

Supplementary Materials for

Electrically Tunable Selectivity Enhancement Overcomes the

Permeability-Selectivity Trade-off in Two-dimensional

Nanofluidic Channels

Wanqi Zhou^a, Wanlin Guo^a, Hu Qiu^{a,*}

[a] Key Laboratory for Intelligent Nano Materials and Devices of Ministry of Education, State Key Laboratory of Mechanics and Control for Aerospace Structures, Nanjing University of Aeronautics and Astronautics, Nanjing 210016, China

*Corresponding author. Email: qiu@nuaa.edu.cn

This PDF file includes:

Figs. S1 to S18

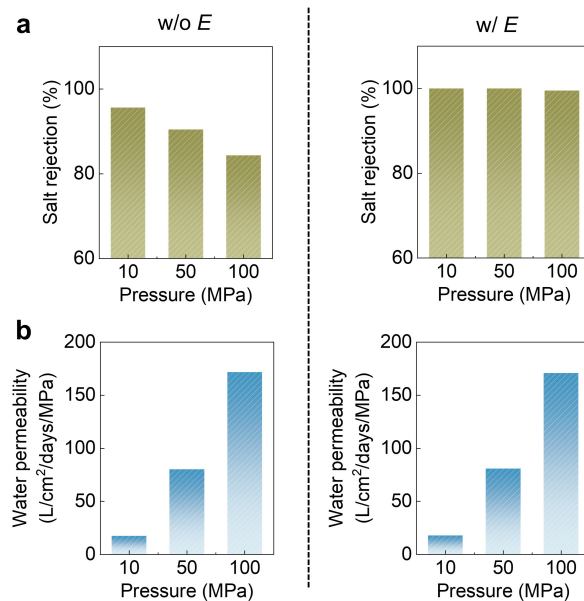


Figure S1: Influence of external pressure on salt rejection (a) and water permeability (b). The left and right panels present the results obtained in the absence and presence of a 1.0 V/nm external electric field, respectively.

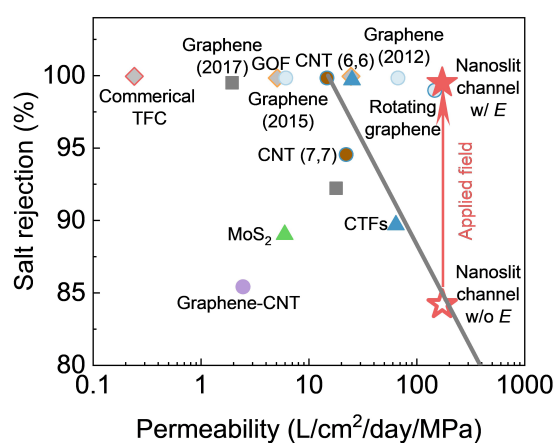


Figure S2: Comparison of our data with previous simulation and experimental results demonstrating the breakthrough of the permeability–selectivity trade-off. The previous data are summarized in *Science Advances* **6**(34), eaba9471 (2020). The gray line serves as a guide to the general trend of the permeability–selectivity trade-off. Red open and solid stars represent the nanoslit channel (9.2 nm height) under zero electric field and under a 1.0 V/nm electric field, respectively.

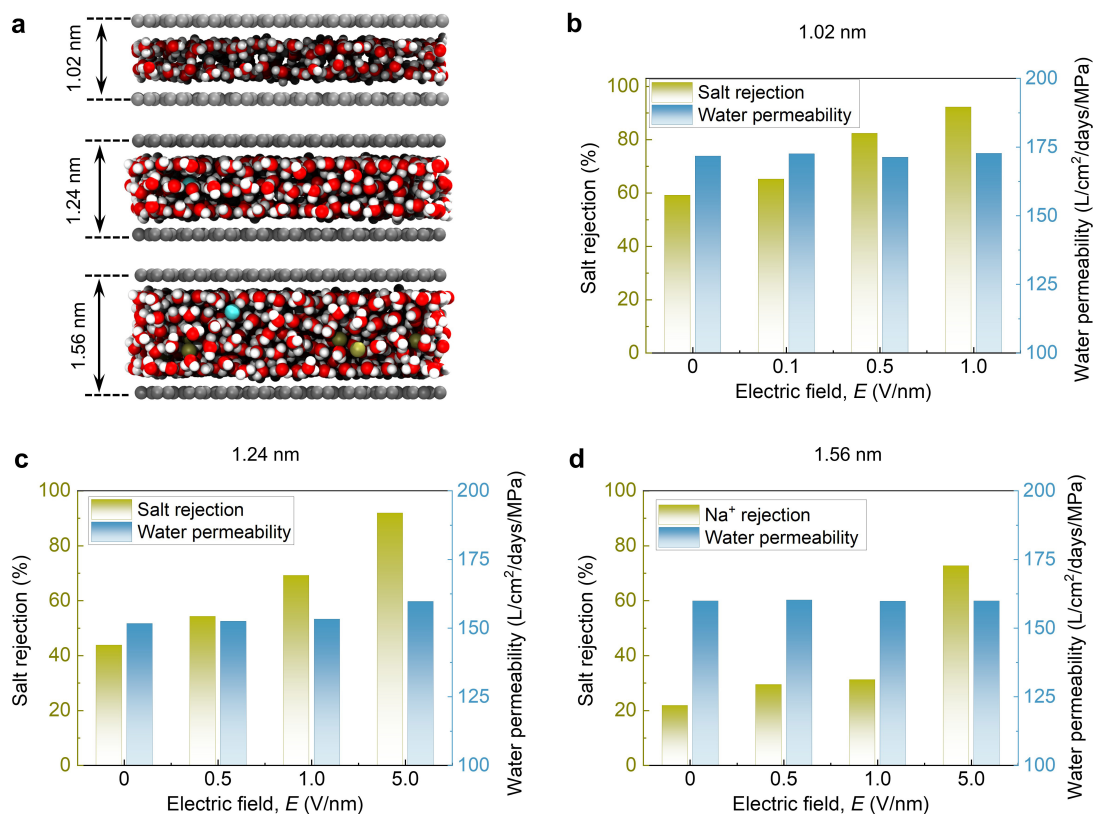


Figure S3: Influence of channel height on desalination performance. (a) Schematic diagrams of nanoslit channels with heights of 1.02, 1.24, and 1.56 nm. (b) to (d) Salt rejection (left axis) and water permeability (right axis) as a function of the applied electric field for channel heights of 1.02 (b), 1.24 (c), and 1.56 (d) nm.

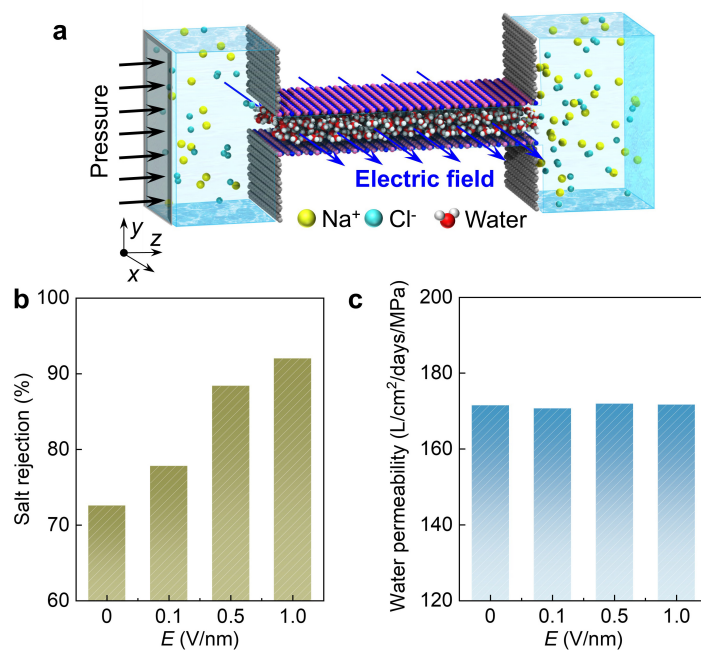


Figure S4: Same as Fig. 1 but for nanoslit channel made from hexagonal boron nitride. The Lennard-Jones parameters for boron and nitrogen atoms were adopted from *Nat. Commun.* **10**(1), 4610 (2019), with partial charges set to $+0.4e$ for boron and $-0.4e$ for nitrogen, respectively.

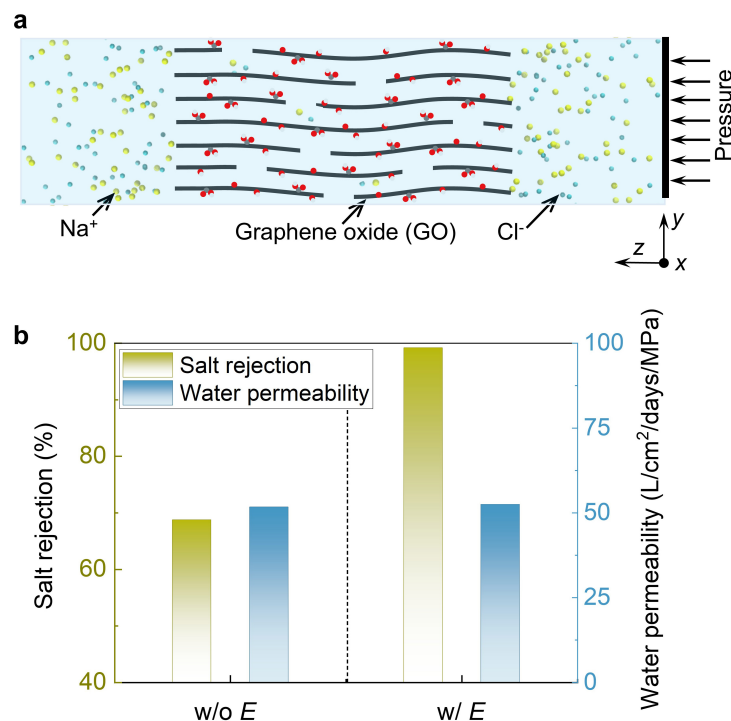


Figure S5: Electric-field-induced enhancement of salt rejection in a graphene oxide (GO) membrane. (a) Schematic illustration of the simulation system. The interlayer distance of channel within GO membrane is 10 Å. The GO flakes measure 6 nm in length and 3.45 nm in width. The system contains a 1 M NaCl solution, with an external pressure of 100 MPa and an electric field of 1.0 V/nm applied along the x -axis. (b) Comparison of salt rejection (left axis) and water permeability (right axis) in the absence and presence of the electric field. See Methods for simulation details.

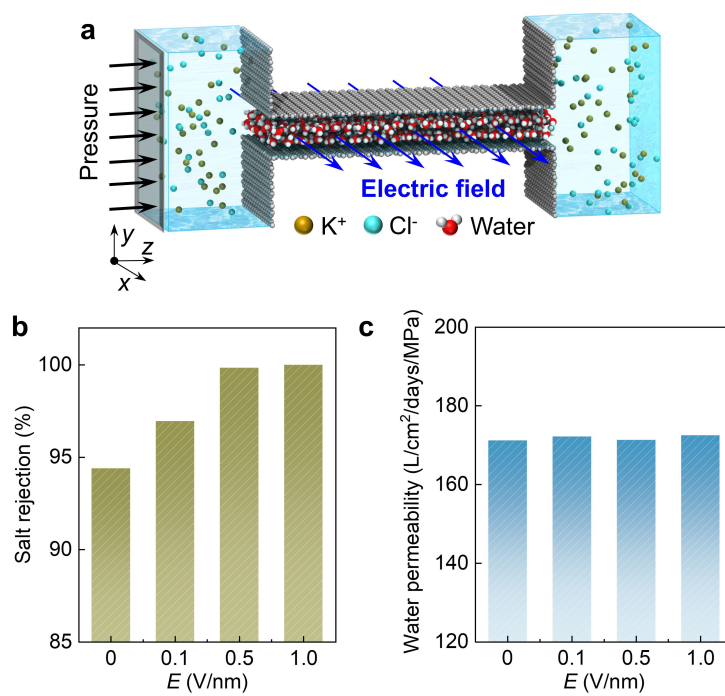


Figure S6: Same as Fig. 1 but for KCl solution.

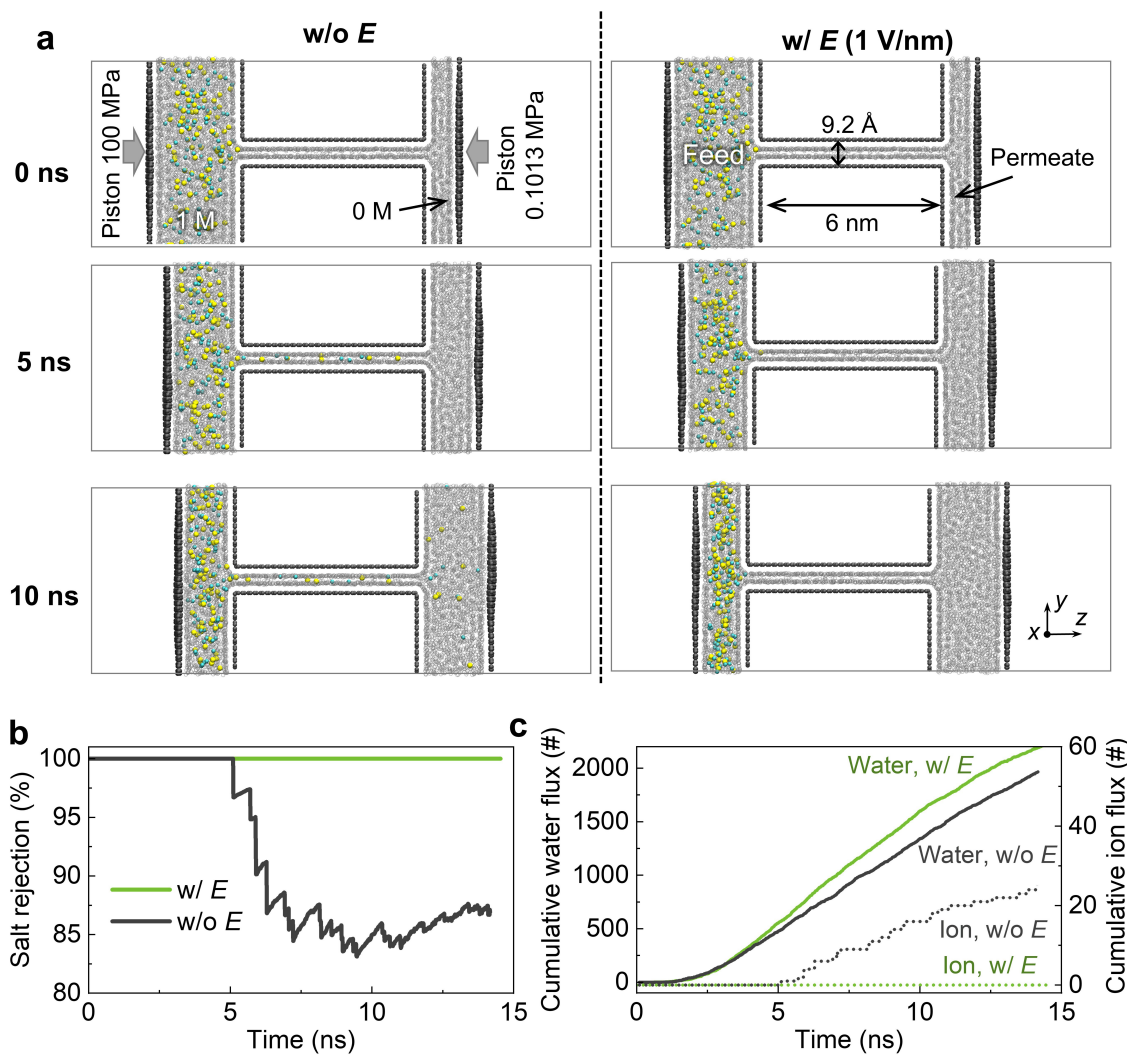


Figure S7: Verification of salt rejection enhancement using the model with two graphene pistons. (a) Snapshots of the simulation system comprising two graphene pistons that separate a saline feed reservoir from a permeate reservoir. The reservoirs are connected by a single nanoslit channel with an interlayer spacing of 9.2 Å. The feed reservoir contains a 1 M NaCl solution, while the permeate reservoir contains pure water (0 M NaCl). The dimensions of the simulation box in the x , y , and z directions are 6.02, 6.38, and 18 nm, respectively. The yellow, cyan, transparent, and gray balls represent Na^+ , Cl^- , water molecules, and C atom, respectively. (b) Time evolution of salt rejection under 0 and 1 V/nm electric field. (c) Cumulative water flux (left axis) and cumulative ion flux (right axis) through the nanoslit under 0 and 1 V/nm electric field.

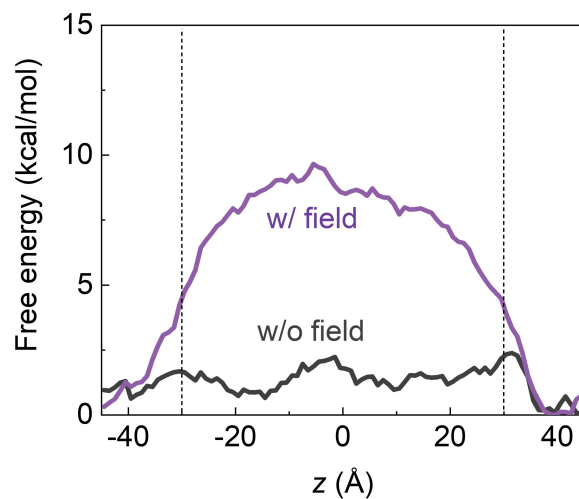


Figure S8: PMF profiles obtained in simulations using the polarizable model. The strength of the electric field is 1.0 V/nm. See Methods for simulation details.

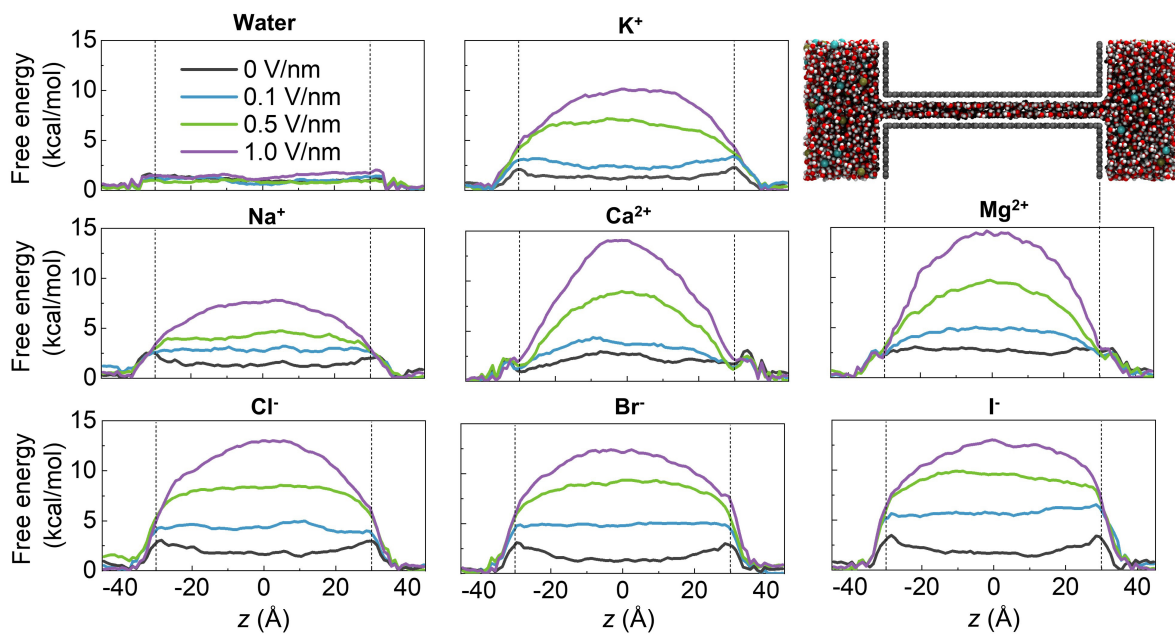


Figure S9: PMF profiles for water, Na^+ , Cl^- , K^+ , Ca^{2+} , Mg^{2+} , I^- and Br^- under different electric field strengths.

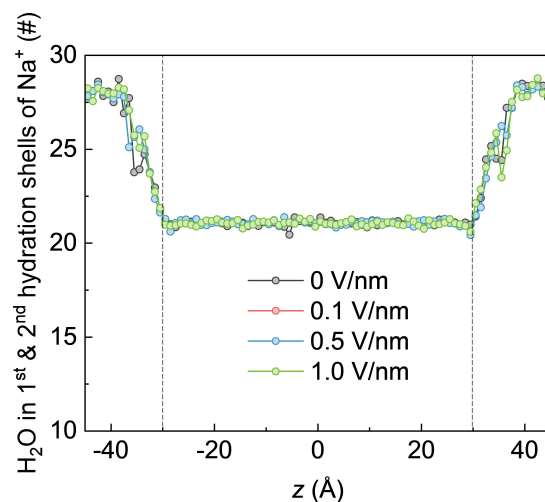


Figure S10: Average number of water molecules in the first and second hydration shells of Na^+ .

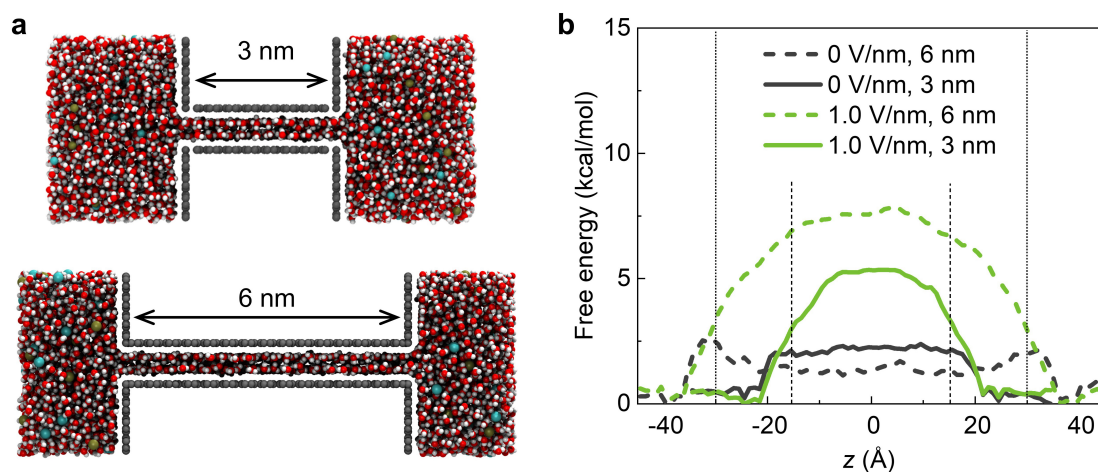


Figure S11: Comparison of free energy profiles (potential of mean force, PMF) for ion translocation through nanoslit channels of varying lengths. **(a)** Simulation systems used for free energy calculations, featuring nanoslit channels with lengths of 3 nm and 6 nm. **(b)** Free energy profiles along the channel axis (z axis) for Na^+ translocation through nanoslit channels with lengths of 3 nm (solid lines) and 6 nm (dashed lines). The green and black curves represent the cases with and without an applied electric field ($E = 1.0$ V/nm), respectively. The vertical black dashed and dotted lines indicate the two ends of the 3 nm and 6 nm nanoslit channels, respectively.

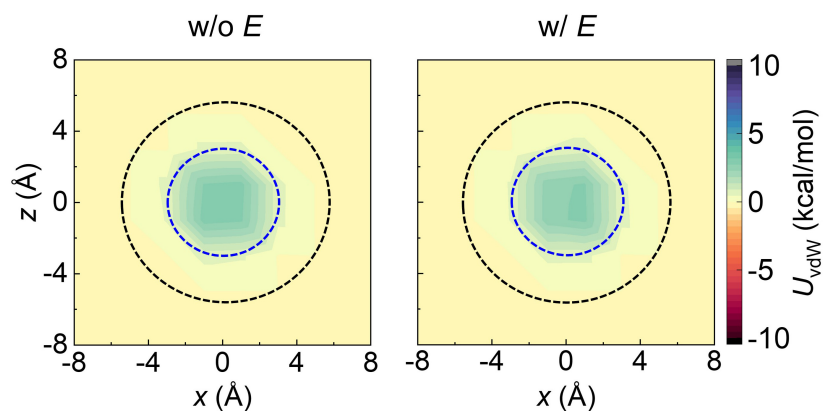


Figure S12: Distribution of the van der Waals (vdW) interaction energy (U_{vdW}) between Na^+ and water molecules around it. The strength of the electric field is 1.0 V/nm.

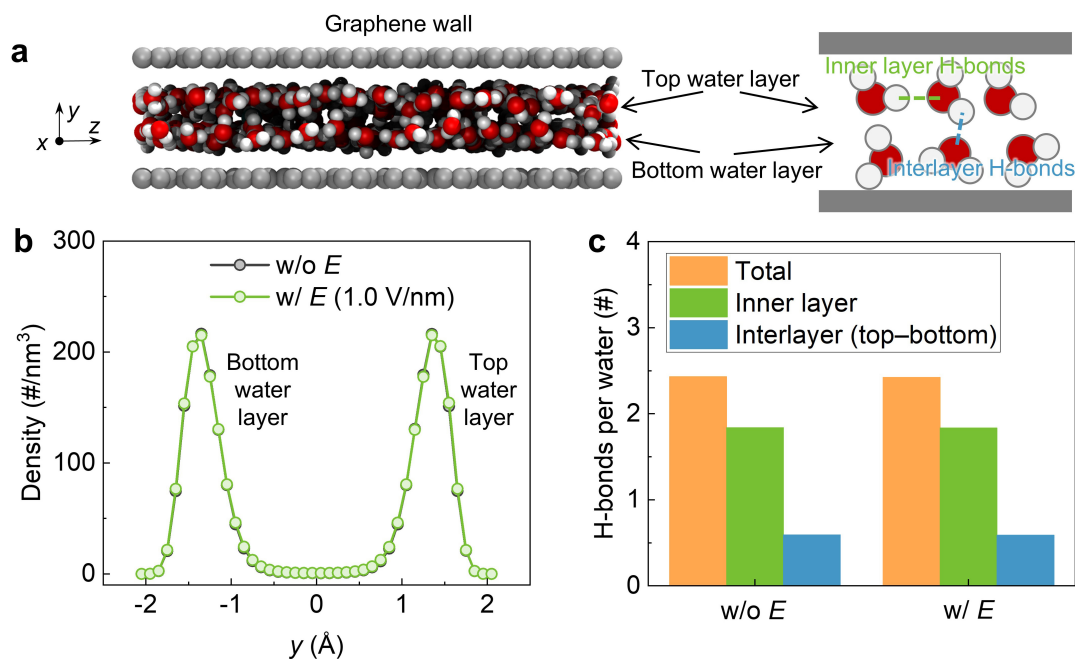


Figure S13: Comparison of water density and hydrogen-bond numbers inside the nanoslit channel with and without an applied electric field. (a) Schematic illustration of the bilayer water structure inside the nanoslit channel. (b) Water density profiles along the y axis in the presence and absence of the electric field. (c) Number of inner layer and interlayer hydrogen bonds (H-bonds) per water layer with and without the applied electric field ($E = 1.0$ V/nm).

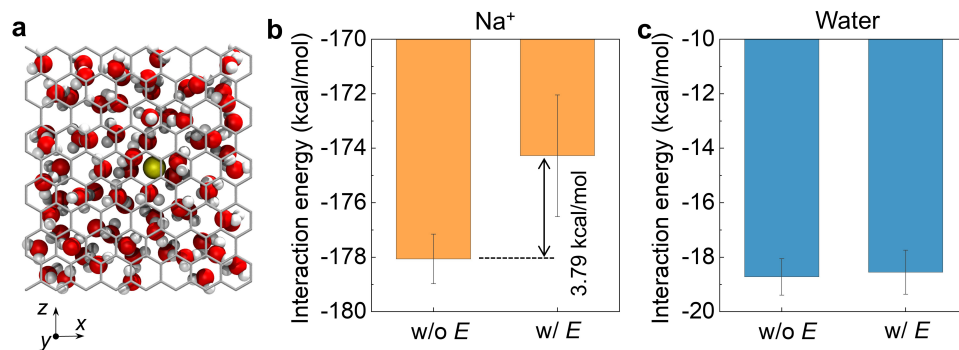


Figure S14: Field-induced change in the interaction between in density functional theory (DFT) calculations. (a) AIMD simulation system containing water molecules and a Na⁺ ion sandwiched between two graphene layers with an interlayer distance of 9.2 Å. (b) Interaction energy between the Na⁺ ion and remaining atoms in the system in the absence and presence of an electric field. (c) Interaction energy between a water molecule and remaining atoms in the system (without the presence of Na⁺ ion) in the absence and presence of an electric field. See Methods for simulation details.

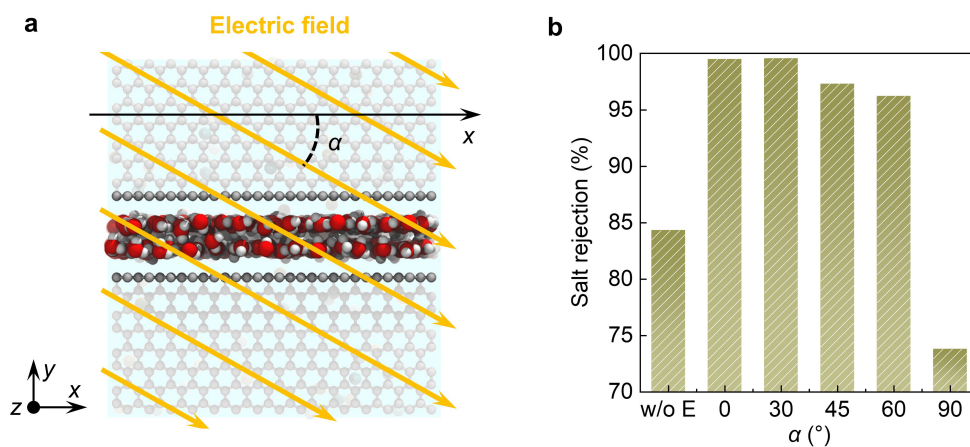


Figure S15: Effect of the in-plane electric field direction on salt rejection. (a) Definition of the field direction angle α . (b) Salt rejection versus α .

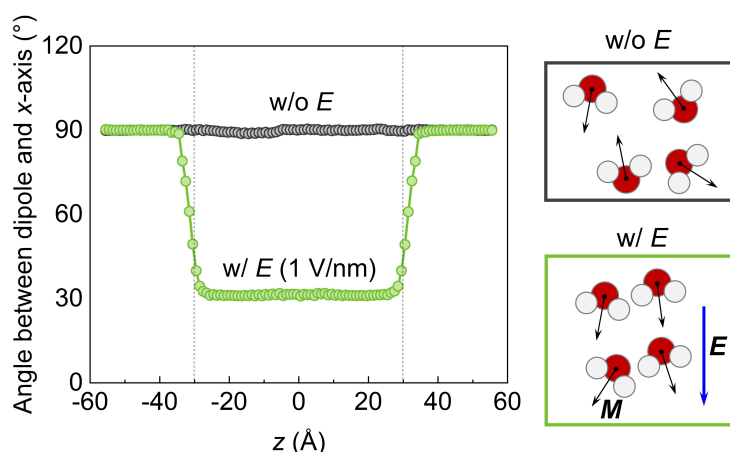


Figure S16: Orientation of water dipoles relative to the x -axis (the direction of the applied electric field) at different z -positions along the channel axis, comparing simulations without an electric field and with a 1 V/nm electric field. The vertical black dashed lines indicate the two ends of the nanoslit channels. The angle is defined as the angle between the dipole of a water molecule and the direction of the applied electric field (x -axis).

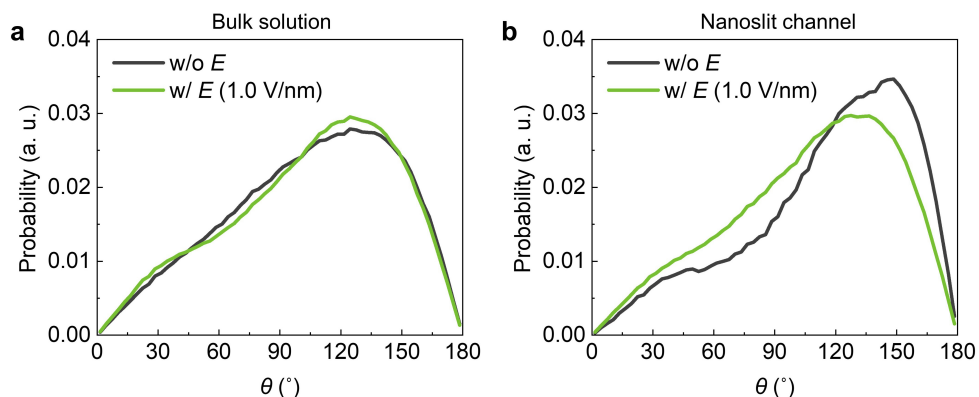


Figure S17: Comparison of the orientation angle (θ) in the bulk region and within the nanoslit channel. (a) Distribution of θ for water molecules in the first and second hydration shells of Na^+ in the bulk region, under 0 and 1 V/nm electric field. (b) Distribution of θ for water molecules in the first and second hydration shells of Na^+ within the nanoslit channel, under 0 and 1 V/nm electric field. The orientation angle θ is defined as the angle between the dipole of a water molecule and the line joining the Na^+ to water oxygen.

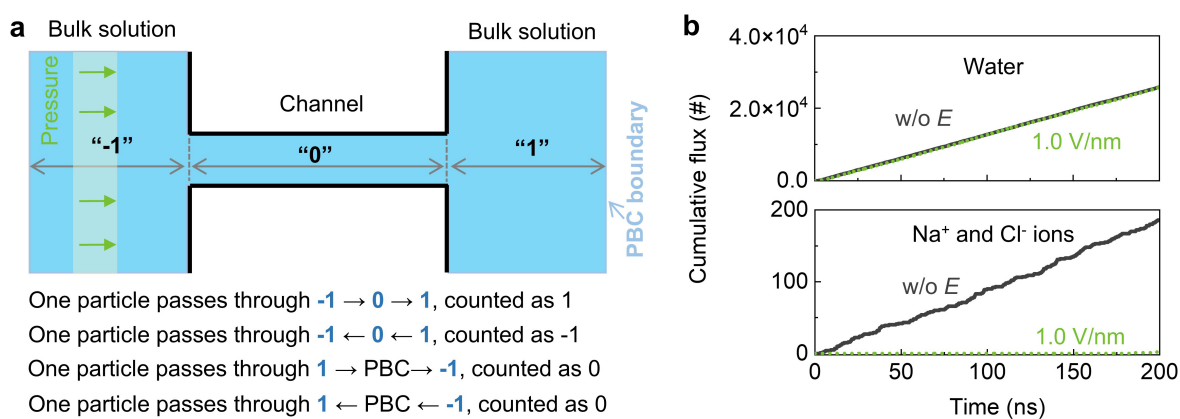


Figure S18: Statistics of the number of particles passing through the nanoslit channel. (a) Schematic illustration of the counting method for ions and water molecules translocating through the channel. **(b)** Cumulative flux of water molecules and ions across the nanoslit channel.