Supporting Information
Nanoscale Gas Accumulation At Solid-Liquid Interfaces: A Molecular Dynamics Study

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Figure S1 shows the solid-liquid contact angle at graphene surface.

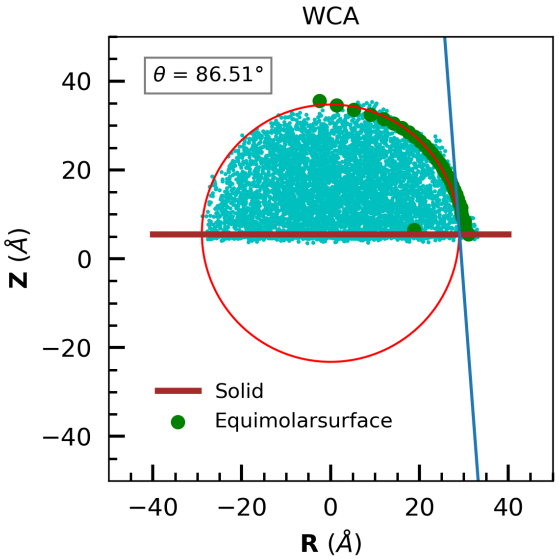

Figure S1: Water nanodrop contact angle at graphite surface

Figure S2 shows the density and pressure for a nitrogen gas molecules in a rectangular box at 50 atmospheric pressure and $300K$.

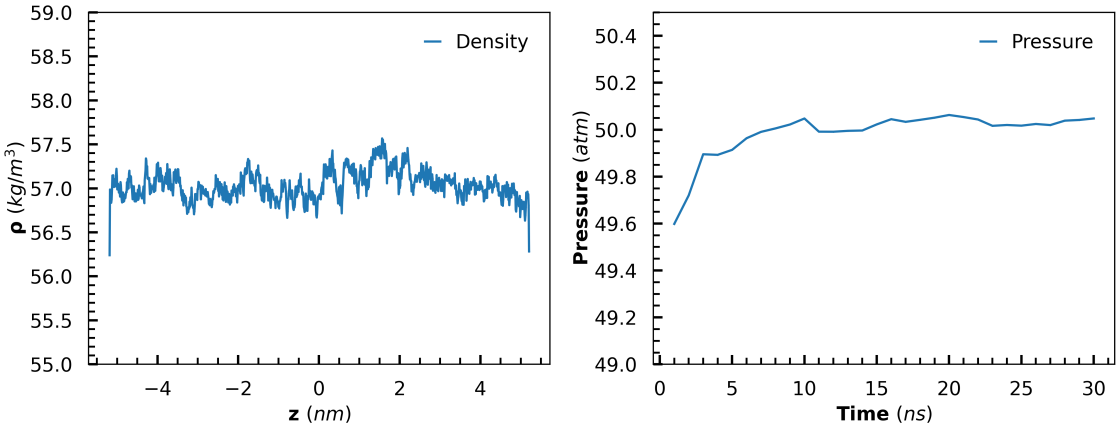

Figure S2: Density and pressure with time as obtained from simulation.

Figure S3 shows the converged surface tension profile at $300K$ for the case of pure water[1] and nitrogen dissolved water. The surface tension can be calculated using the components of the pressure tensor using

$$\gamma = \frac{1}{2}\int_{-\infty}^{\infty}[P_N(z) - P_T(z)]dz \tag{1}$$

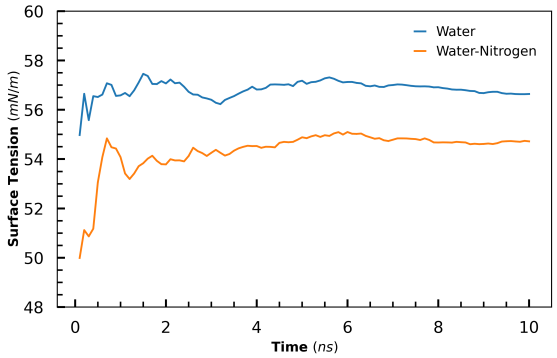

Figure S3: Surface tension for pure water and nitrogen dissolved in water. Note that the values obtained from the simulations are excluding the long range corrections.

Figure S4 shows the density profile of nitrogen and water at 50 atmospheric pressure and 300K. The water box is sandwiched on the top and bottom with nitrogen gas molecules and nitrogen dissolves in water accordingly at the set pressure and temperature. Periodic boundary conditions are stipulated for all three dimensions.

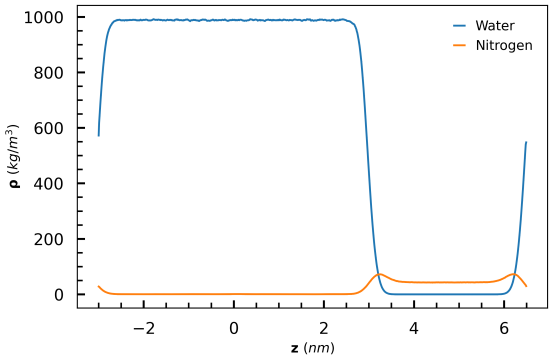

Figure S4: Nitrogen dissolved in water at 50atm and 300K

Figure S5 shows gas accumulation and the density profile for a gas that is approximately 2 times more attracted towards the wall than its intermolecular interaction with itself after a period of 30 ns.

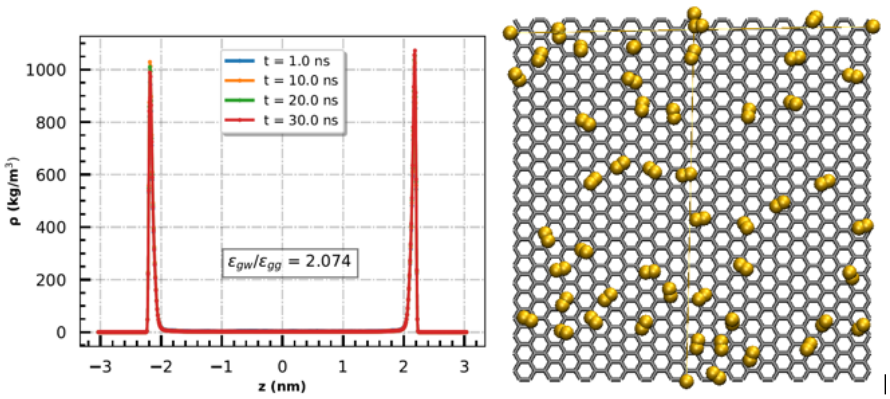

Figure S5: Gas adsorbed on the lower graphene wall. The figure on the left shows the density profile of gas inside channel when the gas-wall interaction strength is increased by factor of 2 i.e eps_{gw}/eps_{gg}. The figure on the right shows the screenshot configuration of gas molecules adsorbed on the lower wall captured from model after a period of 30 ns. The standard error for the five independent simulations is 0.005411 over 608 data points.

It is observed that near the walls, density peaks to 1000 kg/m^3, which represents a high-density state similar to that of a liquid. However, we could not observe multi layering on the walls but rather a closed pack aggregate form of gas molecules adhering to the walls. Note that if we consider a system where there is a 7 times increase in the number of nitrogen molecules than the number appropriate at a pressure of $\sim$ 50 atm, an additional layer of gas molecules can be seen building on the graphene sheet.

Figure S6 and Figure S7 shows density profile in the channel and gas adsorbed on the lower wall of the simulated model when the number of nitrogen molecules inside the channel is increased from 100 to 700. The channel size is 8 * 8 * 6 nm.

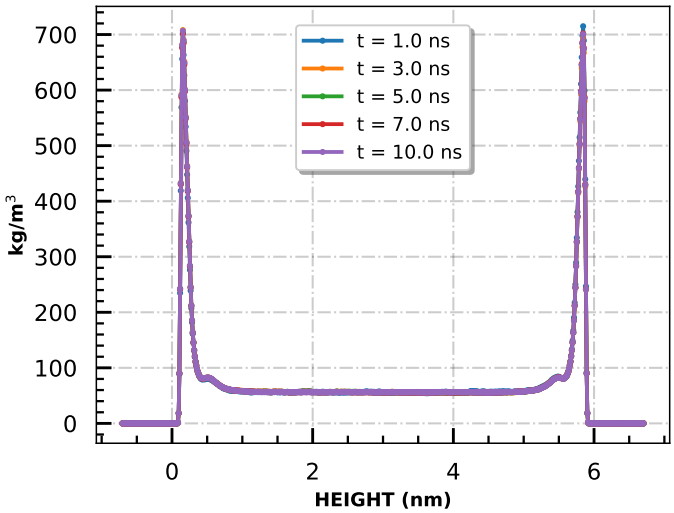

Figure S6: Density profile for higher no of nitrogen

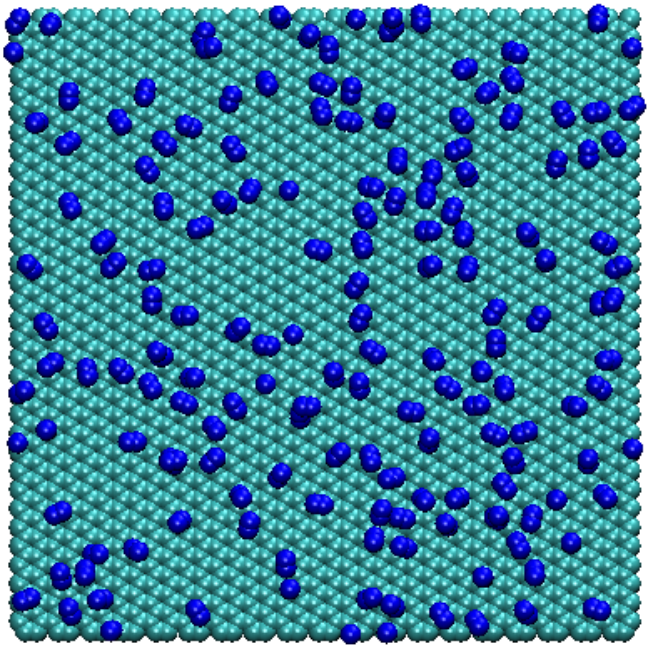

Figure S7: Gas adsorbed at the lower wall for higher no of nitrogen

References

[1] José Alejandre, Dominic J Tildesley, and Gustavo A Chapela. Molecular dynamics simulation of the orthobaric densities and surface tension of water. *The Journal of chemical physics*, 102(11):4574–4583, 1995.