

Investigating organic molecules at the water-platinum interface: From single-crystal surfaces to nanoparticles via density functional tight binding

Stephan N. Steinmann^a, Qing Wang^a, Mingjun Gu^a, Carine Michel^a,
Thomas Niehaus^b

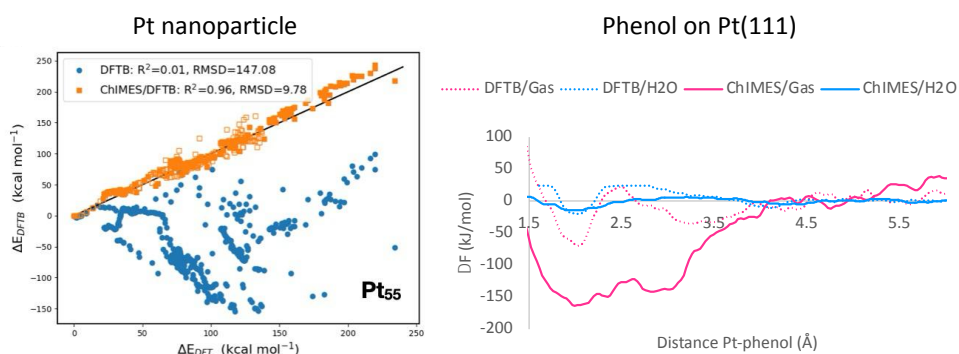
^aLCH, UMR 5182, CNRS, ENS de Lyon, 69342, Lyon, France.

^bILM, UMR5306, Université Claude Bernard Lyon 1, 69622, Villeurbanne, France.

stephan.steinmann@ens-lyon.fr

The platinum/water interface is a prototype to investigate the impact of transitioning from gas-phase petrochemistry to aqueous-phase biomass valorization. The aqueous environment is notoriously difficult to capture for computational methods:[1] implicit solvent models fail to capture near chemisorption on noble-metal surfaces, and DFT-based molecular dynamics are too costly in view of the slow diffusion constants at the solid/liquid interface. Even though QM/MM for the evaluation of the solvation free energy using dedicated force fields such as GAL17 leads to qualitative agreement with experiment.[2] To go beyond additive force fields, we introduce a density functional tight binding (DFTB) parametrization for the Pt/C/H/O system. We quantify the accuracy describing the adsorption and reaction of polyols on Pt(111) as well as the dynamic (morphological) changes of nanoparticles (Pt₃₈, Pt₅₅ and Pt₁₄₇) to be around 10 kcal/mol. Our parametrization efforts show that DFTB requires a corrective three-body potential, introduced via ChIMES[3] to capture water adsorption on Pt(111) and for the structure and reactivity related to nanoparticles. We showcase the difference between DFTB and DFTB/ChIMES on the adsorption energy of phenol on Pt(111) in the gas-phase and in water, see Figure 1.

Figure 1 Comparison of DFTB and DFTB/ChIMES.



References

- [1] S. N. Steinmann, C. Michel, ACS Catal. (2022), **12**, 6294.
- [2] P. Clabaut, B. Schweitzer, A. W. Götz, C. Michel, S. N. Steinmann, J. Chem. Theory Comput. (2020), **16**, 6539.
- [3] N. Goldman, L. E. Fried, R. K. Lindsey, C. H. Pham, R. Dettori, J. Chem. Phys. (2023), **158**, 144112.