École polytechnique fédérale de Lausanne (EPFL) Valais/Wallis
Institute of Chemical Sciences and Engineering (ISIC)
Basic Science Faculty (SB)
Laboratory of molecular simulation (LSMO)
Rue de l'Industrie 17, CH-1950 Sion, Switzerland

SEMINAR

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Investigating the properties of the electrode-electrolyte interface in porous carbon based supercapacitors using idealised coarse-grained models

Céline MERLET
CIRIMAT, CNRS, Université de Toulouse, France

Local fluid structure and ion transport in porous materials are relevant for a number of applications including energy conversion and storage, heterogeneous catalysis and drug delivery. In all these cases, the performance of the systems is highly related to the specific properties of the fluid under confinement. In order to understand fundamentally the macroscopic properties of such systems, it is essential to characterise finely the porous materials used and the structural and dynamic properties of the fluid adsorbed. In this presentation, I will focus on carbon-carbon supercapacitors which store energy at the electrolyte/carbon interface through reversible ion adsorption. I will show how molecular dynamics simulations can be used get a microscopic picture of the phenomena at play. In the past, these simulations have proven useful in explaining the large capacitance increase observed when nanoporous carbons are used as electrode materials1 or in characterising the desolvation of ions upon confinement in the electrode2. Nevertheless, the relationship between the carbon structure, the electrolyte nature and the electrochemical performance is still unclear. In the present work, we use molecular dynamics simulations to investigate the structural and dynamic properties of concentrated electrolytes in contact with various porous carbons (ordered3 and disordered4). The concentrated electrolyte is represented using a simple coarse-grained model which allows us to tune its characteristics (e.g. ion size, ion charge) and analyse the effect of such variations on the properties of the system. In particular, we determine the radial distribution functions of the ions inside the carbon and the diffusion coefficients of the various species. I will also introduce the lattice models we are developing to bridge the gap between molecular simulations and experiments. These models, versatile and very computationnally efficient, allow us to reach experimental length and timescales and predict useful quantities such as quantities of adsorbed ions, NMR spectra or tortuosities5,6.

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CV : Dr Céline Merlet

Dr Céline Merlet received her PhD in 2013 from Université Pierre et Marie Curie (Paris) where she worked under the supervision of Prof. Mathieu Salanne and Dr Benjamin Rotenberg on molecular modeling of ion adsorption in carbon nanopores. She then moved to the University of Cambridge as a postdoctoral fellow to work in the group of Prof. Clare Grey, in collaboration with Prof. Daan Frenkel, on the development of mesoscopic models to improve the interpretation of NMR spectra of electrochemical systems. In 2017, she was appointed as a CNRS researcher at Université Paul Sabatier in Toulouse where she is working towards predicting performances of energy storage systems.