

Understanding COF/Electrode interfaces for electrocatalysis using DFT and molecular simulations

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The structure of electrode/electrolyte interfaces is often studied using density functional theory (DFT), which provides a realistic depiction of the electrode's electronic structure. However, conducting ab-initio molecular dynamics simulations with controlled electrode potential and fluctuating electron numbers is challenging and expensive.

Combining first-principles calculations with force field-based molecular simulations broadens the scope to longer length and time scales, allowing for precise descriptions of dynamics in confined electrode systems. In this study, we use DFT to model COF/electrode systems to understand their behavior in electrocatalysis.

Comparing these models with experimental data from attenuated total reflectance surface-enhanced infrared absorption spectroscopy (ATR-SEIRAS)^[1] reveals insights into the dynamics at the electrode surface under applied potential.

This also allows us to study the effect of confinement on the microscopic electrolyte structure at the surface and how the electrical double layer changes, helping us to understand the impact of confinement on the catalytic process.

[1] K. Tschulik *et al.*, ACS Phys. Chem Au, 2023, 3, 1, 119–129, 10.1021/acspyschemau.2c00044