence time (τ_{res}) describes the average duration a molecule (e.g., water or proton) remains within a specific spatial region (e.g., a coordination shell or confined domain) before leaving. It can be calculated using the residence correlation function $S(t)$:

$$S(t) = \langle S_i(0) \cdot S_i(t) \rangle \quad (\text{S13})$$

where $S_i(t)$ is defined as 1 if particle i is within the region at time t , and 0 otherwise. The mean residence time is obtained by integrating the residence correlation function $S(t)$:

$$\tau_{\text{res}} = \int_0^{\infty} S(t) dt \quad (\text{S14})$$

Alternatively, it can be calculated as the mean of the residence times Δt_i of individual molecules:

$$\tau_{\text{res}} = \frac{1}{N} \sum_i \Delta t_i \quad (\text{S15})$$

where N is the number of molecules considered.

6. Permeation barrier

The permeation barrier, or free energy barrier, is critical for understanding molecular diffusion across membranes or through channels. In MD simulations, the free energy profile along the permeation coordinate (z) is typically calculated using methods such as umbrella sampling or metadynamics.

The potential of mean force (PMF) along the coordinate z is related to the probability distribution $P(z)$:

$$W(z) = -K_B T \ln P(z) \quad (\text{S16})$$

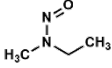
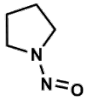
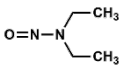
where K_B is the Boltzmann constant and T is the temperature. The energy barrier is given by the difference between the maximum and minimum values of $W(z)$:

$$E_{\text{permeation barrier}} = W(z_{\text{max}}) - W(z_{\text{min}}) \quad (\text{S17})$$

The maximum corresponds to the free energy barrier that needs to be overcome for permeation to occur.

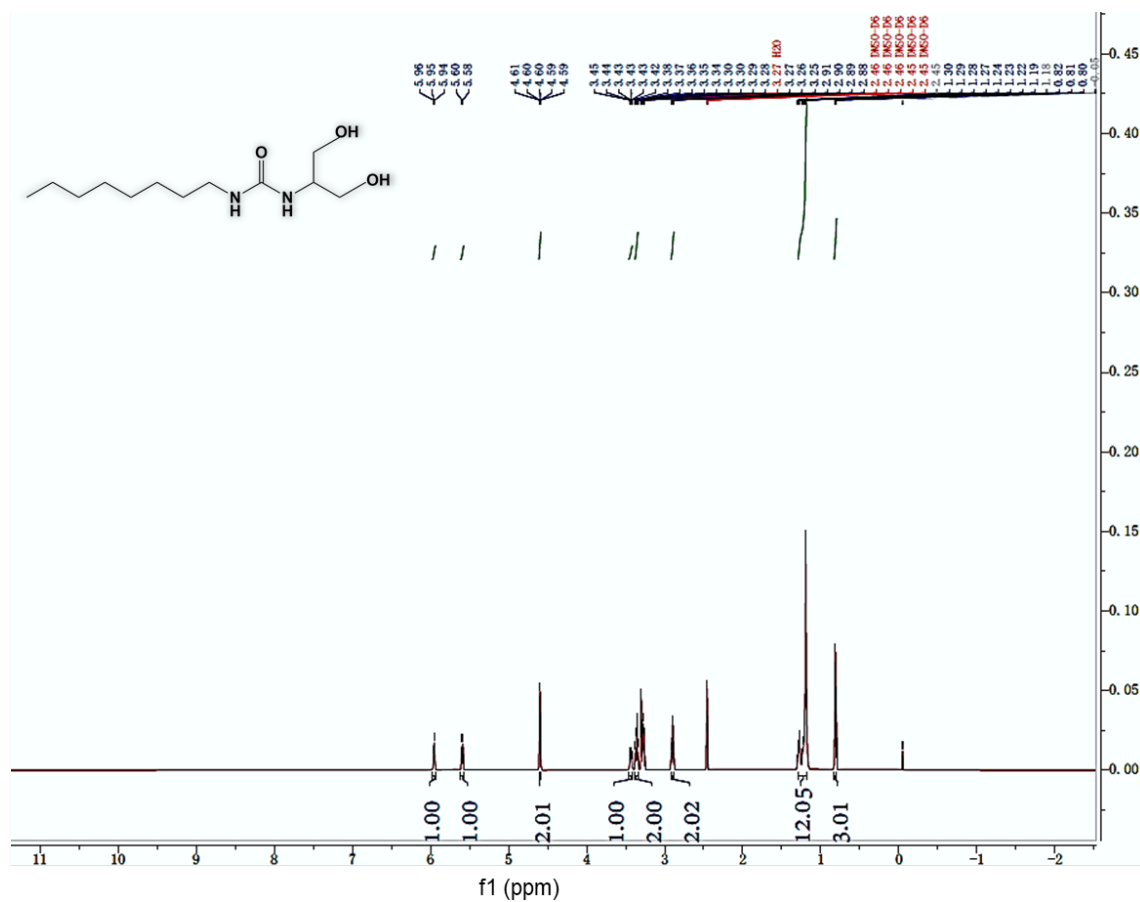
Supplementary Table

Supplementary Table S1. Physicochemical properties and mass spectrometry transitions and parameters of selected nitrosamines

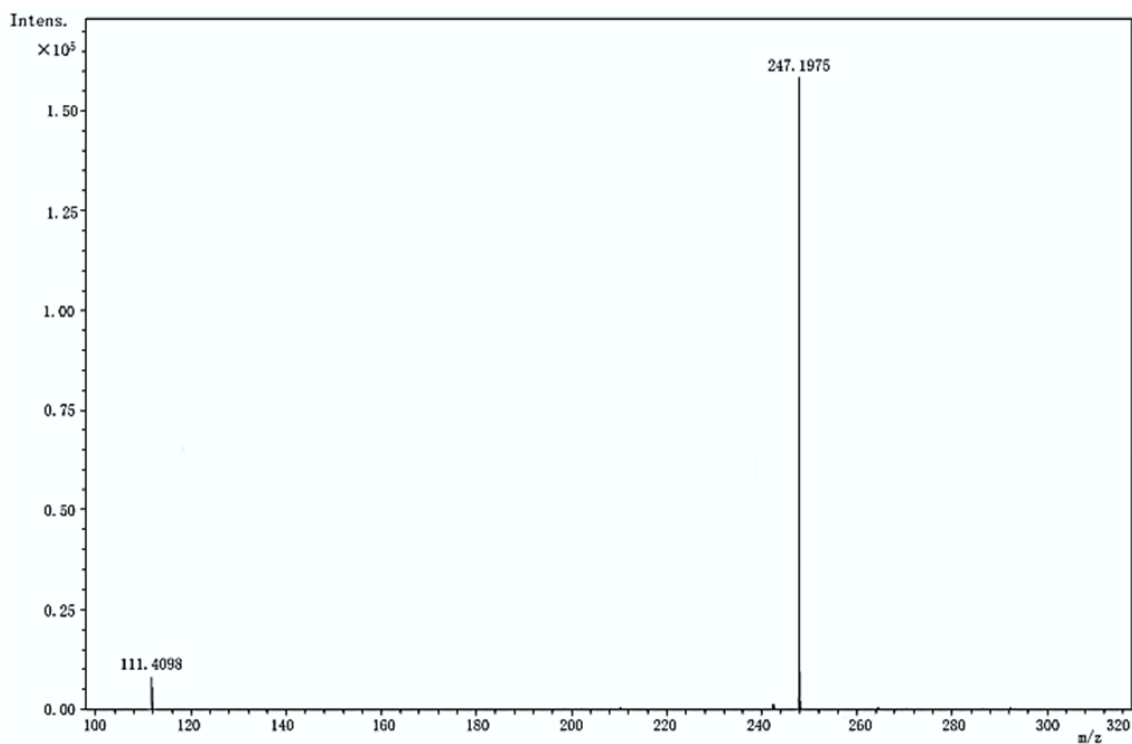
Compound	Structure	Molecular weight	pKa	RT (min)	DP (V)	CE (V)	CXP (V)	Precursor (m/z)	Product (m/z)
NDMA		74.1 Da	3.5	0.49	1	17	28	75..1	58.0
NMEA		88.1 Da	3.4	0.81	1	15	8	89.1	60.9
NPYR		100.1 Da	3.3	0.81	1	21	8	101.1	55.1
NDEA		102.1 Da	3.3	1.58	56	17	10	103.1	75
NPIP		114.1 Da	3.3	1.93	56	21	26	115.1	69.4
NMOR		116.1 Da	3.1	0.58	81	17	12	117.1	87.1

Notes: RT is retention time, DP is declustering potential, CE is collision energy, and CXP is collision cell exit potential.

Supplementary Figures

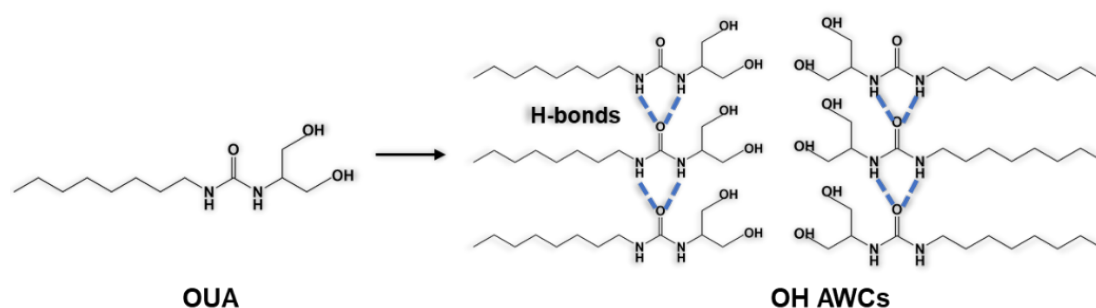


Supplementary Fig. 1. Proton nuclear magnetic resonance (¹H NMR) spectrum of OUA (1-(1,3-dihydroxypropan-2-yl)-3-octylurea).



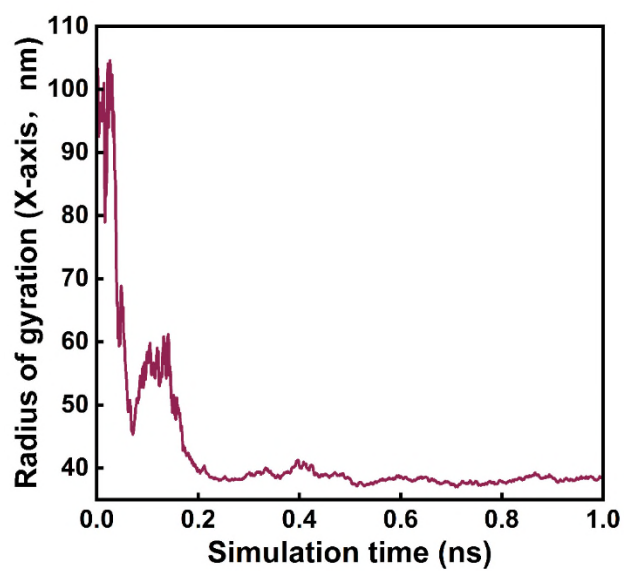
Supplementary Fig. 2. Electrospray ionization mass spectrometry (ESI-MS) spectrum of OUA (1-(1,3-dihydroxypropan-2-yl)-3-octylurea).

Supplementary Note: Proton nuclear magnetic resonance (¹H NMR) and electrospray ionization mass spectrometry (ESI-MS) (Supplementary Figs. 1-2) confirmed the successful synthesis of the precursor OUA (1-(1,3-dihydroxypropan-2-yl)-3-octylurea), which subsequently self-assembled into OH AWCs in a water/ethanol solution.

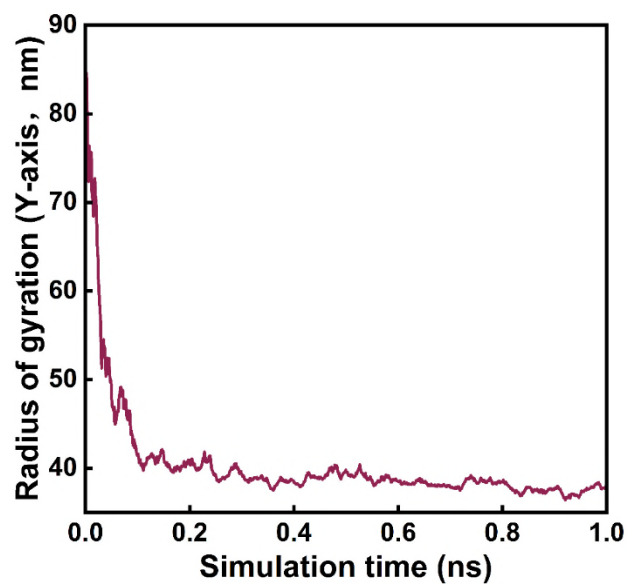


Supplementary Fig. 3. Chemical structures of OUA (1-(1,3-dihydroxypropan-2-yl)-3-octylurea) and hydroxy artificial water channels (OH AWCs) self-assembled from OUA through hydrogen bonds (H-bonds).

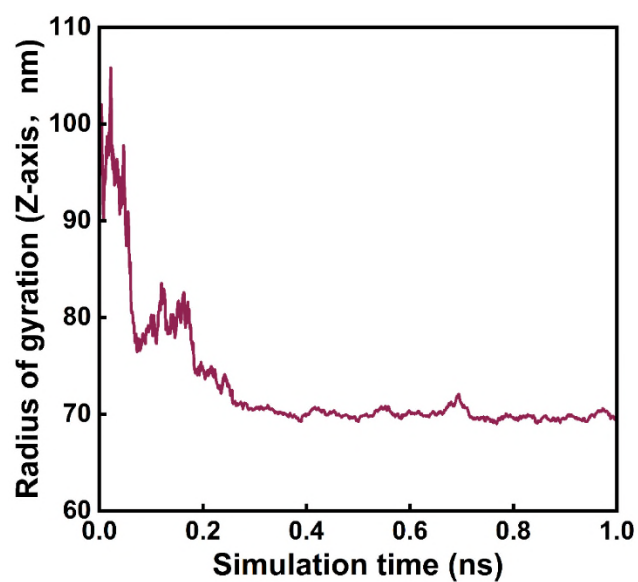
Supplementary Note: The OUA molecule consists of a hydrophobic alkyl chain and a hydrophilic polyhydroxy head group connected by a urea linkage (-NH-CO-NH-). The urea segment is capable of forming directional H-bonds, which drive the formation of an ordered channel structure through H-bonds interactions in water/ethanol solution.



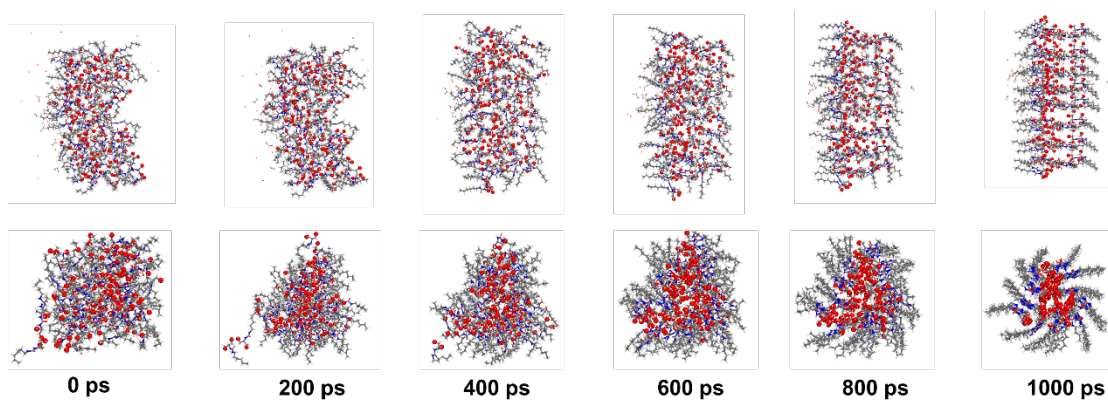
Supplementary Fig. 4. Variation of the radius of gyration (X-axis) of 1-(1,3-dihydroxypropan-2-yl)-3-octylurea (OUA) molecules in water/ethanol solution as a function of simulation time.



Supplementary Fig. 5. Variation of the radius of gyration (Y-axis) of 1-(1,3-dihydroxypropan-2-yl)-3-octylurea (OUA) molecules in water/ethanol solution as a function of simulation time.

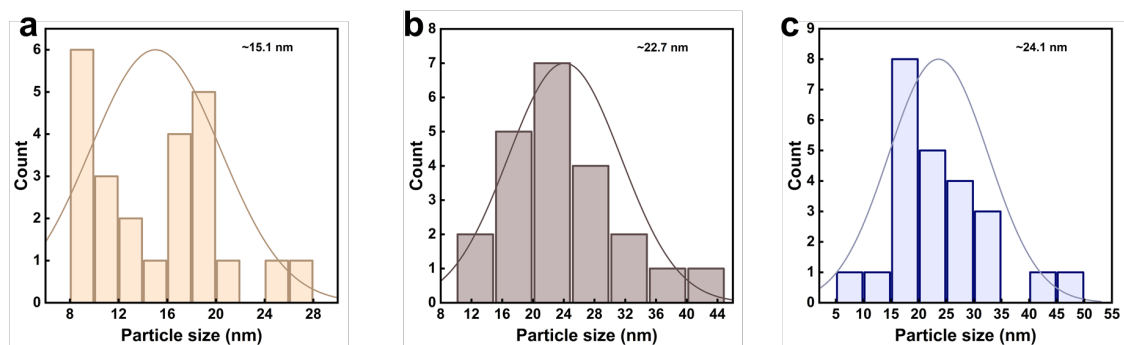


Supplementary Fig. 6. Variation of the radius of gyration (Z-axis) of 1-(1,3-dihydroxypropan-2-yl)-3-octylurea (OUA) molecules in water/ethanol solution as a function of simulation time.



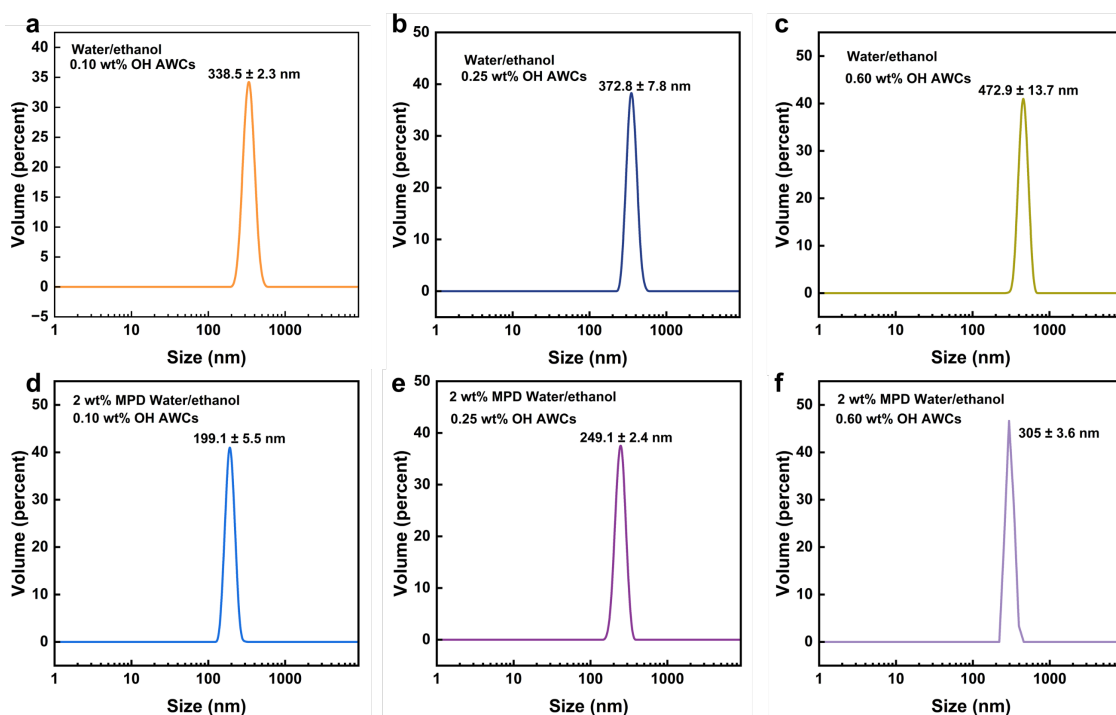
Supplementary Fig. 7. The self-assembly process of 1-(1,3-dihydroxypropan-2-yl)-3-octylurea (OUA) into hydroxy artificial water channels (OH AWCs) in water/ethanol solution.

Supplementary Note: MD simulations further confirmed that the radius of gyration of OUA decreased (the molecules assumed a more compact conformation and underwent self-assembly) in water/ethanol solution (Supplementary Figs. 4-6), and that OUA self-assembled into ordered OH AWCs through H-bond interactions (Supplementary Fig. 7), exhibiting well-defined structural diffraction peaks.



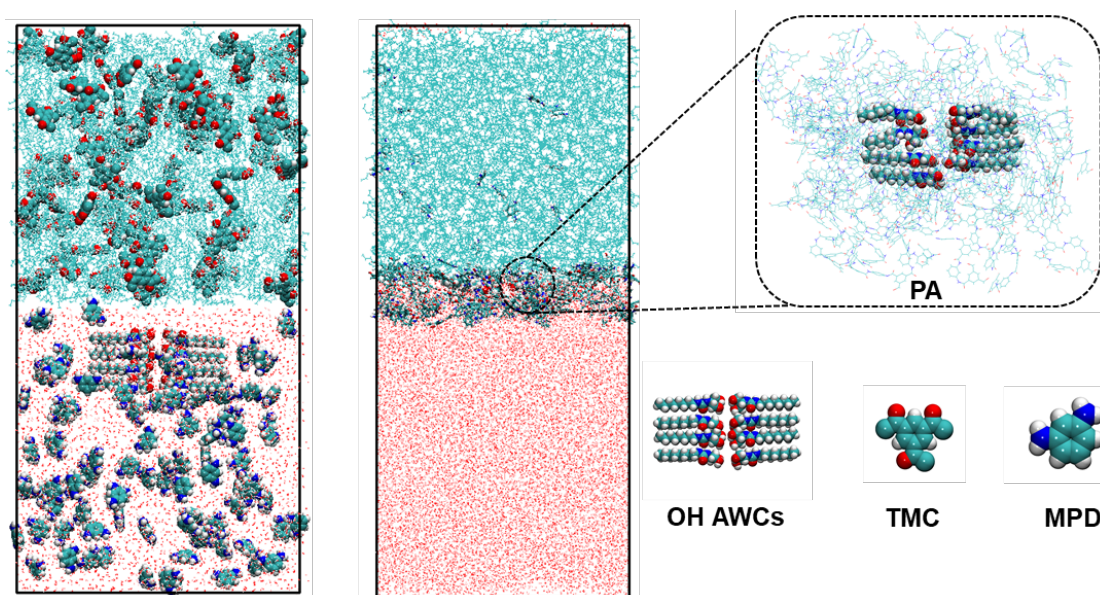
Supplementary Fig. 8. Size distribution of hydroxy artificial water channels (OH AWCs) at (a) 0.10 wt% (24 particles were measured for each sample), (b) 0.25 wt% (13 particles were measured for each sample), and (c) 0.60 wt% (25 particles were measured for each sample), calculated from TEM images using Nano Measurer 1.2 software.

Supplementary Note: As the OH AWC concentration increased from 0.10 to 0.60 wt%, the size of individual OH AWCs increased from ~15.1 nm to 24.1 nm.

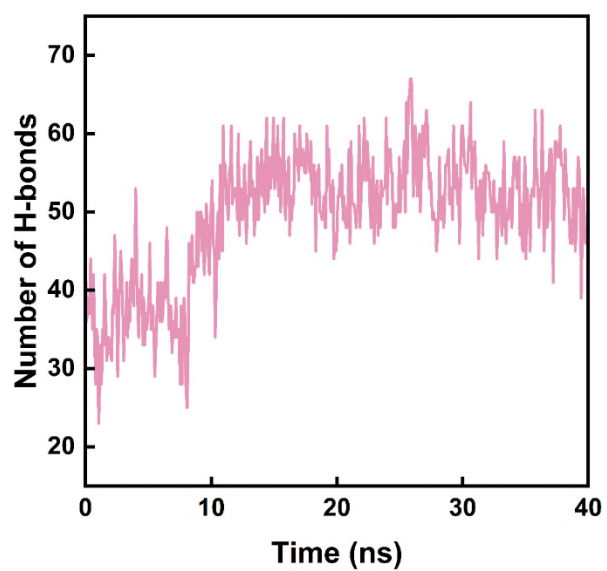


Supplementary Fig. 9. Dynamic light scattering was conducted to investigate the formation of colloidal aggregates of hydroxy artificial water channels (OH AWCs) at various concentrations in water/ethanol and 2 wt% *m*-phenylenediamine (MPD) water/ethanol (water/ethanol volume ratio of 1:1). (a-c) The size distribution of colloidal aggregates formed by varying amounts of OH AWCs in water/ethanol solutions. (d-f) The size distribution of colloidal aggregates formed by varying amounts of OH AWCs in 2 wt% MPD water/ethanol solutions. The data in the figure represented the results from three experiments, with the values presented as the mean values $\pm$ s.d.

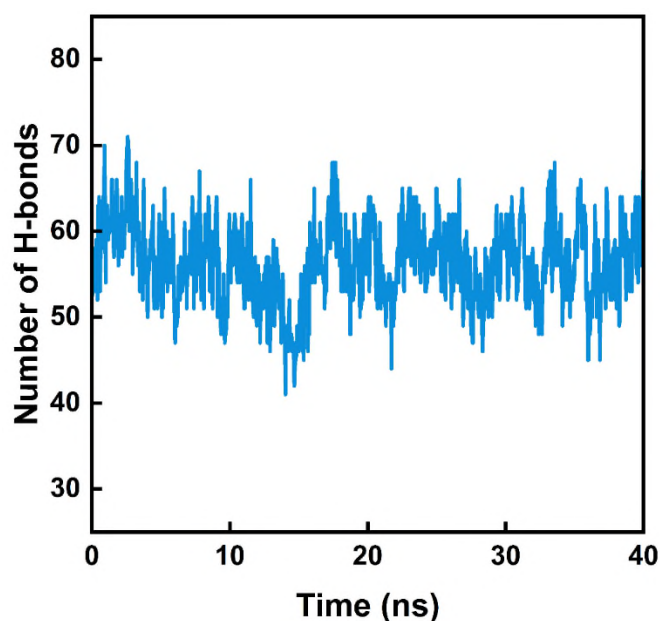
Supplementary Note: The size of colloidal aggregates formed by OH AWCs in water/ethanol solutions was greater than that formed in colloidal aggregates formed by OH AWCs in 2 wt% MPD water/ethanol solutions, indicating the presence of H-bonds interactions between OH AWCs colloidal aggregates and MPD. The H-bonds interactions between MPD and OH AWCs facilitate the integration of OH AWCs into the PA layer during the IP process.



Supplementary Fig. 10. Molecular dynamics (MD) simulation of the process by which hydroxy artificial water channels (OH AWCs) are incorporated into the polyamide (PA) layer of RO membranes.

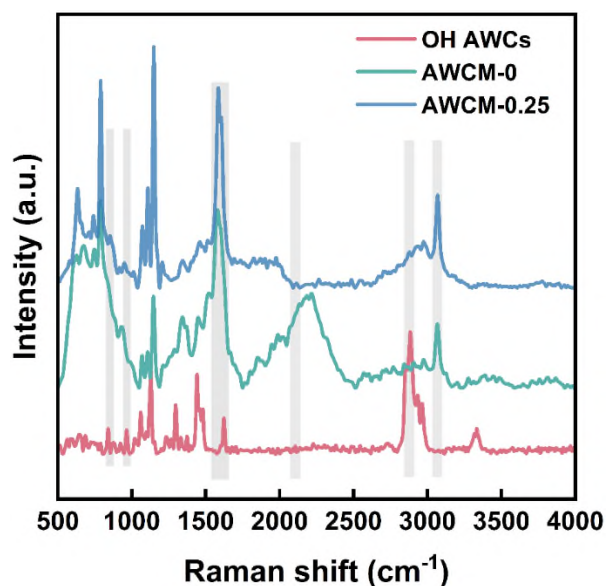


Supplementary Fig. 11. Hydrogen bonds (H-bonds) interactions between hydroxy artificial water channels (OH AWCs) and *m*-phenylenediamine (MPD).



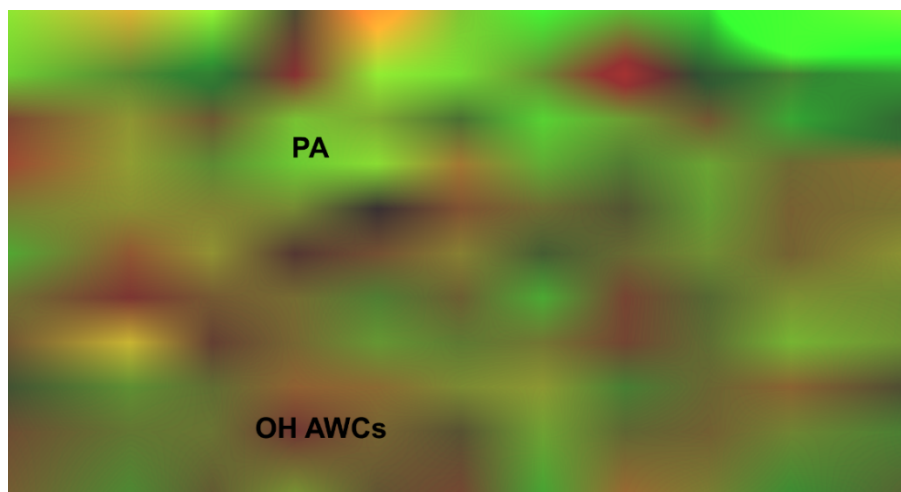
Supplementary Fig. 12. Hydrogen bonds (H-bonds) interactions between hydroxy artificial water channels (OH AWCs) and polyamide (PA) layer.

Supplementary Note: MD simulations further confirmed that the urea groups on the branched chains of OH AWCs interact with the -NH_2 groups of MPD via H-bonds to form MPD-OH AWCs assembled phases, which subsequently react with trimesoyl chloride (TMC) to enable the incorporation of OH AWCs into the PA layer (where OH AWCs are fixed within the PA matrix through H-bonds interactions) (Supplementary Figs. 10-12).



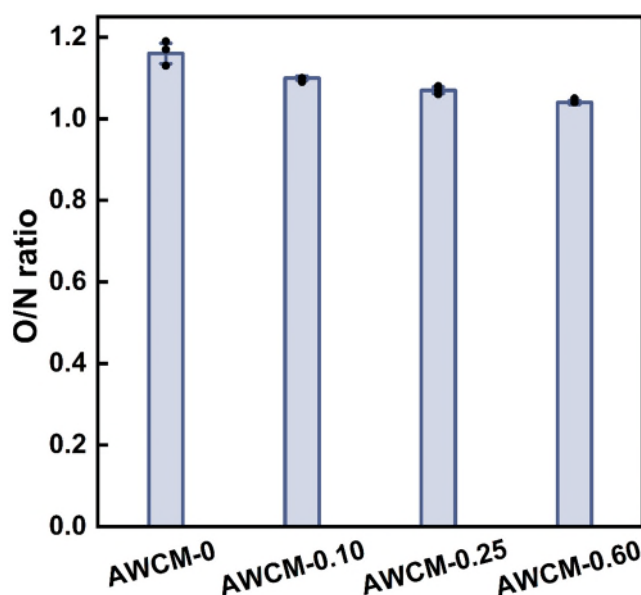
Supplementary Fig. 13. Raman spectra of hydroxy artificial water channels (OH AWCs), AWCM-0 (without OH AWCs) and AWCM-0.25 (RO membranes modified with OH AWCs self-assembled from 0.25 wt% 1-(1,3-dihydroxypropan-2-yl)-3-octylurea) at entire Raman shift range (500-4000 cm^{-1}). The light-gray regions indicate the characteristic peaks of OH AWCs.

Supplementary Note: Peaks from both OH AWCs and PA were simultaneously detected on the surface of AWCM-0.25, indicating the successful incorporation of OH AWCs into the PA layer.



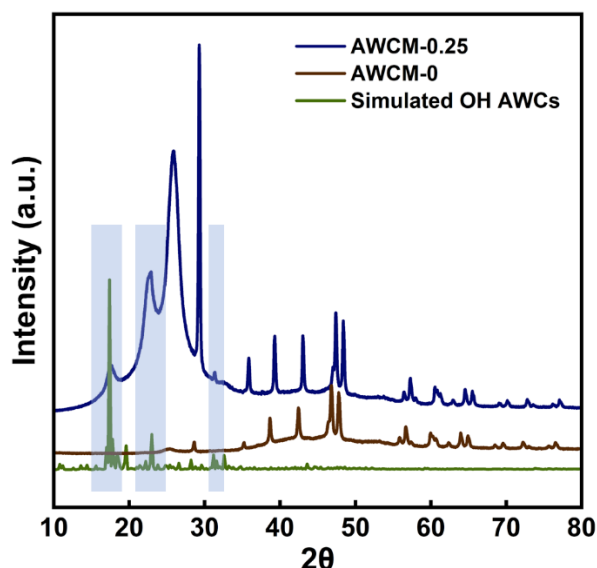
Supplementary Fig. 14. Raman spectroscopic mapping images of AWCM-0.25 (RO membranes modified with OH AWCs self-assembled from 0.25 wt% 1-(1,3-dihydroxypropan-2-yl)-3-octylurea). The red color represents OH AWCs, and the green color represents PA.

Supplementary Note: Raman mapping demonstrated the coexistence of peaks from both OH AWCs and PA on the surface of AWCM-0.25, further confirming the integration of OH AWCs into the PA layer.



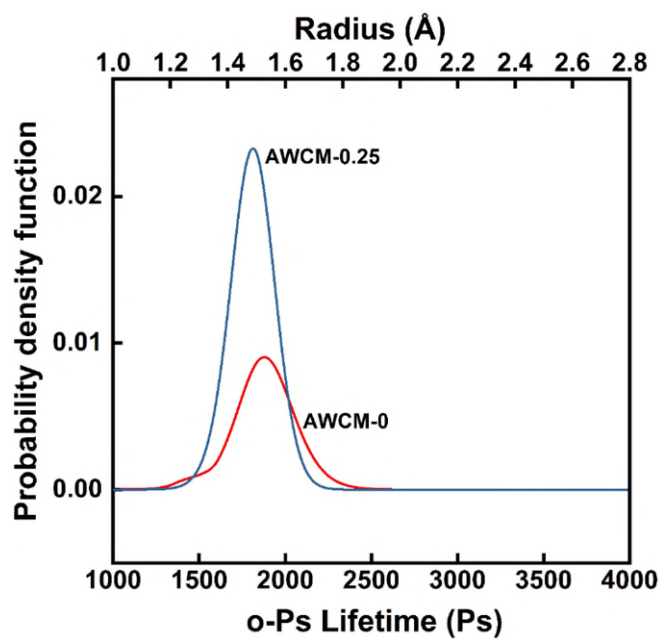
Supplementary Fig. 15. The O/N ratio of AWCM-0 (without OH AWCs), AWCM-0.10, AWCM-0.25, and AWCM-0.60 membranes (AWCM-0.10, AWCM-0.25, and AWCM-0.60 represented RO membranes modified with OH AWCs self-assembled from 0.1 wt%, 0.25 wt%, and 0.6 wt% 1-(1,3-dihydroxypropan-2-yl)-3-octylurea, respectively). Error bars represent the s.d. ($n = 3$) and data are presented as mean values $\pm$ s.d.

Supplementary Note: The O/N ratio of the AWCM decreased with increasing OH AWCs content, possibly because the increased cross-linking degree of the PA (which leads to a decrease in the O/N ratio) offset the enhancement in O/N ratio resulting from the incorporation of OH AWCs.

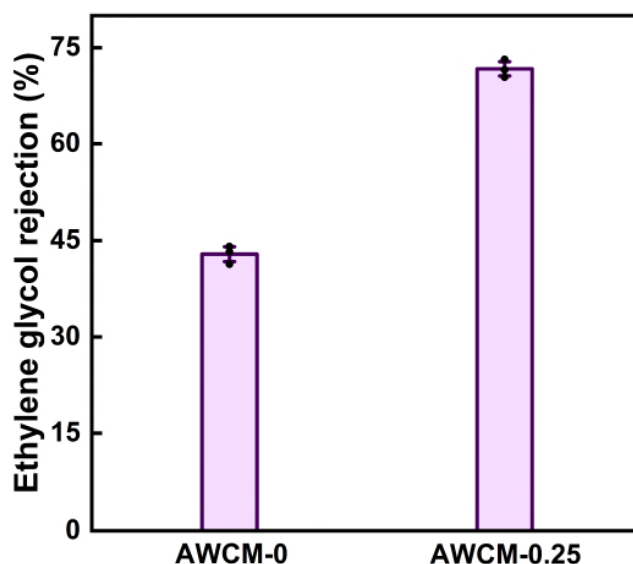


Supplementary Fig. 16. Simulated X-ray diffraction (XRD) pattern of OH AWCs and experimentally measured XRD patterns of AWCM-0 (without OH AWCs) and AWCM-0.25 (RO membranes modified with OH AWCs self-assembled from 0.25 wt% 1-(1,3-dihydroxypropan-2-yl)-3-octylurea). The light-blue regions indicate the simulated XRD peaks of OH AWCs.

Supplementary Note: X-ray diffraction characterization was further performed to analyze the structure of OH AWCs within the PA layer. Compared with the AWCM-0, the AWCM-0.25 exhibited a red shift in the diffraction peak, which was attributed to the incorporation of OH AWCs into the PA layer. Moreover, characteristic diffraction peaks of OH AWCs were detected on the surface of the AWCM membrane, indicating that OH AWCs retained their intrinsic structural features within the PA layer.

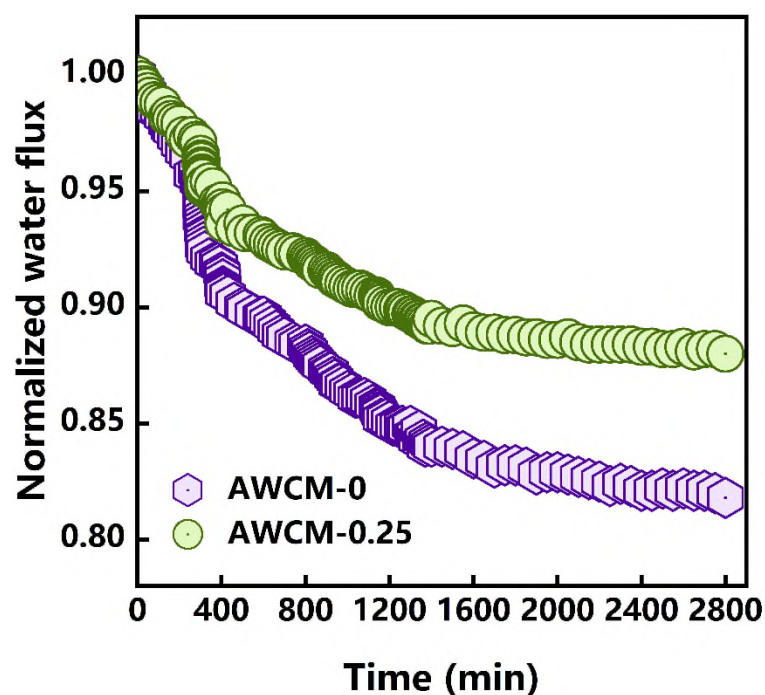


Supplementary Fig. 17. The probability density distributions of pore sizes in AWCM-0 (without OH AWCs) and AWCM-0.25 (RO membranes modified with OH AWCs self-assembled from 0.25 wt% 1-(1,3-dihydroxypropan-2-yl)-3-octylurea) were obtained from the positron annihilation lifetime spectrum.

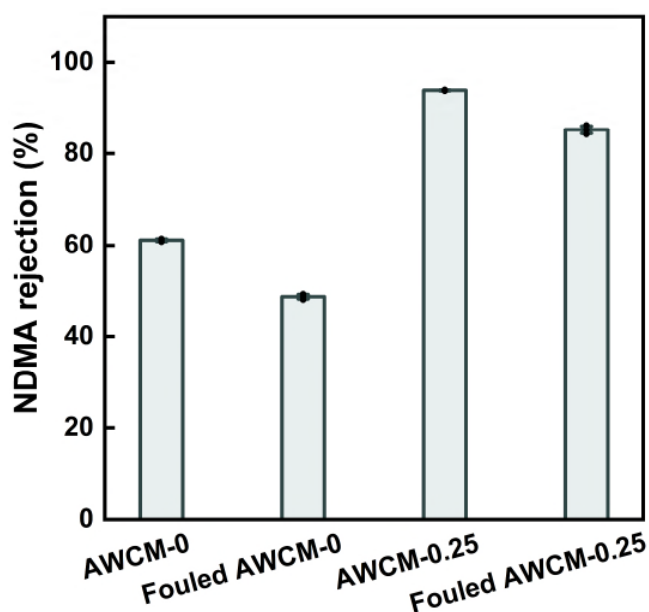


Supplementary Fig. 18. Rejection of ethylene glycol by AWCM-0 (without OH AWCs) and AWCM-0.25 membranes (RO membranes modified with OH AWCs self-assembled from 0.25 wt% 1-(1,3-dihydroxypropan-2-yl)-3-octylurea) (concentration: 40 mg L⁻¹; applied pressure: 16 bar; temperature: 25°C). Error bars represent the s.d. ($n = 3$), and data are presented as mean $\pm$ s.d.

Supplementary Note: Positron annihilation lifetime spectroscopy and neutral molecular rejection experiments confirmed that AWCM-0.25 exhibits a narrower pore size distribution (Supplementary Figs. 17-18).

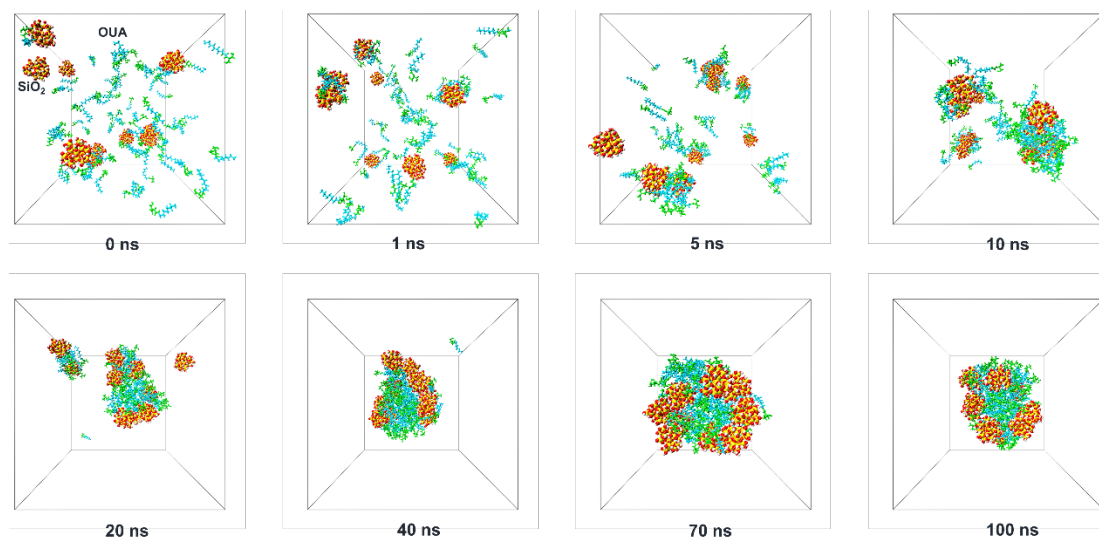


Supplementary Fig. 19. Normalized water flux as a function of time for AWCm-0 (without OH AWCs) and AWCm-0.25 membranes (RO membranes modified with OH AWCs self-assembled from 0.25 wt% 1-(1,3-dihydroxypropan-2-yl)-3-octylurea) during the antifouling experiments. The flux decline curves represent averaged data from duplicate fouling tests. A mixed solution containing 100 mg L⁻¹ HA and 2000 mg L⁻¹ NaCl was used to evaluate the antifouling performance of the membranes (initial flux of ~35 L m⁻² h⁻¹, pH=7, 25°C).

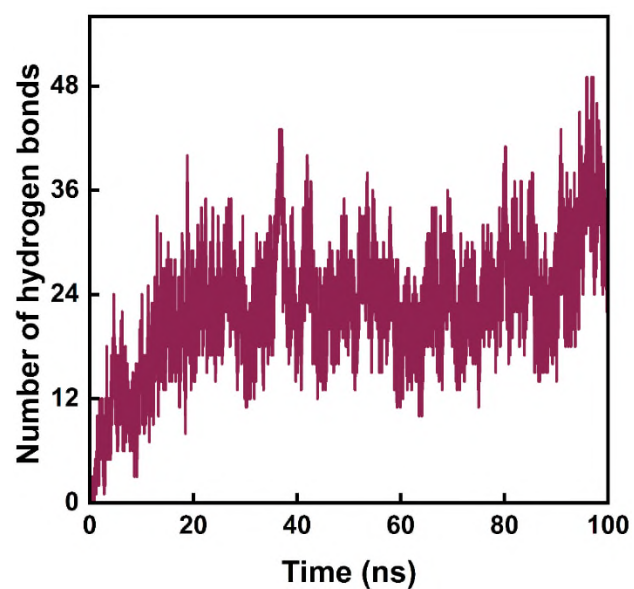


Supplementary Fig. 20. *N*-nitrosodimethylamine (NDMA) rejection of AWCM-0 (without OH AWCs) and AWCM-0.25 membranes (RO membranes modified with OH AWCs self-assembled from 0.25 wt% 1-(1,3-dihydroxypropan-2-yl)-3-octylurea) before fouling and under the same degree of HA fouling. Error bars represent the s.d. ($n = 3$), and data are presented as mean $\pm$ s.d.

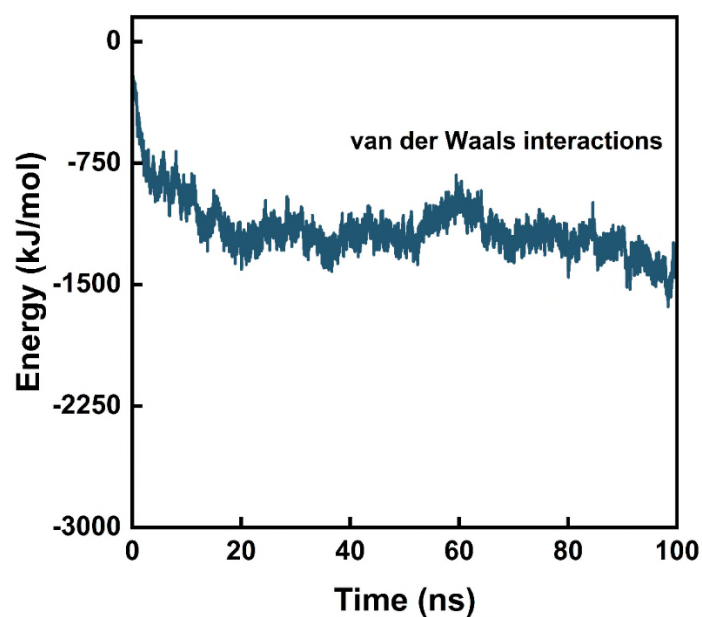
Supplementary Note: AWCM-0.25 exhibited superior resistance to humic acid (HA) fouling compared with AWCM-0 (Supplementary Fig. 19), indicating that the incorporation of OH AWCs into the RO membranes not only enhanced the rejection of neutral small molecule pollutants but also imparted antifouling performance to the membranes. Notably, when the membranes were fouled to the same degree, NDMA rejection decreased for both AWCM-0 and AWCM-0.25 (Supplementary Fig. 20). Compared with their pristine counterparts, AWCM-0.25 (decrease of $\sim 9.3\%$) exhibited a smaller decline than AWCM-0 (decrease of $\sim 20.9\%$), indicating that the incorporation of OH AWCs into the RO membrane could partially mitigate the adverse effects of membrane fouling.



Supplementary Fig. 21. Snapshots of the overall structures of SiO_2 and 1-(1,3-dihydroxypropan-2-yl)-3-octylurea (OUA) self-assembly at different simulation times.

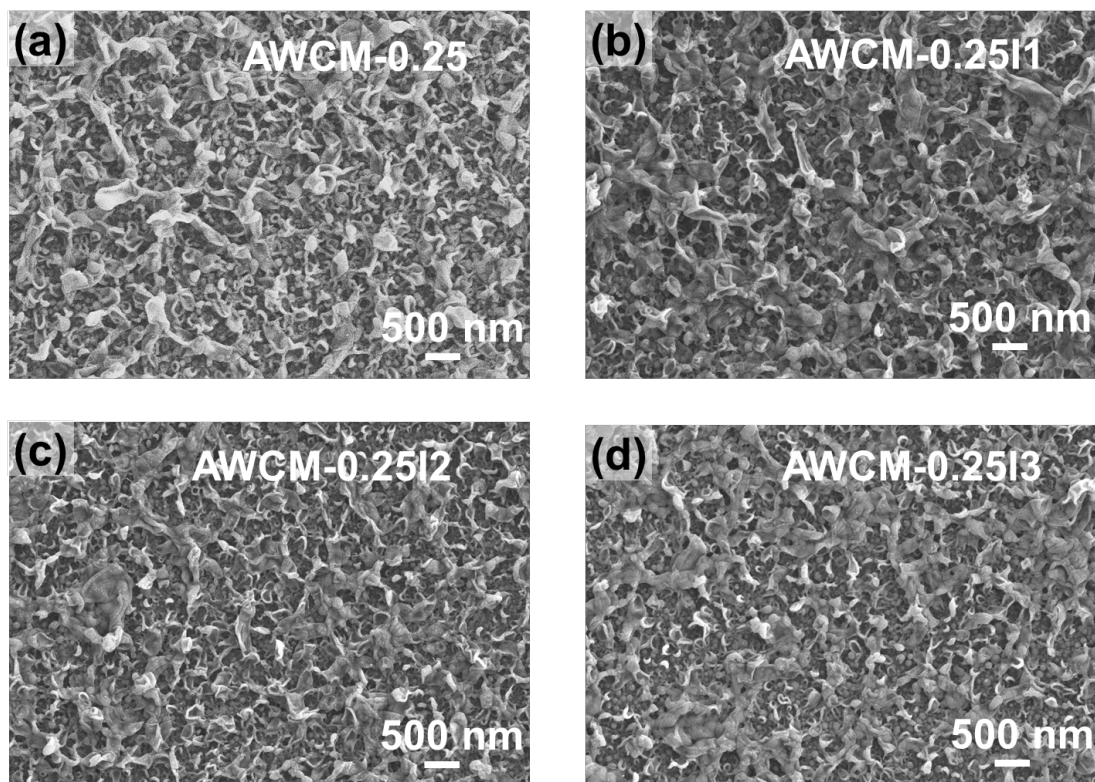


Supplementary Fig. 22. Changes in the number of H-bonds between SiO₂ and 1-(1,3-dihydroxypropan-2-yl)-3-octylurea (OUA) during the self-assembly simulation.



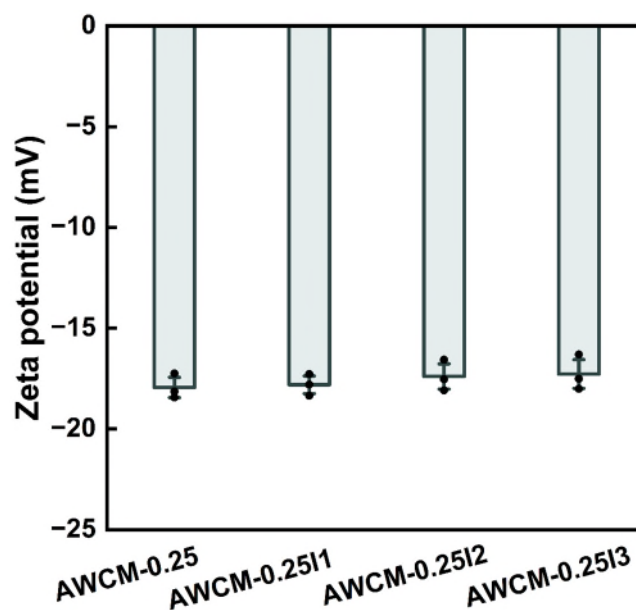
Supplementary Fig. 23. Van der Waals interactions between SiO₂ and 1-(1,3-dihydroxypropan-2-yl)-3-octylurea (OUA) during the self-assembly simulation.

Supplementary Note: MD simulations revealed that SiO₂ and OUA in a water/ethanol solution self-assemble into a stable aggregate via H-bonds (formed between Si-OH groups and the -OH, -C=O, and -NH groups in the OUA structure) and van der Waals interactions (arising from the alkyl chains of OUA and Si atoms) (Supplementary Figs. 21-23), thereby inhibiting the formation of OH AWCs.

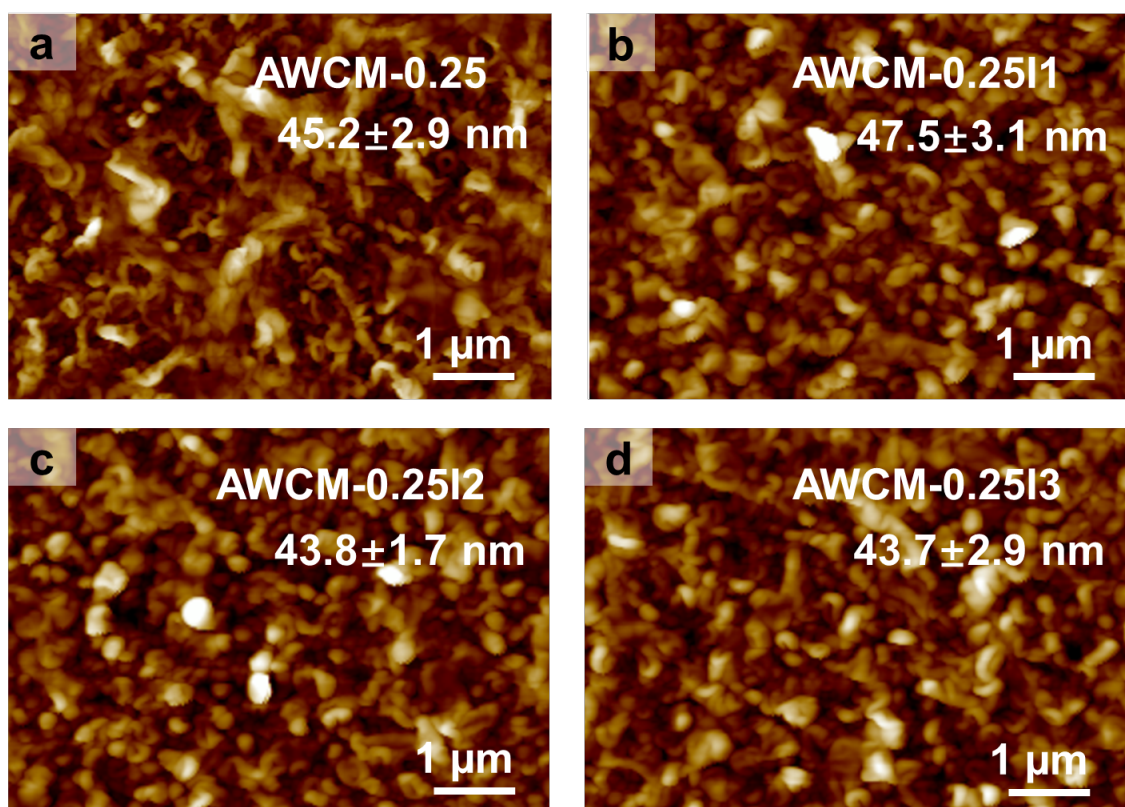


Supplementary Fig. 24. Morphology of the AWCM-0.25, AWCM-0.25I1, AWCM-0.25I2, and AWCM-0.25I3 membranes (AWCM-0.25, AWCM-0.25I1, AWCM-0.25I2, and AWCM-0.25I3 represent the SiO₂@OUA modified RO membranes assembled at SiO₂ and OUA mass ratios of 0, 1:4, 1:2, and 1:1, respectively).

Supplementary Note: Compared to AWCM-0.25, there were no significant changes in the surface morphology of the AWCM-0.25I membranes (Supplementary Fig. 24), indicating that the incorporation of SiO₂@OUA did not affect the IP process.

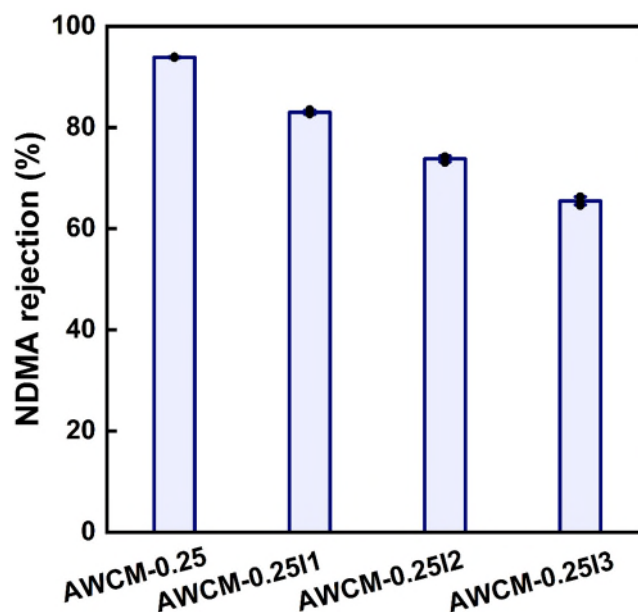


Supplementary Fig. 25. The surface charge of AWCM-0.25, AWCM-0.25I1, AWCM-0.25I2, and AWCM-0.25I3 membranes (AWCM-0.25, AWCM-0.25I1, AWCM-0.25I2, and AWCM-0.25I3 represent the SiO₂@OUA modified RO membranes assembled at SiO₂ and OUA mass ratios of 0, 1:4, 1:2, and 1:1, respectively). Error bars represent the s.d. ($n = 3$) and data are presented as mean values $\pm$ s.d.



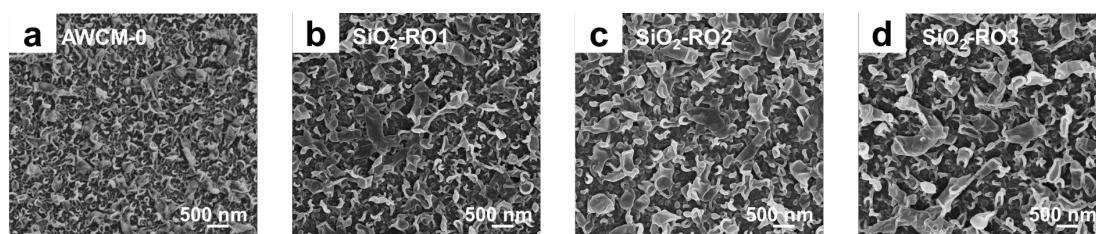
Supplementary Fig. 26. Atomic force microscopy (AFM) images of AWCM-0.25, AWCM-0.25I1, AWCM-0.25I2, and AWCM-0.25I3 membranes (AWCM-0.25, AWCM-0.25I1, AWCM-0.25I2, and AWCM-0.25I3 represent the SiO₂@OUA modified RO membranes assembled at SiO₂ and OUA mass ratios of 0, 1:4, 1:2, and 1:1, respectively). The corresponding membrane surface roughness data were shown in the AFM images. The data in the figure represented the results from three experiments, with the values presented as the mean values $\pm$ s.d.

Supplementary Note: Compared to AWCM-0.25, there were no significant changes in the surface charge (Supplementary Fig. 25) and surface roughness (Supplementary Fig. 26) of the AWCM-0.25I membranes.

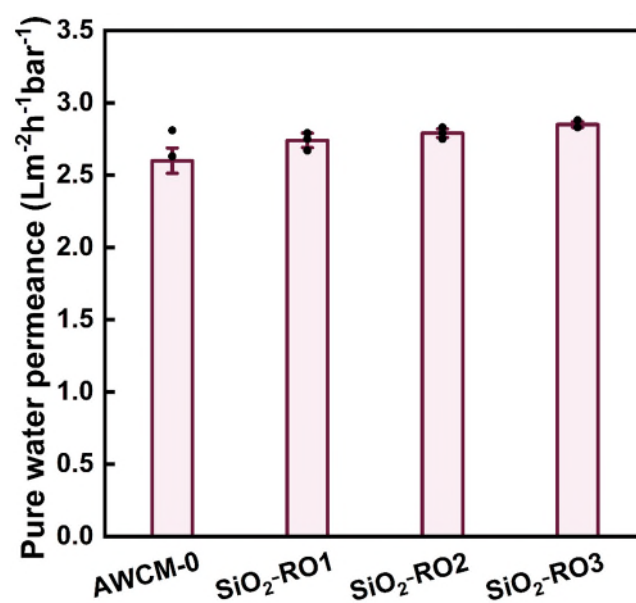


Supplementary Fig. 27. The *N*-nitrosodimethylamine (NDMA) rejection of the AWCM-0.25, AWCM-0.25I1, AWCM-0.25I2, and AWCM-0.25I3 (AWCM-0.25, AWCM-0.25I1, AWCM-0.25I2, and AWCM-0.25I3 represent the SiO₂@OUA modified RO membranes assembled at SiO₂ and OUA mass ratios of 0, 1:4, 1:2, and 1:1, respectively). Error bars represent the s.d. ($n = 3$), and data are presented as mean $\pm$ s.d.

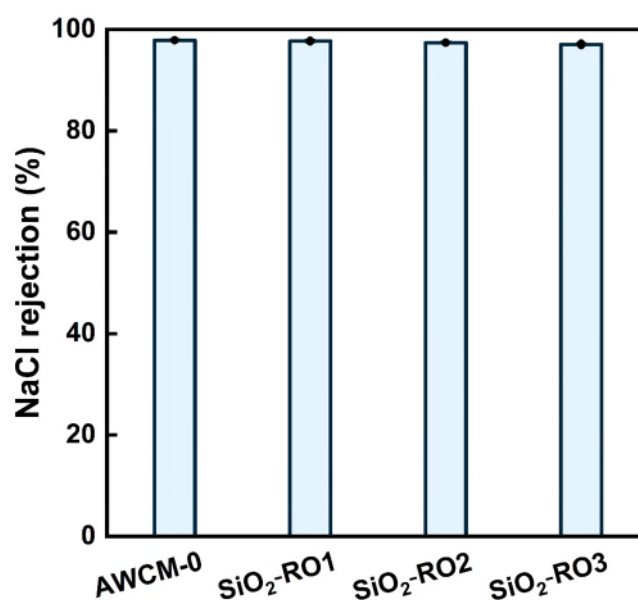
Supplementary Note: The NDMA rejection of the AWCM-0.25I decreased from $93.9 \pm 0.1\%$ to $65.5 \pm 0.8\%$ (Supplementary Fig. 27).



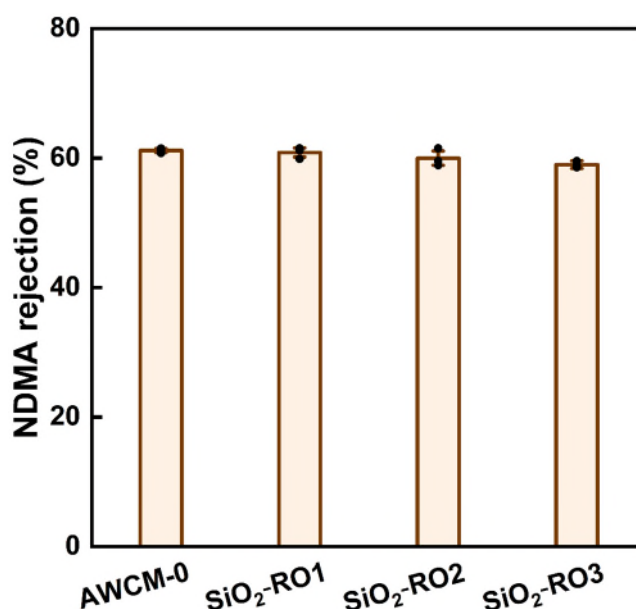
Supplementary Fig. 28. Morphology of the AWCM-0 (without OH AWCs), SiO₂-RO1, SiO₂-RO2, and SiO₂-RO3 membranes (SiO₂-RO1, SiO₂-RO2, and SiO₂-RO3 membranes denote RO membranes modified with different amounts of SiO₂ used to inhibit OUA self-assembly).



Supplementary Fig. 29. Pure water permeance of AWCm-0 (without OH AWCs) and SiO₂ modified RO membranes (SiO₂-RO1, SiO₂-RO2, and SiO₂-RO3 membranes denote RO membranes modified with different amounts of SiO₂ used to inhibit OUA self-assembly). Error bars represent the s.d. ($n = 3$), and data are presented as mean $\pm$ s.d.

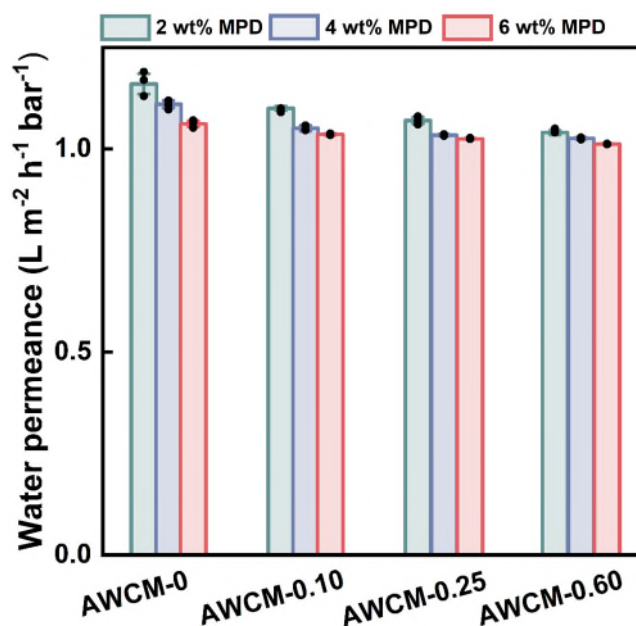


Supplementary Fig. 30. NaCl rejection of AWCm-0 (without OH AWCs) and SiO₂ modified RO membranes (SiO₂-RO1, SiO₂-RO2, and SiO₂-RO3 membranes denote RO membranes modified with different amounts of SiO₂ used to inhibit OUA self-assembly). Error bars represent the s.d. ($n = 3$) and data are presented as mean values $\pm$ s.d.



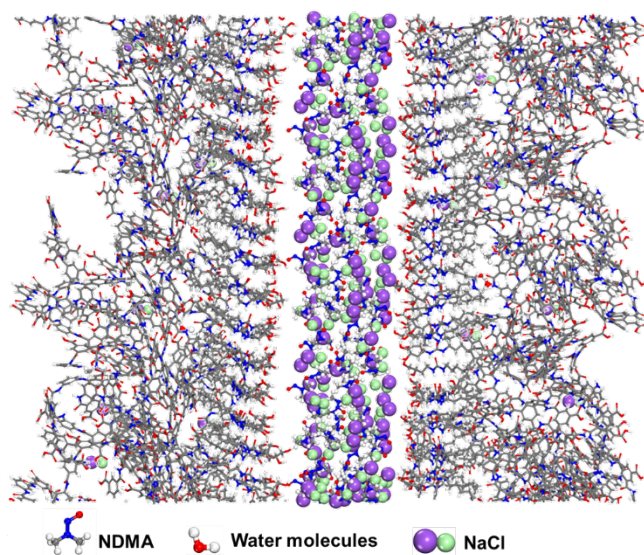
Supplementary Fig. 31. *N*-nitrosodimethylamine (NDMA) rejection of AWCM-0 (without OH AWCs) and SiO₂ modified RO membranes (SiO₂-RO1, SiO₂-RO2, and SiO₂-RO3 membranes denote RO membranes modified with different amounts of SiO₂ used to inhibit OUA self-assembly). Error bars represent the s.d. ($n = 3$) and data are presented as mean values $\pm$ s.d.

Supplementary Note: Compared with the AWCM-0 membrane, the surface morphology, pure water permeance, and NaCl and NDMA rejection of RO membranes modified solely with SiO₂ showed no significant changes (Supplementary Figs. 28-31), indicating that the decline in the separation performance of the AWCM-0.25I membrane was primarily attributed to the reduced quantity of available OH AWCs for water molecule and NaCl transport within the membrane, which shifted the primary transport pathways toward the PA matrix.

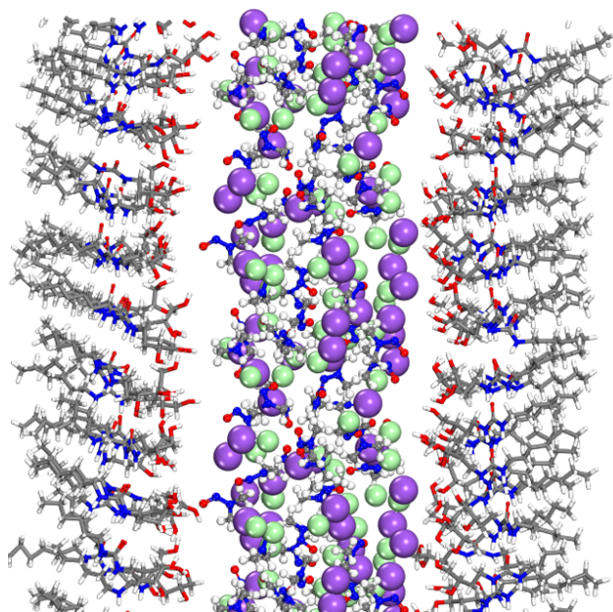


Supplementary Fig. 32. The O/N ratio of the AWCM-0 (without OH AWCs), AWCM-0.10, AWCM-0.25, and AWCM-0.60 membranes (AWCM-0.10, AWCM-0.25, and AWCM-0.60 represented RO membranes modified with OH AWCs self-assembled from 0.1 wt%, 0.25 wt%, and 0.6 wt% 1-(1,3-dihydroxypropan-2-yl)-3-octylurea, respectively) prepared with different *m*-phenylenediamine (MPD) monomer concentrations. Error bars represent the s.d. ($n = 3$) and data are presented as mean values $\pm$ s.d.

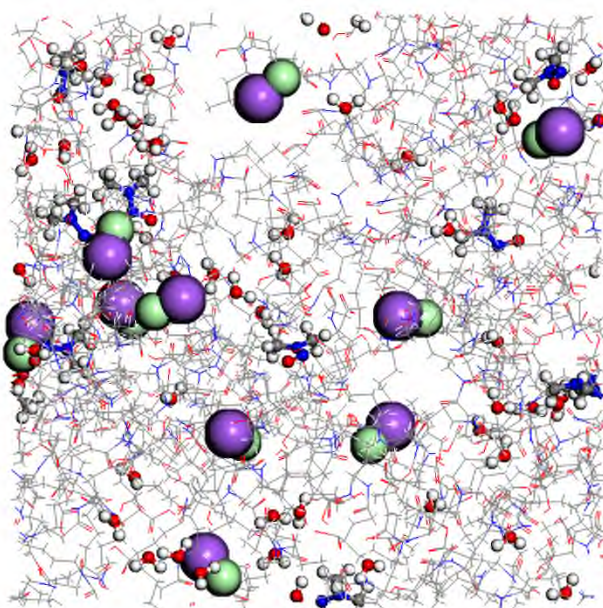
Supplementary Note: Increasing the concentration of MPD monomer leads to the formation of a denser PA layer, as evidenced by the decreased O/N ratio (Supplementary Fig. 32).



Supplementary Fig. 33. The hydroxy artificial water channels-polyamide (OH AWCs-PA) hybrid model used for molecular dynamics simulations.

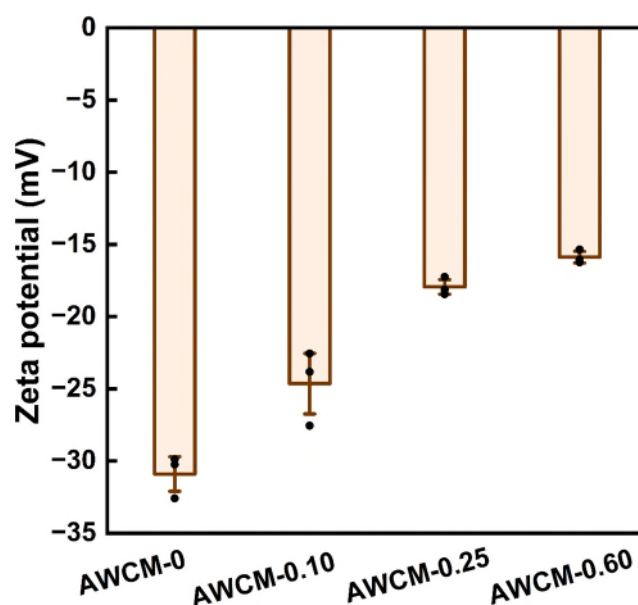


Supplementary Fig. 34. The hydroxy artificial water channels (OH AWCs) model used for molecular dynamics simulations.



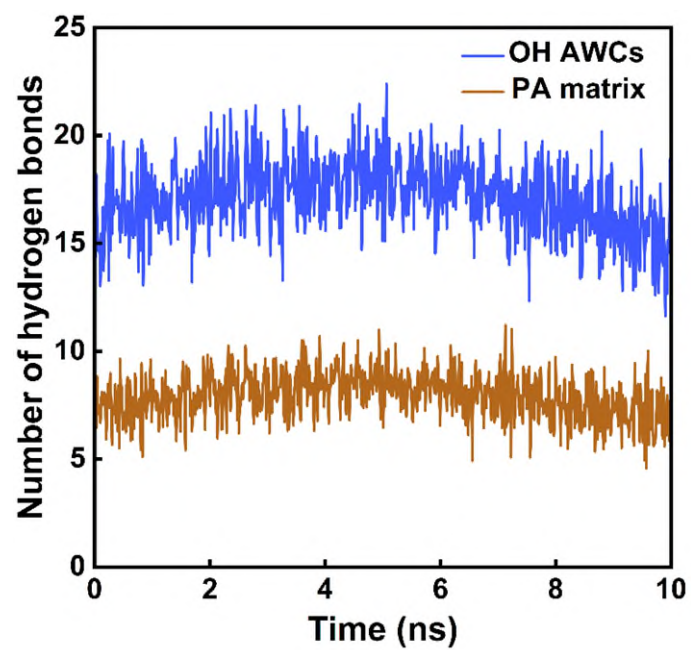
Supplementary Fig. 35. The polyamide (PA) model used for molecular dynamics simulations.

Supplementary Note: The AWCs-PA hybrid model (Supplementary Fig. 33), OH AWCs model (Supplementary Fig. 34), and PA matrix model (Supplementary Fig. 35) were employed to investigate the molecular mass transfer mechanisms.

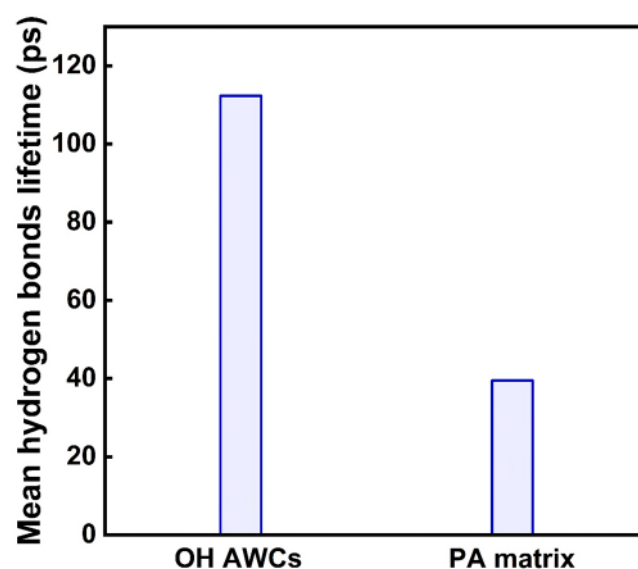


Supplementary Fig. 36. The surface charge of AWCM-0 (without OH AWCs), AWCM-0.10, AWCM-0.25, and AWCM-0.60 membranes (AWCM-0.10, AWCM-0.25, and AWCM-0.60 represented RO membranes modified with OH AWCs self-assembled from 0.1 wt%, 0.25 wt%, and 0.6 wt% 1-(1,3-dihydroxypropan-2-yl)-3-octylurea, respectively). Error bars represent the s.d. ($n = 3$), and data are presented as mean values $\pm$ s.d.

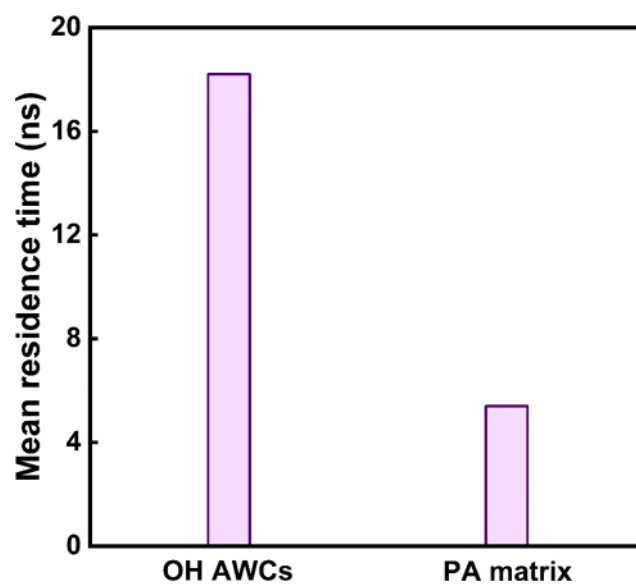
Supplementary Note: Under neutral conditions, GNPs are negatively charged and exhibit similar surface charge behavior to RO membranes (Supplementary Fig. 36).



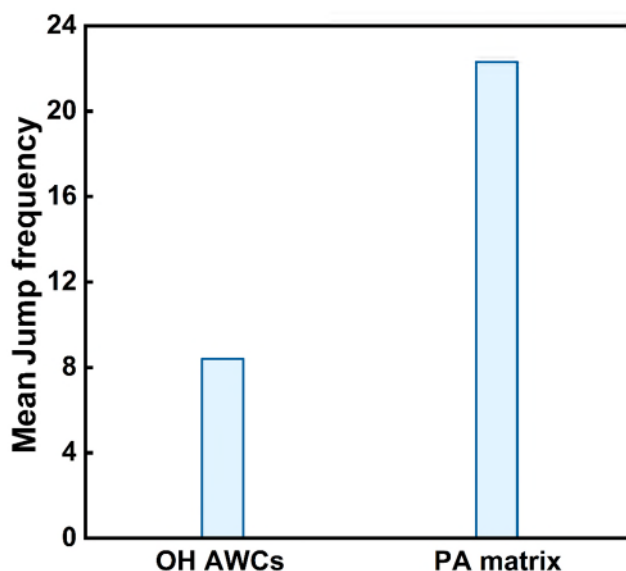
Supplementary Fig. 37. Number of hydrogen bonds formed during *N*-nitrosodimethylamine (NDMA) transport in hydroxy artificial water channels (OH AWCs) and the polyamide (PA) matrix.



Supplementary Fig. 38. Mean hydrogen bonds lifetime during *N*-nitrosodimethylamine (NDMA) transport in hydroxy artificial water channels (OH AWCs) and the polyamide (PA) matrix.

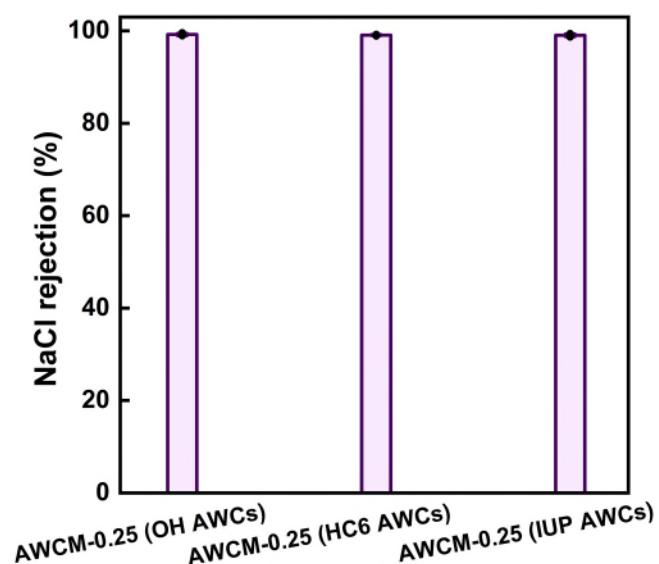


Supplementary Fig. 39. Mean residence time of *N*-nitrosodimethylamine (NDMA) during transport in hydroxy artificial water channels (OH AWCs) and the polyamide (PA) matrix.



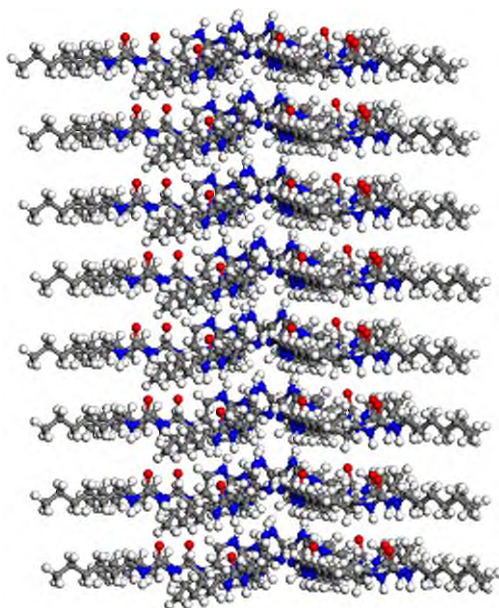
Supplementary Fig. 40. Mean jumping frequency of *N*-nitrosodimethylamine (NDMA) during transport in hydroxy artificial water channels (OH AWCs) and the polyamide (PA) matrix.

Supplementary Note: For NDMA, during transport through the OH AWCs, a greater number of H-bonds were formed than in the PA matrix (Supplementary Fig. 37), and these H-bonds exhibited longer lifetimes (Supplementary Fig. 38), leading to prolonged residence times and lower jumping frequencies (Supplementary Figs. 39-40), consequently OH AWCs mediated transport pathways conferred a higher permeation energy barrier for NDMA and superior rejection in the AWCs.

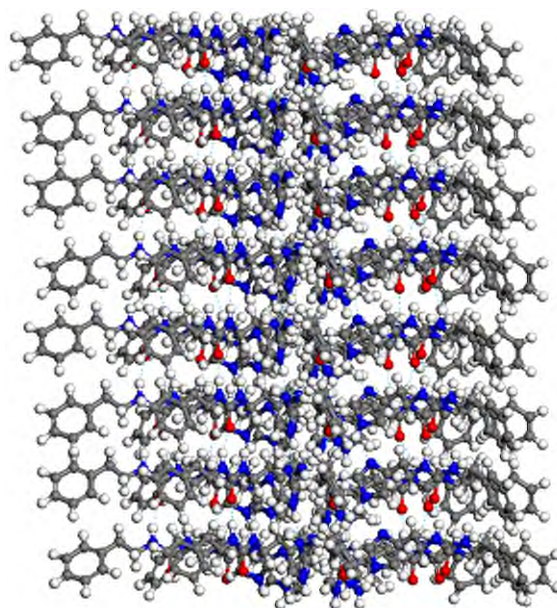


Supplementary Fig. 41. NaCl rejection of RO membranes modified with hydroxy artificial water channels (OH AWCs), I-quartet AWCs (HC6 AWCs), and imidazolylethyl-ureidoethyl-phenyl AWCs (IUP AWCs). Error bars represent the s.d. ($n = 3$) and data are presented as mean values $\pm$ s.d.

Supplementary Note: The NaCl rejection of RO membranes modified with OH AWCs showed no significant difference compared with those modified with HC6 AWCs and IUP AWCs (Supplementary Fig. 41).



Supplementary Fig. 42. Structure of I-quartet AWCs (HC6 AWCs).



Supplementary Fig. 43. Structure of imidazoleylethyl-ureidoethyl-phenyl AWCs (IUP AWCs).

Supplementary Note: The structures of HC6 AWCs and IUP AWCs used for MD simulations were shown in Supplementary Fig. 42 and Supplementary Fig. 43.

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