Finite-Element modeling of the Electric Double-Layer and its application to the prediction of supercapacitor charging dynamics

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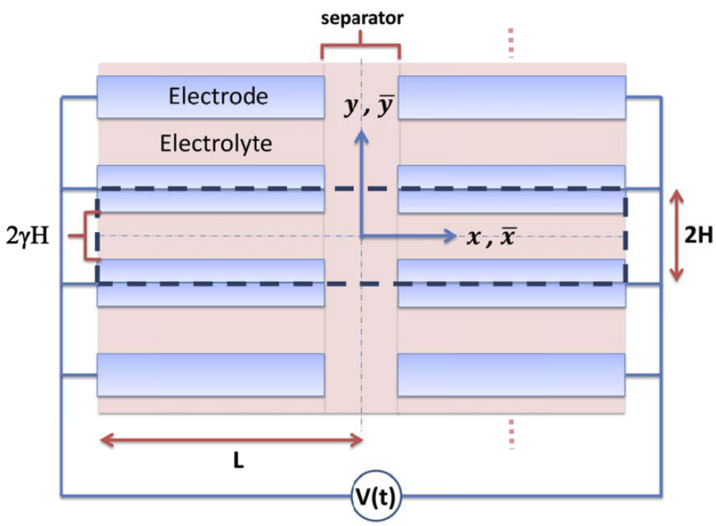
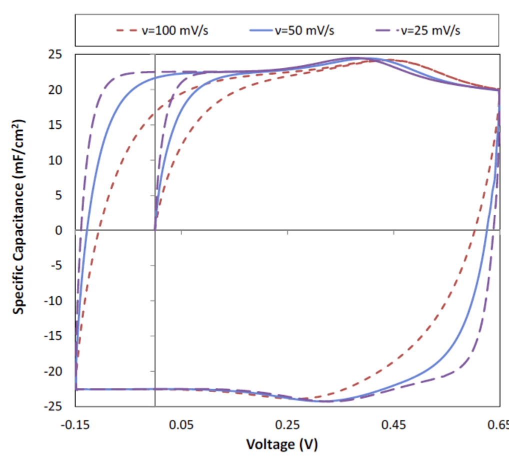
Charge storage in the Electric Double-Layer is used in a myriad of important devices, particularly for water desalination[1] and energy storage.[2] In supercapacitors, charge is rapidly stored and delivered by the re-distribution of charged ions into an adsorbed surface layer (commonly on high surface area carbon electrodes) and a diffuse layer in the electrolyte. While substantial work has and is being done using sophisticated computational techniques to better understand the double layer on the atomic scale, there is a need for models that can more reliably account for macroscopic aspects of these devices, being able to directly predict performance characteristics and thus guide their design.

In this work, we report on the development of a Finite Element approach to modeling of EDL devices, based on the Stern-Gouy-Chapman double layer model, which relates the local electric potential and ion concentrations in the electrolyte, and the modified Poisson-Nernst-Planck model,[3] which governs the dynamics of the charging process, while accounting for the finite size of the ions. Importantly, we can account for the effect of both ionic and electronic conductivity in the electrolyte and electrode, respectively, and show both contribute significantly to correctly predicting the behavior of realistic scale systems.

Unlike previous work, we deploy our model on structures with realistic dimensions, mimicking slit-pore geometries of varying size, and packed spheres similar to onion-like carbon electrodes. Predicted values of surface specific capacitance agree well with experiments on carbons nanomaterials, as do the charging dynamics as studied by cyclic voltammetry at different rates.





References:

1. S. Porada, L. Weinstein, R. Dash, A. van der Wal, M. Bryjak, Y. Gogotsi and P. M. Biesheuvel, *ACS Appl. Mater. Interfaces*, 2012, **4**, 1194-1199.
2. P. Simon and Y. Gogotsi, *Acc. Chem.* Res., 2012, doi: 10.1021/ar200306b.
3. M. S. Kilic, M. Z. Bazant and A. Ajdari, *Physical Review E*, 2007, **75**, 021503.