Supplementary Information

Nanoscale Two-Phase Flow of Methane and Water in Shale Inorganic Matrix

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Temperature as a function of simulation time

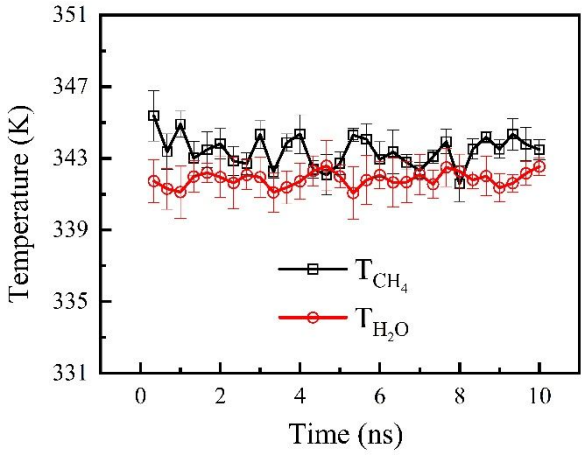

Fig. S1 Temperature as a function of simulation time

Calculation of Average Streaming Velocity

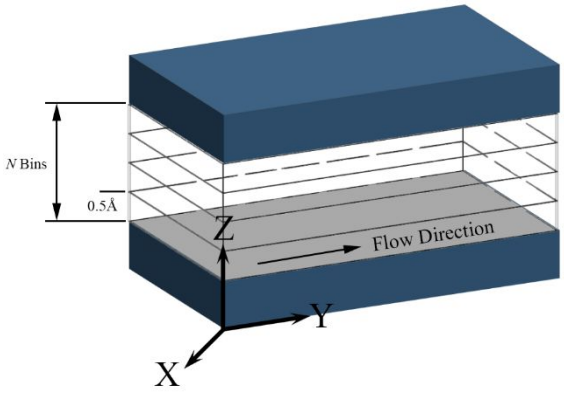

Fig. S2 Schematic diagram of ASV.

Based on the bin method[1], we compute the average streaming velocity, as shown in Fig. S2 and equation (1). The slit is divided into N bins in the Z direction. The width of each bin is equal to 0.5 Å. For each bin, the center-of-mass velocity of the confined fluid at steady state is extracted from time step J_n to J_m of the trajectories

$$v_{bin} = \frac{\sum_{j=J_n}^{J_m} \sum_{i \in bin} m_i v_{iy,j}}{\sum_{i \in bin} m_i} \tag{1}$$

where m_i is the mass of the particle i in the bin, v_{iy} is they-component velocity of the particle i at time step j in the bin.

No slug flow for water at a = 0.002 nm/ps^2

To explore no slug flow for water, we observe the distribution of water molecules involved in the water bridge flowing through the nanopore at different time, as shown in Fig. S4. These water molecules are marke in red and the other water molecules are in cyan. It is observed that water molecules transfer between water film and water bridge when water flows in the nanopore. Many water molecules in the water bridge enter into the water film and many water molecules in the water film move into the water bridge. They may exchange their velocities and still remain the flow characterization of Poiseuille flow. That is, water near the surface has a small velocity due to the strong effect of the surface and water in the central region of the nanopore has a high velocity because of the weak effect of the surface.

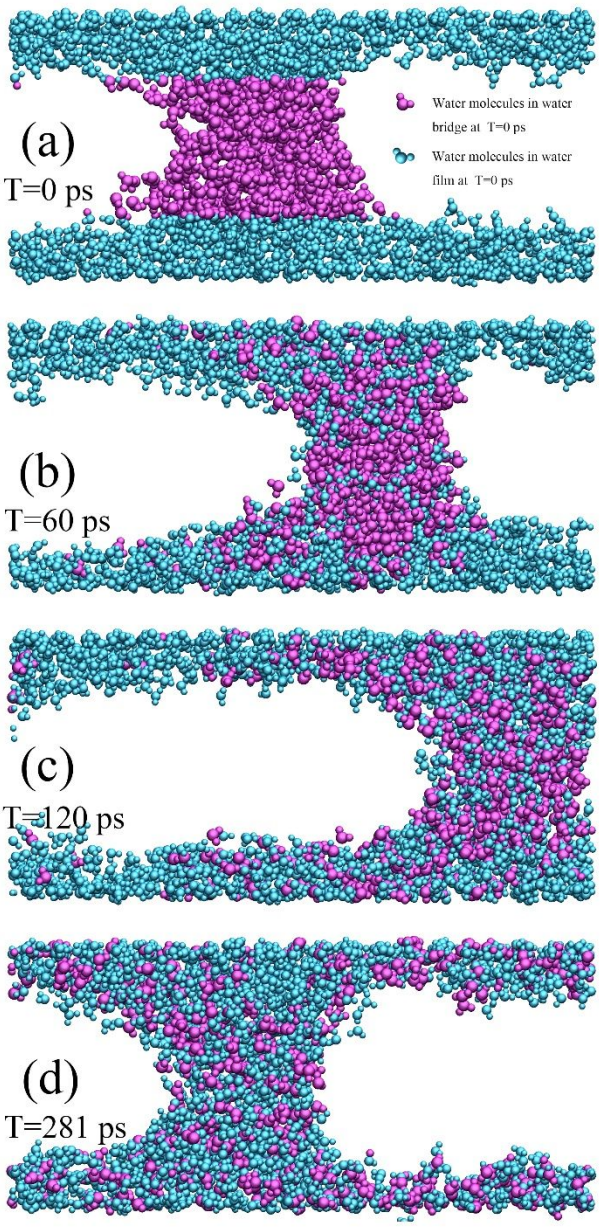

Fig. S4 Distribution of water molecules in water bridges at different time.

To compare with slug flow of water, we carry out the additional mulecular dynamics simulation on the two-phase flow of water and methane in a slit-shaped graphene nanopore, as shown in Fig. S5. It shows that the structure of water-in-methane is formed and no water film near the surface occurs. A sharp jump is observed in the velocity profile near the surface, implying a slug flow occurs inside the nanopre, as shown in Fig. S5d. Such slug flow is the natural outcome of the low friction between the water molecules and the graphene surfaces due to the weak interaction of water with the surface[2]. Fig. S5 also shows that water molecules transfer between the region near the surface and the central region of the nanopore. They exchange their velocities and yet remain the characterization of slug flow.

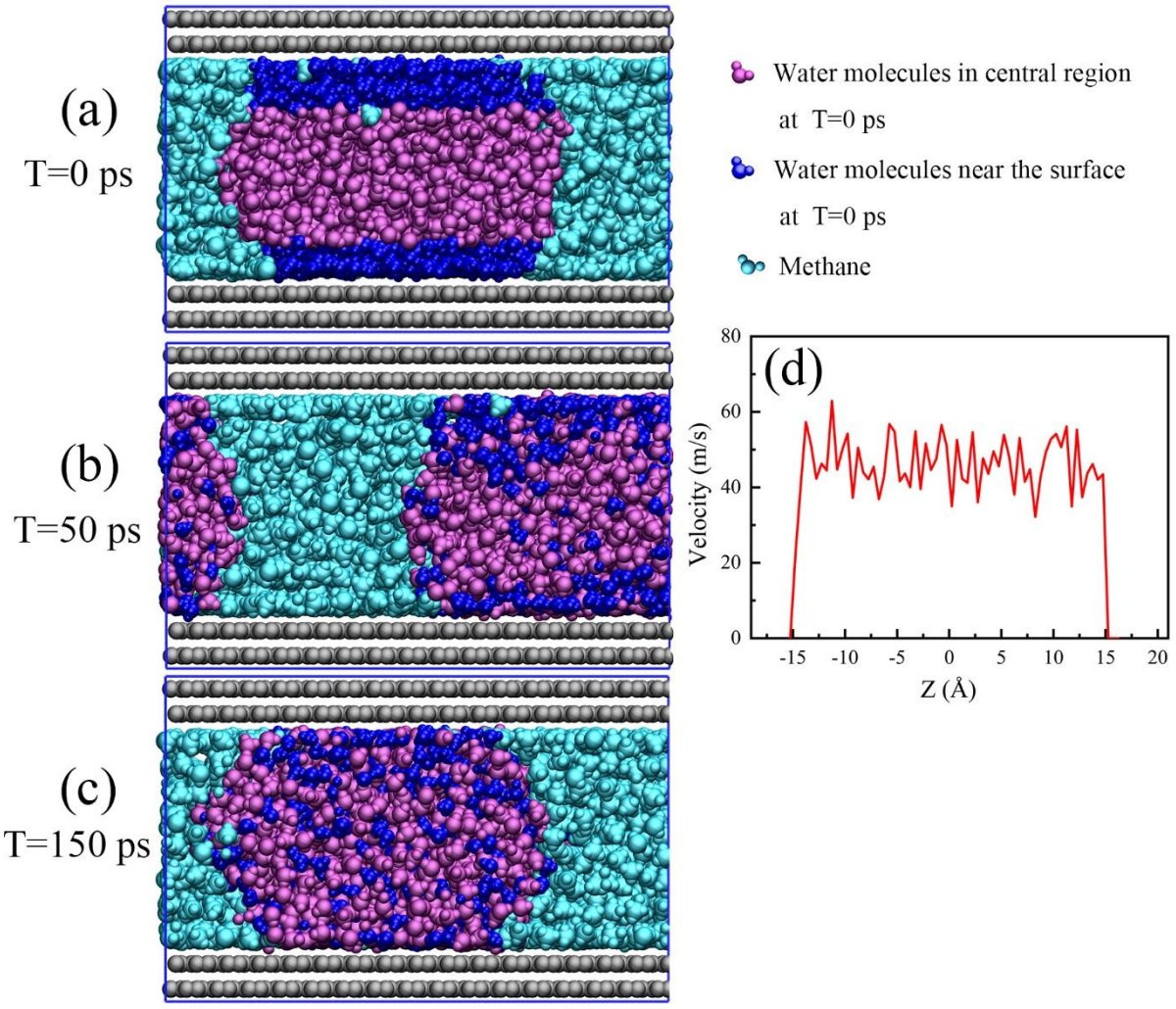

Fig. S5 (a)-(c) Distribution of water and methane molecules at different time and (d) the velocity profile in graphene nanopore.

Reference

(1) Hansen, J. S.; Todd, B.; Daivis, P. J., Prediction of Fluid Velocity Slip at Solid Surfaces. *Physical Review E* **2011,** *84*, 016313.

(2) Liu, B.; Wu, R.; Baimova, J. A.; Wu, H.; Law, A. W.; Dmitriev, S. V.; Zhou, K., Molecular Dynamics Study of Pressure-Driven Water Transport through Graphene Bilayers. *Phys. Chem. Chem. Phys.* **2015,** *18*, 1886-1896.