Thermal-driven flow inside graphene channels for water desalination

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Abstract

A novel concept of membrane process in a thermal-driven system is proposed for water desalination. By means of molecular dynamics simulations, we show fast water transport through graphene galleries at a temperature gradient. Water molecules are driven to migrate through nanometer-wide graphene channels from cold reservoir to hot reservoir by the effect of thermal creep flow. Reducing the interlayer spacing to 6.5 Å, an abrupt escalation occurs in water permeation between angstrom-distance graphene slabs. A change from disordered bulklike water to quasi-square structure has been found under this extremely confined condition. This leads to a transition to subcontinuum transport. Water molecules perform collective diffusion behaviors inside graphene channels. The special transport processes with structure change convert thermal energy into motion without dissipation, resulting in unexpectedly high water permeability. The thermal-driven system reaches maximum flowrate at temperature variance of 80 K, corresponding to the quantity at pressure difference up to $10^5$ bar in commercial reverse osmosis processes and 230 bar in pressure-driven slip flow. Our results also reveal the movement of saline ions influenced by thermophoretic effect, which complements the geometry limitation at greater layer spacing, enhancing the blockage of ions. This finding aims to provide an innovational idea of developing a high-efficiency desalination technology able to utilize various forms of energy.

1. Introduction

The shortage of water resources has been severe in recent years due to ever-increasing global population, rapid industrialization and expanding urbanization [1–4]. Desalination of saline water has now been accepted as a promising way to gain fresh water [5–8]. Well-established desalination technologies can be classified as membrane processes (without phase change) and thermal processes (with phase change). Membrane processes utilize a physical barrier (membrane) to separate the dissolved ions from the feed water, with low consumption of energy and high efficiency of water product, but membrane-based technologies such as reverse osmosis (RO) processes usually demand high-grade energy (electric energy) [6–9]. Thermal processes vaporize fresh water from saline water, so various forms of energy, such as fossil fuel sources, solar energy, or waste heat, can be directly used in thermal desalination technologies [6, 10–12]. However, the great latent heat of vaporization makes the thermal processes energy-intensive [1, 6]. Those limitations of membrane processes and thermal processes have stalled the development of desalination technologies.

Nowadays, the blooming of advanced materials in nanoscale applied in desalination brings many chances to get out of the dilemma [13–20]. Water interlaminated between two-dimensional (2D) materials, such as graphene oxide (GO) membranes, is proved to show unique flow behaviors and unconventional mechanisms. Unimpeded permeation of water through GO membranes has been experimentally investigated by many researchers, and it is found that the permeation rates of water are several orders of magnitude greater than the classic viscous flow prediction [21–24]. This unexpected fast flow was attributed to the slippery nature of water flow in confined channels with great
Knudsen numbers, and the pristine graphene regions in GO sheets offer a super-lubrication boundary to enhance the slip effect [25–33]. Recent experimental and theoretical studies have demonstrated the existence of ordered water structures within nanofluidic spaces [34–36]. This solid-like ordered water performs collective diffusion, which causes fast water transport through GO membranes at high humidity [37]. Though the nano-scaled materials to date have improved the membrane performance to some extent, it still depends on a pressure-driven system for operating. Is there some way to combine the high-efficiency of membrane processes with the wide-use of energy of thermal processes?

Here we propose a membrane process for water desalination in a thermal-driven system, through performing molecular dynamics (MD) simulations. It may help to develop an innovative method for high-efficiency desalination, which is able to utilize various forms of energy. The molecular structure is shown in figure 1. Two saline water reservoirs are separated by a graphene channel, and a heat current is applied to the whole system, maintaining the two reservoirs at different temperature. We have found that water molecules are driven to transport from the cold side to hot side by the effect of thermal creep flow, while saline ions are driven to move at reverse direction owing to the thermophoretic force. These two effects help water separate out of a pressure-based system. What’s more, a subcontinuum transport has been revealed in extremely confined water molecules, exhibiting collective diffusion behaviors and convert thermal energy into directed motion with high efficiency. It shows an extraordinary ability of water permeation, corresponding to that of commercial RO membrane at a pressure difference of 10^5 bar and 230 bar in pressure-driven slip flow.

2. Model and computational details

2.1. MD simulations

The molecular model is shown in figure 1. Two saline water reservoirs, at the opposite of ends of the simulation box, are divided by a graphene channel with a length of 10 nm. Period boundary conditions are utilized in the in-plane x and y directions. In the z direction, both sides of system are bounded by rigid pistons to apply pressure with same value and opposite direction. It aims to guarantee the water molecules’ entrance into the channel between graphene lamellas. Each saline water reservoir contains 4000 water molecules and 40 pair of Na^+ and Cl^- (with the concentration of 0.56 M). The two graphene laminates in channel have the width of 3 nm in y directions, and we consider five sets of interlayer spacing between two laminates, corresponding to 6.5 Å, 9 Å, 12 Å, 17 Å, and 22 Å. Simulations are firstly performed in the NVT ensemble lasting 500 ps for relaxation. After that, the system is switched to the NVE ensemble, and two specific regions at the opposite ends are thermostatic to perform a temperature gradient through the whole system. To avoid the spurious effect on water molecules entering into the channel, the thermostatic regions are set to begin at the position 4 nm away from the channel entrance, till the edge of the simulated box. The temperature differences between hot and cold reservoirs range from 30 K to 80 K at interval of 10 K, within the limits of 280 K to 350 K. Langevin thermostat is utilized to control the temperature of hot and cold water reservoirs in our simulations. The time step to integrate the equation of motion is 0.5 fs. Simulations of thermal-driven flow are implemented after equilibration at average temperature of cold water and hot water using the Nosé–Hoover thermostat with a damping time constant of 50 fs. The number of water molecules and ions from two reservoirs are recorded every 5 ps to get converged results, and last for a sufficiently long time (20 ns) for data collection. The rigid simple point charge effective pair (SPC/E) model is used to describe the potential of water molecules. This model has been widely used in previous works, and it successfully predict structures and thermodynamic properties of confined water [38]. We have also discussed the properties of interlayer molecules by using different models, and obtained consistent results from all models. The interactions between water
molecules and carbon atoms in graphene is considered as parameters $\sigma_{\text{C-C}} = 0.319 \text{nm}$ and $\varepsilon_{\text{C-C}} = 4.063 \text{meV}$, which predicts a water contact angle for graphene corresponding to the value measured experimentally. The van der Waals interactions are truncated at 1.2 nm, and the long-range Coulomb interactions are computed by utilizing the particle–particle–particle-mesh (PPPM) algorithm. All potential parameters are given in table 1. The characteristic length $\sigma$ and energy parameter $\varepsilon$ between different atoms are employed by the common Lorentz–Berthelot combination rule. All MD simulations are employed using the large-scale atomic/molecular massively parallel simulator (LAMMPS) package [39]. The post-processing is made by visual molecular dynamics (VMD) [40] and The Open Visualization Tool (OVITO) [41].

### Table 1. Potential parameters of atoms.

| Atom  | $\sigma$ (Å) | $\varepsilon$ (Kcal/mole) | Charge ($q$) |
|-------|--------------|---------------------------|--------------|
| C(–C) | 3.550        | 0.070                     | 0            |
| O(H$_2$O) | 3.166        | 0.155                     | −0.8476      |
| H(H$_2$O) | 0.000        | 0.000                     | 0.4238       |
| Na$^+$ | 2.586        | 0.105                     | 1            |
| Cl$^-$ | 4.402        | 0.105                     | −1           |

2.2. Calculation of the diffusive coefficients

To elucidate the diffuse behaviors of water confined to inner graphene channels, we construct a sandwiched structure with water molecules intercalated between two graphene sheets. The size of graphene sheet is $10 \text{nm} \times 3 \text{nm}$, equal to the simulations of thermal-driven flow. Specified quantity of water molecules are placed between graphene laminations according to the molecular number inner thermal-driven channel, with $N_w$ from 329 to 1763. The variation of water molecules between graphene slabs with different $d$-spacing have been shown in figure S12, and the average number of molecules in converged region is chosen as the specified value $N_w$. The molecular structures are equilibrated at NVT ensemble for 2 ns, and switched to NVE ensemble for data collection. The molecular diffusive coefficient $D$ can be obtained from the mean square displacement (MSD). It can be calculated from the long-time limit of MSD by $D_{\text{MSD}} = \lim_{t \to \infty} \langle (\vec{r}(t) - \vec{r}(0))^2 \rangle / 2dt$, where $|\vec{r}(t) - \vec{r}(0)|$ is the displacement of center of mass from its initial position, $d_t$ is the dimension of space, $t$ is the simulation time, and $\langle \cdots \rangle$ is the ensemble average. To investigate the flow in the inner graphene valley in detail, we have decomposed the diffusion coefficient into components along $x$ and $y$ directions by the directional displacement.

3. Results and discussion

3.1. Thermal-driven creep flow

The temperature field plays a significant role in inducing flow, especially for water flow in nano-scaled geometries. It has been reported that water can be thermally pumped across carbon nanotube (CNT) membranes [42–44], because of the thermal Brownian fluctuations of surface atoms [42] or the thermo-osmotic flow of water molecules [45–47]. The previous literature considers water flow as a continuous process and ignores the kinestate inside the channel. However, in nano-confined systems where the motion scope of the water molecules is comparable to the scale of the flow domain, the applicability of the continuum based model is uncertain. Thus, the kinetics of the molecules near the boundary should be taken into consideration on the fluid flow. Combined with kinetic theory and classic hydrodynamics, a net flow can be driven along a temperature gradient on a diffusely reflecting wall, which is known as thermal creep flow [48–52]. For the flow confined between two flat plates with a temperature gradient $\partial T / \partial x$, the induced velocity $u_s$ can be estimated as [53, 54],

$$u_s = \frac{3 + j}{4} \left( \frac{\eta}{\rho T} \frac{\partial T}{\partial x} \right).$$

(1)

Here, $j$, $\eta$, $\rho$, and $T_i$ is the degree of internal freedom, viscosity, density, and temperature of fluid molecules, respectively. For a water molecule, a rigid triatomic molecule without non-linear arrangement, the degree of internal freedom $j$ should be 3 here. Because the spatial distribution of density in the direction perpendicular to the graphene sheets is inhomogeneous (figure S14), the average density can be used in the formula. We consider the averaged velocity $u_m$ along the whole channel to assess the comprehensive thermal creep flow, which is defined as:

$$u_m = \frac{1}{L} \int_0^L \frac{3 + j}{4} \left( \frac{\eta}{\rho (T_0 + \frac{\partial T}{\partial x})} \frac{\partial T}{\partial x} \right) dx,$$

$$dx = \frac{3 + j}{4} \cdot \frac{\eta}{\rho L} \cdot \ln \left( 1 + \frac{L \eta \partial T}{T_0 \partial x} \right).$$

(2)

Here, $L$ is the length of the channel and $T_0$ is the temperature of the channel entrance. The explanations of thermal creep flow in detail have been expounded in supplementary information section 1 (figure S1 (stacks.iop.org/2D Mat 6/035018/mmmedia)). To elucidate the thermal-driven creep flow inner graphene channel, a passage with $d$-spacing of 12 Å is investigated through the thermostatic system, with cold reservoir at 293 K and hot reservoir at 323 K. Figure 2(a) portrays the temperature distribution of water on $x$–$z$ plane and along $z$ direction. The temperature is linearly increased from cold reservoir to hot reservoir through the graphene channel, promising a stable temperature gradient during the simulation. It is noted that the geometrical spacing between two graphene sheets is greater than the actual flowing width of the channel, because of the repulsive interaction between water molecules and carbon atoms in graphene. The actual width of the channel is measured as 8 Å according to the temperature contour figure. The flowrate of water
through the graphene channel is calculated through the effective flow area, equal to the product between actual width of channel and the slabs length in \( y \) direction. The whole simulation box is divided into three parts by the graphene channel, and the temporal evolution of number of water molecules in each portion has been tracked in figure 2(b). It is clearly that the water molecules move from cold reservoir to hot reservoir through the graphene channel. The flow direction is identical to the way of positive temperature gradient, consistent with the prediction of thermal creep flow in equation (1). The flow profiles show that the flow rate of water is stable in time, corresponding to 1916 L/cm²/day. Surprisingly, the water flowrate driven from a temperature difference of 30 K is equivalent to that of the existing commercial RO membranes from a pressure difference of almost 10⁴ bar.

To give a further insight into the flow characteristics, we investigate the thermal creep flowrate at different temperature gradient by setting series of temperature difference from 40 K to 80 K (figures S2–S9). The flow rates as a function of temperature difference are plotted in figure 3. For graphene channels with \( d \)-spacings beyond 9 Å, the flowrate increases with the increase of temperature difference, but the growth rate of flowrate shows decay at large temperature difference. It accords with the tendency of natural logarithm as portrayed in the solid curves (figure 3), consistent with the prediction in equation (2). The flowrate also varies a lot among different graphene channels. That with smaller \( d \)-spacing exhibits superior capability for thermal-driven flow. It is noted that the graphene channel with \( d \)-spacing of 6.5 Å shows distinctive flow characteristics compared to others. The flowrate through the narrowest graphene channel greatly exceeds that beyond 6.5 Å and it increases linearly without decay even at larger temperature difference. All the above proves that the thermal creep flow is strongly related to the spacing inner graphene channels, and it is important to figure out the size effects on water flowrate.

![Figure 2. Thermal-driven creep flow in graphene channel with \( d \)-spacing of 12 Å. (a) Temperature distribution of water on \( x-z \) plane (left panel) and along \( z \) direction (right panel). (b) Temporal evolution of water molecules.](image)

![Figure 3. Dependence of water flowrate through graphene channel with variable \( d \)-spacing of temperature difference. Black, red, blue, and green spheres represent the value of \( d \)-spacing corresponding to 6.5 Å, 9 Å, 12 Å, and 17 Å. Lines are guides to the trend of flowrate evolution. Dashed straight line denotes the linear trend, and solid curve lines denote the trend of natural logarithm with the form of \( y = A\ln(1 + Bx) \).](image)
3.2. Size effects: transition to subcontinuum transport

Variable kinds of graphene channels with \(d\)-spacing from 6.5 Å to 22 Å at \(\Delta T = 30\) K have been performed to investigate the size effects on water thermal creep flow (figures S10–12). In graphene channels with \(d\)-spacing larger than 9 Å, we found that the flowrate monotonically decreases with increasing layer spacing (figure 4). This trend is like the decreasing flow enhancement of pressure-driven flow in CNTs with growing diameter, due to the impair size effect for large \(d\)-spacing. It is interesting that a growth spurt occurs when the layer spacing reduces from 9 Å to 6.5 Å, which suggests a transition to subcontinuum transport (figure 4). By reducing interlayer spacing, the water structure changes from bulk to high order as shown in the snapshots inserted in figure 4. We have calculated the out-of-plane permittivity [55] of confined water between graphene sheets with different layer spacing at temperature of 300 K (figure S18). The permittivity of confined water is smaller than that of bulk water because the rotational freedom of water dipoles is expected to decrease at nanoconfinement. With the decrease of \(d\)-spacing, the permittivity of confined water is reducing and approaches the minimum value of 4.48 at the \(d\)-spacing of 6.5 Å, which is close to the permittivity of ice as reported in previous works [56]. It demonstrates that water at extreme confinement will form ordered structure like ice. To clearly exhibit the difference of water structures and flow characteristics at variable \(d\)-spacing, we implement simulations of a sandwiched structure with water molecules intercalated between two graphene sheets (figure S13). The spatial density distribution of confined water perpendicular to the graphene layer has been depicted in figure S14. It is clear that the number of water layers is getting less as the layer spacing decreases, and it reduced to only one layer at \(d\)-spacing of 6.5 Å.

We provide the pair correlation functions atoms in water molecules inner graphene gallery with different spacing (figure S15). The first peak of \(g(O-O)\) and \(g(O-H)\) curves vanish with the increase of \(d\)-spacing, which gradually changes toward the shape of bulk water. This transformation indicates that the scale effect on water inner graphene gallery is progressively weakened with \(d\)-spacing increasing from 9 Å to 22 Å. To quantify the variation in nanoconfined flow with layer spacing, we perform the calculation of total energy interaction between water molecules and graphene laminations. The average energy per molecule reduces with expanding layer space as shown in figure S16. It arises from the fact that the intercalated water molecules increase fast while the contact area is limited for channel with a certain length. Since the thermal-driven flow is due to the interaction force of the wall applying to water molecules, the reduction of interaction energy will inevitably cause the drop of flowrate.

When the layer distance dwindles to an extent which only can hold mono-layer water molecules, the structure of water shows significant difference compared to others. The first peak of \(g(O-O)\) curves in \(d\)-spacing of 6.5 Å is much higher than that beyond, and another peak occurs after the first peak. As portrayed in the snapshots of figure 5(c), in the extreme confined space (i.e. inner graphene gallery with the layer distance of 6.5 Å), water molecules are highly ordered and form the structure of quasi-square lattice. These ice-like molecules perform collective behaviors to transport through graphene channel in high efficiency. We investigate the in-plane displacement of water molecules under two graphene slabs within 30 ps as shown in figure 5(b). The displacement is decomposed in \(x\) and \(y\) directions, and the distribution of molecular diffusion distance have been calculated for 6.5-angstrom-wide and 22-angstrom-wide channel.
at 300 K (figure 5(b)). It is observed that the diffusion is isotropic in larger channel with the \(d\)-spacing of 22 Å, whereas it is anisotropic with the \(d\)-spacing of 6.5 Å. It demonstrates that the diffusion of single-layer molecules with ordered structure exhibits unusual characteristics in contrast to bulk water. The collective diffusion coefficients of intercalated water have been investigated from the trajectories of center of mass of molecules (figure 5(a)). Below the temperature of 310 K, the coefficient \(D\) in \(x\) direction exceeds that in \(y\) direction, which proves that the collective diffusion is able to enhance water transport through graphene channel. However, this regular structure no longer exists as increase of \(T\), which can be presented in the peak decay of correlation curves (figure S15). What’s more, the collective diffusion behaviors do not predominate in \(x\) direction beyond 310 K (figure 5(a)). The distribution of diffusion distance presents the trend of anisotropy–isotropy transition in greater temperature (figure 5(b)). Besides, the out-of-plane permittivity of water also increases when the temperature rises from 300K to 340K (figure S19). These results indicate that the ordered quasi-square structure is sensitive to temperature, and it gets collection at low temperature while collapse at high temperature.

In the thermal-driven system with \(\Delta T = 30\) K, water molecules are in different temperature at the entrance (293.15 K) and the outlet (323.15 K) of graphene channel. It makes a transition of water structures from ice-like ordered to free along the direction toward positive temperature gradient. Water molecules entering the graphene gallery form an ordered structure and release heat to cold reservoir. Then ice-like water performs a collective transport through the graphene channel to the hot water reservoir. When arriving at the outlet, ordered water molecules gradually transform to free and absorb heat from hot reservoir. The diagram of molecular mechanism is portrayed in figure 5(d). The collective transport of water molecules inner gallery with low energy dissipation and the directly transformation from heat energy into molecular motion, provide more directional and higher efficient transport compared to the diffusion in bulk water. Thus, the graphene channel with \(d\)-spacing of 6.5 Å can achieve such high flowrate.

3.3. Thermophoretic effect on ions
In pressure-driven system, ions filtration is usually dependent on the geometry designation of pores or channels on membranes, which should be smaller than the size of hydrated ions. The hydration radii of some monovalent salt ions (like Na\(^+\) and K\(^+\)) are always at sub-nanometer, so most 2D materials have to downsize the \(d\)-spacing between laminations at the value not exceeding 0.8 nm to obtain an acceptable salt rejection. Saline water can be considered as the fluid mixture of several large particles (hydrated ions) around many tiny particles (water molecules). When the system is
exerted in a temperature gradient, the large particles receive imbalanced collisions from tiny particles in hot sides and cold sides with different momentum. The comprehensive result is a thermophoretic force applied to large particles (saline ions), which get a drift along the negative temperature gradient. It is reverse to the direction of water flow. Thus, there is an additional thermophoretic effect to help impeded ions besides geometry filtration in thermal-driven system. The mass flux for thermophoresis can be written as, $J = -D \nabla c - cD_T \nabla T$. Here $D$ is the usual Brownian diffusion coefficient, and $D_T$ is generally called the thermal diffusion coefficient in simple liquid mixtures [57, 58]. If $\nabla T$ is applied to a solution confined between two parallel plates, the system reaches a steady-state concentration gradient as, $\nabla c = -cS_T \nabla T$, where $S_T = D_T/D$ is called the Soret coefficient. It reaches a relative concentration difference $\Delta c/c = -S_T \Delta T$ at steady state. From previous study of diffusion coefficient [59] and thermophoretic mobility [60], we can predict that the relative concentration difference could be 3%–80% in our system. To explicitly indicate the dynamic motion of ions affected only by thermophoretic force, we construct the saline water reservoir at the same

Figure 6. Temporal evolution of ions concentration in the case of graphene channel with $d$-spacing of 12 Å. Left panel portrays the ions concentration of Na$^+$, and right panel portrays that of Cl$^-$.  

Figure 7. Snapshots of representative ions transport through graphene channel with $d$-spacing of 12 Å. Green particles denote Na$^+$, and purple particles denote Cl$^-$. Left side represents hot reservoir colored by red, and right side represents cold reservoir colored by blue. The black arrows are guide to eye for the same ions.
ionic concentration. For graphene channel with layer distance of 6.5 Å, smaller than size of hydrated ions, it presents the ability of blocking saline ions absolutely (figure S17), owing to the size filtration of geometry. As for larger $d$-spacing of 12 Å, the ionic concentrations of $\text{Na}^+$ and $\text{Cl}^-$ declines in hot reservoir (permeation side), and in contrast, those increase in cold reservoir (feeding side) during the whole process of thermal-driven flow (figure 6). It proves that water in hot reservoir can be purified even on the weak effect of geometry. To explore the motions of ions inner graphene channel, we track the trajectories of some representative ions for 10 ns. It can be seen that ions hesitate in the gallery and finally migrate from hot reservoir to cold reservoir (figure 7). It is because that two external forces, viscous stress by relative shift of water flow and thermophoretic force by temperature gradient, are both exerted to saline ions. At temperature difference corresponding to 30 K, the major factor is the thermophoretic effect, resulting in the ionic motion from hot side to cold side.

3.4. Comparisons with pressure-driven flow

Finally, we make a comparison of water flowrate between thermal-driven system and pressure-driven system. To complement the pressure-driven flow, we perform the simulations of water flow through the same molecular structures with $d$-spacing of 6.5 Å at 192 bar. Fast slip flow has been found for water inside the interlayer gallery between graphene slabs, which has also been investigated in previous works. We linearly extrapolate the flowrate at different pressure differences as plotted in the purple straight line (figure 8), through Darcy’s law, $\bar{v} = \gamma (\Delta P)$. Here, $\Delta P$, $L$, and $\gamma$ is the pressure difference across the channel, the channel length, hydraulic conductivity, respectively. The flowrate of graphene galleries with $d$-spacing of 17 Å can attain the extent of slip flow at $\Delta P = 10–30$ bar, and with $d$-spacing of 9 Å to 12 Å it reaches the extent of slip flow at $\Delta P = 20–70$ bar. The maximum flowrate is obtained through the graphene channel with layer distance of 6.5 Å. A surprising flowrate at pressure difference up to 230 bar in slip flow is equal to that in thermal-driven system at a temperature difference of 80 K.

According to MD results, graphene channels under temperature gradient exhibit remarkable permeability of water molecules, with a flowrate of 732–18 402 l/cm$^2$/day. This quantity corresponds to the infiltration capacity of existing commercial membranes at the pressure difference of $10^4$ to $10^5$ bar, as shown in the blue arrows of figure 8. Such an incredible flowrate is due to the huge temperature gradient and ultra-thin film thickness (10 nm). Practically, the thickness of the film is not such thin, so it is not enough to reach such a large temperature gradient. According to equation (2) in the manuscript, the flow velocity is approximately linear to the temperature gradient $\partial T/\partial x$. If it reaches the thickness of commercial RO membranes (about 1 μm), the water flow of this system is still comparable to the flow rate at the working pressure of the existing RO systems (~10 MPa). However, in this system the heat can be directly converted into the driving force of water flow, which is able to utilize various forms of energy. It can get rid of the shortcomings of traditional RO systems that rely heavily on electrical energy.

4. Conclusions

In summary, we propose a novel membrane process for water desalination in thermal-driven system, through nanoscale graphene channels. Our MD
various forms of energy. We have done is to provide a new idea of developing a process and 230 bar in pressure-driven slip flow. All Haifeng Jiang ORCID iDs Center of Wuhan University.

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References

[1] Gude V G 2015 Water Res. 89 87
[2] Gude V G 2015 Renew. Sustain. Energ. Rev. 45 52–68
[3] Elimelech M and Phillip W A 2011 Science 333 712–7
[4] Shannon M A, Bohn P W, Elimelech M, Georgiadis J G, Marinas B J and Mayes A M 2008 Nature 452 301–10
[5] 1963 J. Franklin Inst. 276 183–4
[6] Khawaja A D, Kutubkhanah I K and Wie J M 2008 Desalination 221 47–69
[7] Periade B and Garcia-Rodriguez I 2012 Desalination 284 1–8
[8] Pérez-González A, Urtiaga A M, Ibáñez R and Ortiz I 2012 Water Res. 46 267
[9] Lee K P, Arnot T C and Mattia D 2011 J. Membr. Sci. 370 1–22
[10] Gude V G 2015 Appl. Energy 137 877–98
[11] Ghaffour N, Bundschuh J, Mahmoudi H and Goosen M F A 2015 Desalination 356 94–114
[12] Bennett A 2011 Filtr. Separat. 48 24–7
[13] Zhou L et al 2016 Nat. Photon. 10 393–8
[14] Lin L C and Grossman J C 2015 Nat. Commun. 6 8335
[15] Yang H Y, Han Z J, Yu S F, Pey K L, Ostrikov K and Karnik R 2013 Nat. Commun. 4 2220
[16] Kim S J, Ko S H, Kang K H and Han J 2010 Nat. Nanotechnol. 5 297
[17] Li W, Yang Y, Weber J K, Gang Z and Zhou R 2016 ACS Nano. 10 1829
[18] Cohen–Tanugi D and Grossman J C 2012 Nano Lett. 12 3602–8
[19] Heinrianan M, Fariman A B and Alurun N R 2015 Nat. Commun. 6 8616
[20] Chen W F et al 2013 ACS Nano 7 5308
[21] Yoshi K R et al 2014 Science 343 753–4
[22] Sun P, Wang K and Zhu H 2016 Adv. Mater. 28 2287–310
[23] Nair R, Wu H, Jayaram P, Grigorieva I and Geim A 2012 Science 335 442–4
[24] Huang H et al 2013 Nat. Commun. 4 2979
[25] Wei N, Peng X and Xu Z 2013 Phys. Rev. E 89 012115
[26] Yoshida H and Boucquet L R 2016 J. Chem. Phys. 144 234701
[27] Cohen–Tanugi D, Lin L–C and Grossman J C 2016 Nano Lett. 16 1027–33
[28] Wei N, Peng X and Xu Z 2014 ACS Appl. Mater. Interfaces 6 5877–83
[29] Chen B, Jiang H, Liu X and Hu X 2017 J. Phys. Chem. C 121 1321–8
[30] Chen B, Jiang H, Liu X and Hu X 2017 ACS Appl. Mater. Interfaces 9 22826
[31] Xue G et al 2017 ACS Appl. Mater. Interfaces 9 15052–7
[32] Chen B, Jiang H, Liu X and Hu X 2018 Phys. Chem. Chem. Phys. 20 92760–6
[33] Radha B et al 2016 Nature 538 222
[34] Kwac K, Kim I, Pascal T A, Goddard W A, Park H G and Jung Y 2017 J. Phys. Chem. C 121 16021–8
[35] Alghara–Siller G et al 2015 Nature 519 443
[36] Deng M, Kwac K, Li J, Jung Y and Park H G 2017 Nano Lett. 17 2342
[37] Jiao S and Xu Z 2017 ACS Nano 11 11152–61
[38] Jiao S, Duan C and Xu Z 2017 Sci. Rep. 7 2646
[39] Plimpton S 1995 J. Comput. Phys. 117 1–19
[40] Humphrey W, Dalke A and Schulte K 1996 J. Mol. Graph. 14 33–8
[41] Stukowski A 2009 Model. Simul. Mater. Sci. 18 015012
[42] Oyarzua E, Walther J C, Georgiadis J G, Koumoutsakos P and Zambrano H 2017 Nano 11 1997–10002
[43] Zhao K and Wu H 2015 Nano Lett. 15 3664–8
[44] Zhao K and Wu H 2017 Phys. Chem. Chem. Phys. 19 28496
[45] Bregulla A P, Wärmer A, Günther K, Mertig M and Cichos F 2016 Phys. Rev. Lett. 116 186803
[46] Fu L, Merabia S and Joly L 2018 J. Phys. Chem. Lett. 9 2086–92
[47] Fu L, Merabia S and Joly L 2017 Phys. Rev. Lett. 119 214501
[48] Mohammadzadeh A, Rana A S and Struchtrup H 2015 Phys. Fluids. 27 420–8
[49] Shahabi V, Baier T, Roohi E and Hardt S 2017 Sci. Rep. 7 41412

Simulations results show that water molecules are driven to migrate through nanometer-wide graphene galleries from cold reservoir to hot reservoir by the effect of thermal creep flow. It derives to the imbalanced momentum exchange between graphene slabs and water molecules from two reservoirs. The flow velocity is strongly related to the temperature gradient and get an increasing trend as the form of natural logarithm with greater temperature difference, corresponding to the prediction of kinetic theory. The size effect is another significant factor on water flow. The flowrate monotonically decreases with increasing layer spacing beyond 9 Å, showing impaire size effect for greater $d$-spacing. The abrupt escalation in water permeation between the 9 and 6.5 angstrom-distance graphene slabs suggests a transition to subcontinuum transport, which is consistent with the change from disordered water to quasi-square structure with decreasing $d$-spacing. The collective diffusion of water molecules inside graphene channels and special transport processes with structure change lead to the unexpectedly high water permeability through graphene galleries. Movement of saline ions are also influenced by the system with temperature gradient, driven to move at a reverse direction of water flow owing to the thermophoretic force. On one hand, the graphene channels could reject ions by geometry limitation ($d$-spacing of 6.5 Å), and on the other hand, the graphene channels could reject ions by geometry through graphene galleries. Movement of saline ions is another significant factor on water flow owing to the thermophoretic force. On one hand, the graphene channels could reject ions by geometry limitation ($d$-spacing of 6.5 Å), and on the other hand, the graphene channels could reject ions by geometry.
[50] Sharipov F and Selezniev V 1998 J. Phys. Chem. Ref. Data. 27 657–706
[51] Ohwada T, Sone Y and Aoki K 1989 Phys. Fluids A 1 1588–99
[52] Sone Y 2007 J. Phys. Soc. Japan 76 1836–7
[53] Schaaf S A and Chambré P L 2017 Flow of Rarefied Gases (Princeton, NJ: Princeton University Press)
[54] Kennard E H 1938 Kinetic Theory of Gases with an Introduction to Statistical Mechanics (London: McGraw-Hill)
[55] Zhang C, Gygi F and Galli G 2013 J. Phys. Chem. Lett. 4 2477–81
[56] Fumagalli L et al 2018 Science 360 1339
[57] Talbot I, Cheng R K, Schefer R W and Willis D R 1980 J. Fluid. Mech. 101 737–58
[58] Piazza R and Parola A 2008 J. Phys.: Condens. Matter 20 153102
[59] Chowdhuri S and Chandra A 2001 J. Chem. Phys. 115 3732–41
[60] Vigolo D, Rusconi R, Stone H A and Piazza R 2010 Soft Matter. 6 3489