

Towards Predicting And Controlling Ionic Hydration Patterns In Nanopores

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MLB is looking for a postdoc position, ideally commencing in April 2021

Intro

Numerous biophysical and technological processes require control of the selective and conductive properties of the constituent ion-transporting biological or artificial membranes [1,2]. These properties are largely defined by the ionic hydration barrier, that emerges when the water molecules, surrounding the ion, are detached or redistributed [3]. This water structures (patterns) are complex and highly-fragmented as routinely reported in molecular dynamics (MD) simulations, while their analytical prediction has been overlooked. Here [4], we

- provide an analytical method predicting the hydration patterns,
- demonstrate the pore's features it grasps, and
- discuss the method's potential applications in biophysics and nanotechnology.

Methods

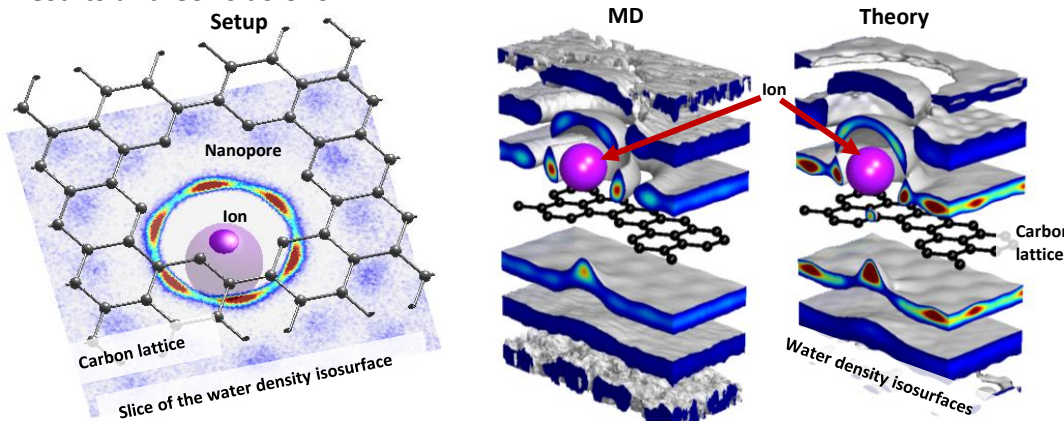
Water density ρ is described by

$$\frac{\rho(\mathbf{r}|\{\mathbf{r}_m^c\}, \mathbf{r}^i)}{\rho_0} = g^{iw}(|\mathbf{r} - \mathbf{r}^i|) \cdot \prod_m^{N^c} g^{cw}(|\mathbf{r} - \mathbf{r}_m^c|)$$

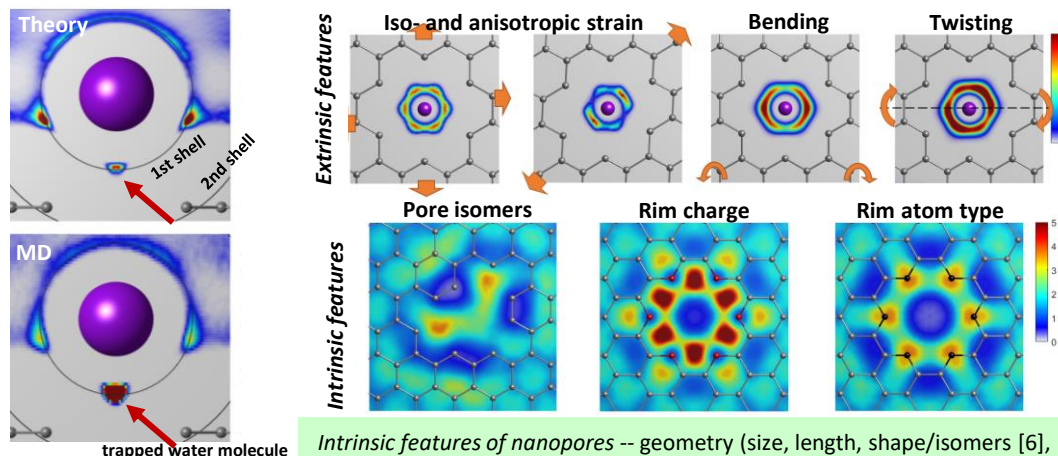
where ρ_0 is the bulk density, \mathbf{r} is the position of interest, \mathbf{r}^i and \mathbf{r}_m^c are the locations of the ion and lattice atoms (N^c in total), g^{iw} and g^{cw} are the bulk radial density functions (RDFs). This method extends the approach by Hummer and co-workers [5] by explicitly accounting for the ion's presence.

Classical all-atom MD simulations were run in NAMD to calculate the water densities. For more details, see Ref. [4]. Ions and lattice atoms are clamped throughout the simulation.

Results and Conclusions



Good agreement between MD and our method is found. Arbitrary ionic positions can be considered (see dynamic [movie1](#) and [movie2](#) online).



Method successfully predicts the locations of trapped water molecules

Intrinsic features of nanopores -- geometry (size, length, shape/isomers [6], number of layers, offset eclipse) and their **Extrinsic features** -- effect of iso- and anisotropic strain, bending, and twisting -- can all be accounted for by our method. Other lattices, e.g. hBN or MoS₂, can be considered as well.

Applications

The proposed method can provide fundamental insights into the electrostatics, dielectric properties, and dynamics of ions in sub-nanopores. It can be further used to predict analytically the ionic potential of the mean force (PMF) – the key descriptor of energy landscape in nanopores. This opens the way to design and optimisation of novel nanoionic devices (including biomimetic ones) via the control of the hydration patterns. Attractive targets involve

- **“blue energy” harvesting:** the trade-off between energy harvesting efficiency and achievable power output [2]
- **water desalination:** the water quality and energy efficiency (i.e. minimal energy cost) [1,2]
- **DNA sequencing:** detection of specific nucleotide via ionic currents, or slowing down the DNA's permeation [2,5,6].

Further work includes

1. Extending the method carefully to yield the PMF, paying specific attention to the dielectric permittivity and electrostatics at nanoscale. The latter topic has been a subject of intense research recently and still remains an unsolved problem.
2. Generalization of the method to describe hydration during non-equilibrium processes, such as translocation events.

Acknowledgements

The work was partially supported by a PhD Scholarship from the Faculty of Science and Technology of Lancaster University, by the Engineering and Physical Sciences Research Council UK (grants EP/M016889/1 and EP/M015831/1), and by a Leverhulme Trust Research Project Grant RPG-2017-134. MLB, CG, WATG, DGL and PVEMcC gratefully acknowledge the use of the High End Computing facility at Lancaster University. AS gratefully acknowledges support from the Materials Genome Initiative.

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