Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supporting Information for

Structure-dependent water transport across nanopores of carbon nanotubes: toward selective gating upon temperature regulation

Kuiwen Zhao and Huiying Wu *

School of Mechanical Engineering, Shanghai Jiao Tong University, Shanghai, 200240, China.

Corresponding Author: whysrj@sjtu.edu.cn

1. Methods

Molecular dynamics simulations were performed using the LAMMPS [1]. Water was modeled using the TIP4P/2005 rigid water model, one of the best models for describing condensed phases of water, both solid and liquid [2]. The TIP4P/2005 rigid water model gives an excellent prediction of the thermodynamic properties, including self-diffusion coefficient, density, and phase diagram, over the temperature range from 123 K to 573 K [2], which are very important for this work. The freezing point of TIP4P/2005 water model at 1 atm is 252.1 K, about 21 K below the experimental value. The Lennard-Jones parameters of carbon atoms are σ_{cc}= 3.55 Å and ε_{cc}= 0.07 kcal/mol, corresponding to the aromatic carbon in the CHARMM27 force field[3]. The van der Waals parameters of different species of atoms were obtained by the Lorentz–Berthelot

combining rules. A 12-Å cutoff distance for the pair interactions was used. The particle-particle particle-mesh (PPPM) algorithm was used to calculate the long-range electrostatic interaction. Harmonic restraining force (1.0 kcal/mol/Å^2) was applied to the carbon atoms of the nanotubes to prevent their displacements in the simulations**.** Periodic boundary conditions were imposed in all directions. The equations of motion were integrated in time using the leapfrog scheme with a time step of 2 fs. Structures were saved every 1 ps. Simulation systems were maintained constant at 1 atm using Nosé−Hoover barostat.

2. Comparison with continuum predictions

To compare our results with continuum predictions, we conducted two additional simulations of pressure-driven water flow across nanopores with d = 0.798 nm at 303.15 and 373.15 K following the reference [4]. To avoid the damage of water structures in nanopores induced by a high pressure difference, a small pressure difference of 5 MPa (still very large for practical applications) is imposed across the nanoporous membrane. We obtain a net water flux of 6.54 ns^{-1} at 373.15 K (Figure S1a). However, no obvious net water flux is observed as the temperature decreases to 303.15 K (Figure S1b).

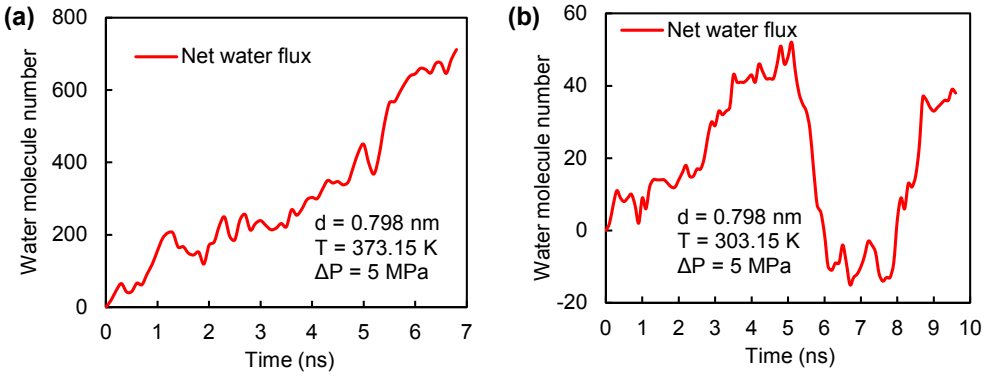

Figure S1. Net water flow across nanoporous membrane of carbon nanotubes under a pressure difference of 5 MPa at 373.15 K (a) and 303.15 K (b). The membrane consists of 16 nanopores

with d = 0.798 nm.

According to no-slip Hagen–Poiseuille equation in continuum hydrodynamics theory, the water flux across a cylindrical shaped channel under a pressure difference can be determined from [4-6]

$$Q = \frac{\pi d^4}{128\mu}\frac{\Delta P}{L} \quad (1)$$

where d is the inner diameter of channel, ΔP is the pressure difference, μ is the dynamic viscosity of water, and L is the length of channel. A water flux of 3.9×10^{-20} m^3/s, corresponding to 1.3 ns^{-1}, can be predicted by Equation 1 for d = 0.798 nm and L = 1.6 nm at 303.15 K. The net water flux for Figure S1b is definitely smaller than it. For T = 373.15 K, the net water flux obtained from MD simulations (Figure S1a) is 1.8 times of the prediction of no-slip Hagen–Poiseuille equation, which is consistent with previous literature [6].

References

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