Multi-scale models to study dynamics and confinement in materials for energy storage applications: From the molecular to the experimental scale

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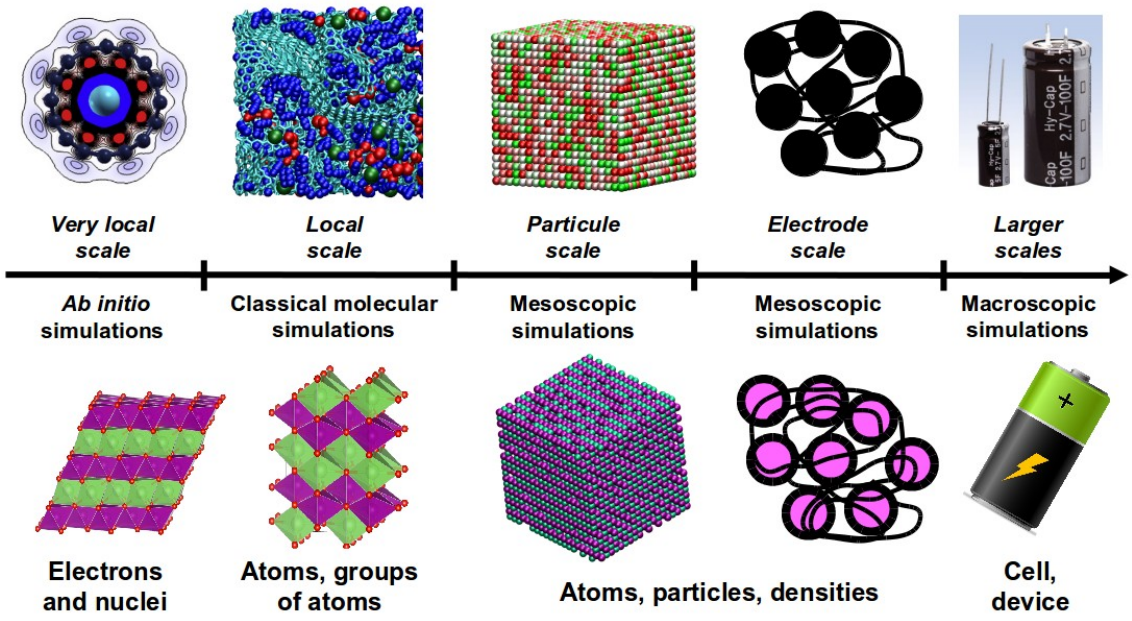
Progress in the development of novel energy storage systems such as batteries and supercapacitors is hampered by our lack of understanding of the microscopic mechanisms that determine their performance. The key issue is that phenomena on the atomistic scale have consequences on macroscopic length and timescales. In particular, the effects of ionic confinement and diffusion are crucial for device performance, yet experiments that probe properties related to local structure and diffusion are challenging and difficult to interpret without a parallel modelling approach.

In this talk, I will show how classical molecular dynamics simulations have been used to explore the electrode-electrolyte interface in carbon-carbon electrical double layer capacitors, and explain the large capacitance increase observed when nanoporous carbons are used as electrode materials [1]. I will then introduce the lattice models I am currently developing to bridge the gap between molecular simulations and NMR experiments (Nuclear Magnetic Resonance). These models, versatile and very computationnally efficient, allow me to reach experimental length and timescales and predict useful quantities such as NMR spectra or tortuosities. I will first illustrate this modelling technique on the case of ions diffusing through a carbon particle [2, 3], relevant to the study of supercapacitors, before introducing the case of a battery material, namely $LiMnO_2$.



[1] C. Merlet, B. Rotenberg, P. A. Madden, P.-L. Taberna, P. Simon, Y. Gogotsi and M. Salanne, *Nature Mater.*, **11**, 306 (2012).
[2] A. C. Forse, C. Merlet, P. K. Allan, E. K. Humphreys, J. M Griffin, M. Aslan, M. Zeiger, V. Presser, Y. Gogotsi and C. P. Grey, *Chem. Mater.*, **27**, 6848 (2015).
[3] C. Merlet, A. C. Forse, J. M Griffin, D. Frenkel and C. P. Grey, *J. Chem. Phys.*, **142**, 094701 (2015).