

Bridging Scales in Electrode Systems: Investigating the Phase Behavior of Fluids in Nanoporous Systems with Enhanced Sampling

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Electrode systems are fundamental components of energy storage and catalysis devices, and their properties at different length scales are essential for optimizing their performance. Nano-porous materials are particularly promising due to their advantageous physical properties and very high specific surface area. However, the physics of confined fluids in nanometer-scale pores differs significantly from that of bulk fluids, as the atomistic nature of fluid molecules and pore-walls becomes important. The modelling of such systems necessitates consideration of quantum mechanical details, which become important at this length scale.

These quantum mechanical properties are determined from ab-initio calculations and incorporated in classical MD models in order to reach the relevant time and length scales. Additionally, for materials with metallic properties such as graphene, we utilize a constant potential scheme with parameters that are tuned to quantum density functional theory [2]. We are interested in bridging the length and timescales in order to go from microscopic, quantum mechanical details to the macroscopic properties of materials relevant for technical applications. To achieve this, we use coarse-graining procedures that allow us to retrieve continuum models with modified parameters which capture the atomistic details effectively. These procedures involve integrating out the degrees of freedom of individual atoms or molecules to obtain a simplified model that captures the essential features of the system at technically relevant scales [1].

In order to accurately model the behavior of electrode pores, it is crucial to consider them as open systems, where only a small subsystem is explicitly simulated, allowing for particle exchange with an outside reservoir. This approach enables changes in composition due to complex interactions between the wall, fluid, and any applied potential on the electrodes in the system. In simulations, Grand Canonical Monte Carlo methods are commonly used to model these fluctuations in particle numbers. However, these methods can be slow and inefficient, especially for larger systems and when multiple species of fluid molecules are involved. For this reason, we employ the Wang-Landau enhanced sampling method [3]. The Wang-Landau sampling method allows us to efficiently determine the density of states of the system, by iteratively approximating the free energy surface, which can be done in a massively parallel procedure. By using this method, we hope to accurately capture the behavior of electrode systems across different length scales and investigate the behavior of confined fluids in binary and even tertiary mixtures. This is essential because the behavior of confined fluids in these mixtures can differ significantly from bulk fluids, and

the behavior of the electrode can also be affected.

By improving our understanding of the behavior of electrode pores, we can optimize the design of electrode systems and improve their performance for these applications. Our poster highlights the importance of understanding the behavior of pores in electrode systems and the usefulness of the Wang-Landau method in this context.

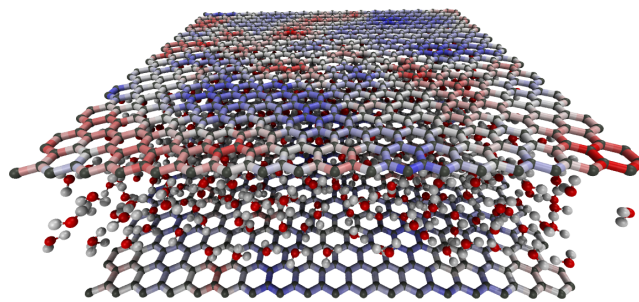


Figure 1: Water molecules confined between graphene sheets. The colors on the graphene sheets visualize the fluctuating surface charge of the metallic graphene. From these simulations, macroscopic parameters are determined, such as the transport coefficients or the dielectric response.

References

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