

Ionic transport in graphene-based single digit nanopores: Insights from molecular modelling

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Understanding the microscopic behavior of aqueous electrolyte solutions in graphene-based nanochannels with heights comparable to a few molecular diameters is important in nanofluidics applications such as water purification, fuel cells, and molecular sensing. Under such extreme nanoscale confinement (< 2 nm), the physical and chemical properties of water and ions differ drastically from those in the bulk phase. In this work, we study the structural and diffusion behavior of several prototypical aqueous solutions of electrolytes (LiCl, NaCl, and KCl) confined in both neutral and positively-, and negatively-charged graphene nanochannels. We perform molecular dynamics simulations of the solutions in the graphene nanochannels with either one, two- or three-layer water structures using the effectively polarizable force field for graphene [1].

While overall fluxes of matter in confined geometries can be measured by macroscopic techniques, computer simulations provide detailed analysis of these fluxes as a function of distance from the surface, position within pores, types of ions forming aqueous solutions, surface composition, charges, and geometry. We overview our techniques for determination of dynamic properties at macroscopic interfaces (solid-liquid interface) and in confined geometries and present representative results.

The dynamic properties of water and ions in aqueous solutions include (i) distance-dependent diffusivity determined in absence of external electric field or pressure gradient, distance-dependent (ii) mobilities determined in the presence of external fields, and (iii) the corresponding overall diffusivities and fluxes through the nanopore. We focus on graphene-based planar nanopores [1,2] and study the role of graphene surface charge, the separation of the graphene planes defining the height of the nanopore, as well as the length of the nanopore. The effect of the magnitude of the external fields (electric field, pressure drop) is also investigated.

The goal of our ongoing work is to help in designing efficient nanodevices applied to aqueous solutions, namely desalination devices based on capacitive deionisation (CDI) [2,3].

References:

- [1] J. Dočkal, F. Moučka, M. Lísal, *J. Phys. Chem. C* 123 (2019) 26379-26396.
- [2] J. Dočkal, M. Lísal, F. Moučka, *J. Mol. Liq.* 353 (2022) 118776.
- [3] Y. H. Teow, A. W. Mohammad, *Desalination* 451 (2019) 2–17.

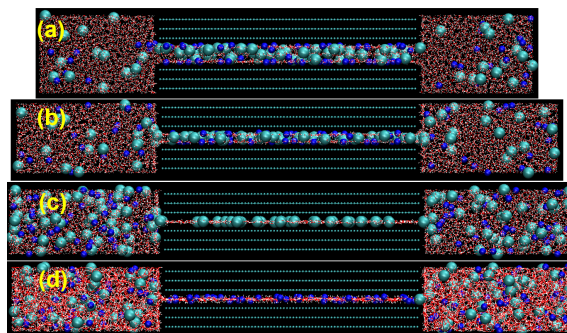


Figure: Examples of the simulation box for the bulk reservoir-graphene nanochannel set-up with an aqueous NaCl solution. (a) Neutral graphene nanochannel of the height $H = 12$ Å with a three-layer water structure, (b) neutral graphene nanochannel of $H = 9$ Å with a two-layer water structure, (c) positively-charged graphene nanochannel of $H = 7$ Å with a one-layer water structure, and (d) negatively-charged graphene nanochannel of $H = 7$ Å with a one-layer water structure.