

Wettability of graphite under 2D confinement

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The thermodynamics of solid/liquid interfaces under nanoconfinement has tremendous implications for liquid transport properties. Here using molecular dynamics, we investigate graphite nanoslits and study how the water/graphite interfacial tension changes with the degree of confinement. We found that, for nanochannel heights between 0.7nm and 2.6nm, graphite becomes more hydrophobic than in bulk, and that the value of the surface tension oscillates before eventually converging towards a constant value for larger slits. The value of the surface tension is correlated with the slip length of the fluid and explained in terms of the effective and interfacial density, hydration pressure and friction coefficient. The study clearly indicates that there is a critical channel height of 0.9nm (achievable experimentally¹) at which the surface tension reaches its highest value, but the water diffusion across the channel is at its minimum. The structural analysis shows that for this pore size a transition between a 2D and 3D hydrogen bond network is accompanied by an abrupt increase in conformational entropy. Our results show that the wettability of solid surfaces can change under nanoconfinement and the data can be used to interpret the experimental permeability data.

REFERENCES

- [1] Williams, C. D., Wei, Z., Shaharudin, M. R. bin & Carbone, P. A molecular simulation study into the stability of hydrated graphene nanochannels used in nanofluidics devices. *Nanoscale* **14**, 3467–3479 (2022).

FIGURES

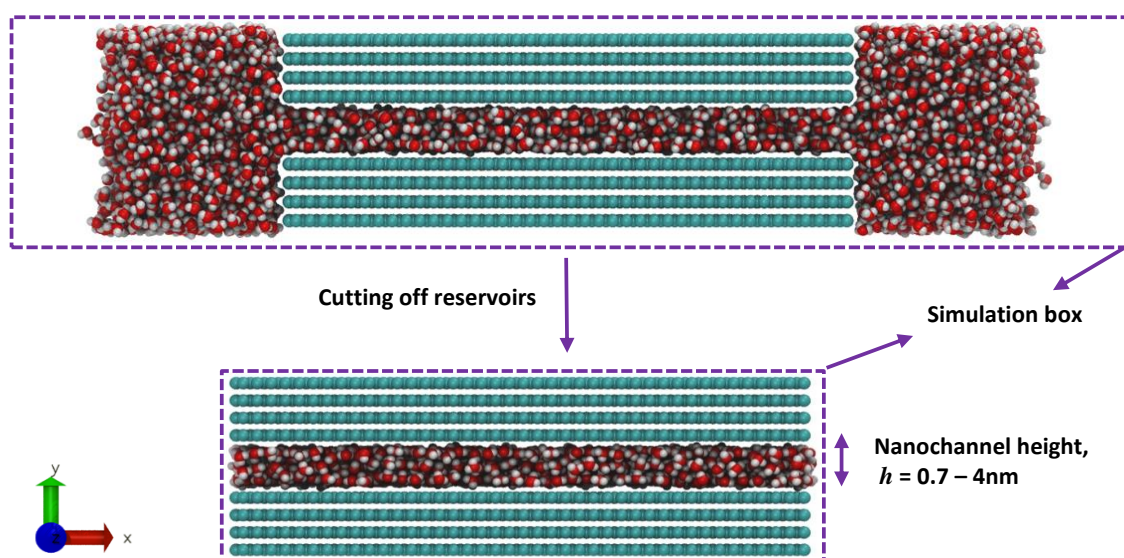


Figure 1: Side-view of water confined in a graphitic nanochannel of 1.2nm height: snapshot of the channel in contact with the two reservoirs after the water has filled the channel (top); snapshot of the channel after the reservoirs have been removed (bottom). The y-direction is normal to the graphite surface. Periodic boundary

conditions are applied in all three directions. The surface area is defined by $A = LxLz$. The red, white, cyan spheres represent oxygen, hydrogen, and carbon atoms respectively.