Séminaire du LCP-A2MC

How Efficient is DFTB for Electrochemical Interfaces? A Benchmark Study at Metal–Water Interfaces

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Modeling charged electrochemical interfaces with density-functional tight-binding (DFTB) requires both a reliable charge-control scheme and an accurate description of interfacial energetics and charge redistribution. Here, we assess SCC-DFTB for Au(111) by benchmarking two charge-control strategies in DFTB$^+$ – an explicit counter-electrode (EC) and a fixed–total–charge scheme (FC) – against grand-canonical DFT. We study H_2O, OH, and H adsorption in vacuum and with explicit water. For Au(111), DFTB yields work-function and potential-of-zero-charge values in good agreement with experiment. EC correctly reproduces the Fermi-level response to interfacial charging, whereas FC underestimates it, highlighting the importance of an explicit counter electrode for electrochemical response properties. Charge partitioning is qualitatively captured, but quantitative offsets remain. Adsorption energies are nearly identical between EC and FC, showing that an explicit counter electrode is not essential for these quantities; however, DFTB overbinds H_2O/OH and underbinds H. Force analysis reveals only moderate agreement in magnitude and poor reproduction of directional trends, especially for simple adsorbates and in solvated systems. Moreover, explicit solvent does not improve the results and often amplifies discrepancies across all properties. Overall, DFTB provides a useful qualitative description of charged Au(111) interfaces, but its quantitative accuracy remains limited by parameterization and by challenges in describing the electrode-electrolyte interface.

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