

TranSIESTA + QM/MM for electrochemistry

Ernane de Freitas Martins (a), Mazhar Iqbal (a,b), Ivan Cole (c), and Pablo Ordejón (a)

(a) Catalan Institute of Nanoscience and Nanotechnology (Spain)

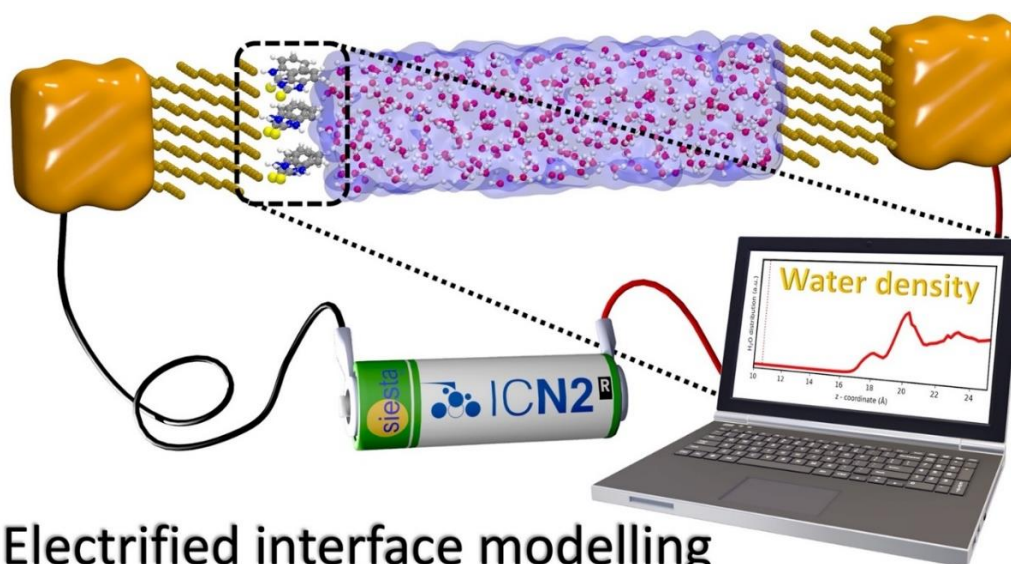
(b) RMIT University (Australia)

(c) Australian National University (Australia)

Although water is the most common solvent and one of the most extensively studied systems, understanding its structure and properties at material surfaces remains an open challenge. Molecular modelling stands out as a powerful technique to investigate critical electrochemical processes involved in the development of batteries, fuel cells, anti-corrosion coatings, and many other technologies where water interacts with metallic, often electrified, surfaces.

This scenario can be addressed using the non-equilibrium Green's function (NEGF) method [1,2], which enables the inclusion of the effect of an applied potential on the electrodes. However, the system sizes required to accommodate a realistic number of water molecules and the time scales needed for accurate sampling make the computational cost prohibitive.

To overcome this limitation, we propose a quantum mechanics/molecular mechanics (QM/MM) approach [3] coupled with the NEGF method, as implemented in the SIESTA package [4,5], to study metal-water interactions, providing an optimal balance between accuracy and computational efficiency. In this workshop, the proposed method will be demonstrated on a test system consisting of liquid water confined between gold electrodes, including a hands-on session.



Electrified interface modelling

- [1] M. Brandbyge et al., Phys. Rev. B 65, 165401 (2002)
- [2] N. Papior et al., Comp. Phys. Comm. 212, 8 (2017)
- [3] C. F. Sanz-Navarro et al., Theo. Chem. Accounts 128, 825 (2011)
- [4] J. M. Soler et al., Journal of Phys. Cond. Matter 14, 2745 (2002)
- [5] A. García et al., The Journal of Chem. Phys. 152, 204108 (2020)