

Study of the Oxygen Reduction Reaction (ORR) using a covalent organic radical framework as an electrocatalyst

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We study the use of 2D Covalent Organic Radical Frameworks (CORFs) based on polychlorotriphenylmethyl (PTM) radicals as electrocatalysts for the oxygen reduction reaction (ORR). The ORR is an essential process in fuel cells, but is limited by slow kinetics. 2D PTM-CORFs have low energy electron accepting states, making them potentially efficient for ORR in basic media [1]. Using density functional theory (DFT) calculations and the standard hydrogen electrode (SHE) as a reference [2], we calculated the stability of the ORR intermediates interacting with the PTM-CORF (see Figure 1). Our systematic study highlights the importance roles of both solvation and charge on the stability of the reaction intermediates and the calculated overpotential. Overall, our results show that 2D PTM-CORFs exhibit promising characteristics as electrocatalysts due to their open accessible structure and flexible electron-accepting electronic character, which allow for effective interactions with the species involved in the ORR. Our findings highlight the potential of 2D radical-based systems for enhancing the efficiency of the ORR in energy applications (e.g. fuel cells, batteries).

- [1] Wu, S.; Li, M.; Phan, H.; Wang, D.; Heng, T. S.; Ding, J.; Lu, Z.; Wu, J. Toward Two-Dimensional π -Conjugated Covalent Organic Radical Frameworks. *Angew. Chem.* 2018, 130(27), 8139–8143.
- [2] Liang, Q.; Brocks, G.; Bieberle-Hüttner, A. Oxygen evolution reaction (OER) mechanism under alkaline and acidic conditions. *J. Phys.* 2021, 3(2), 026001.

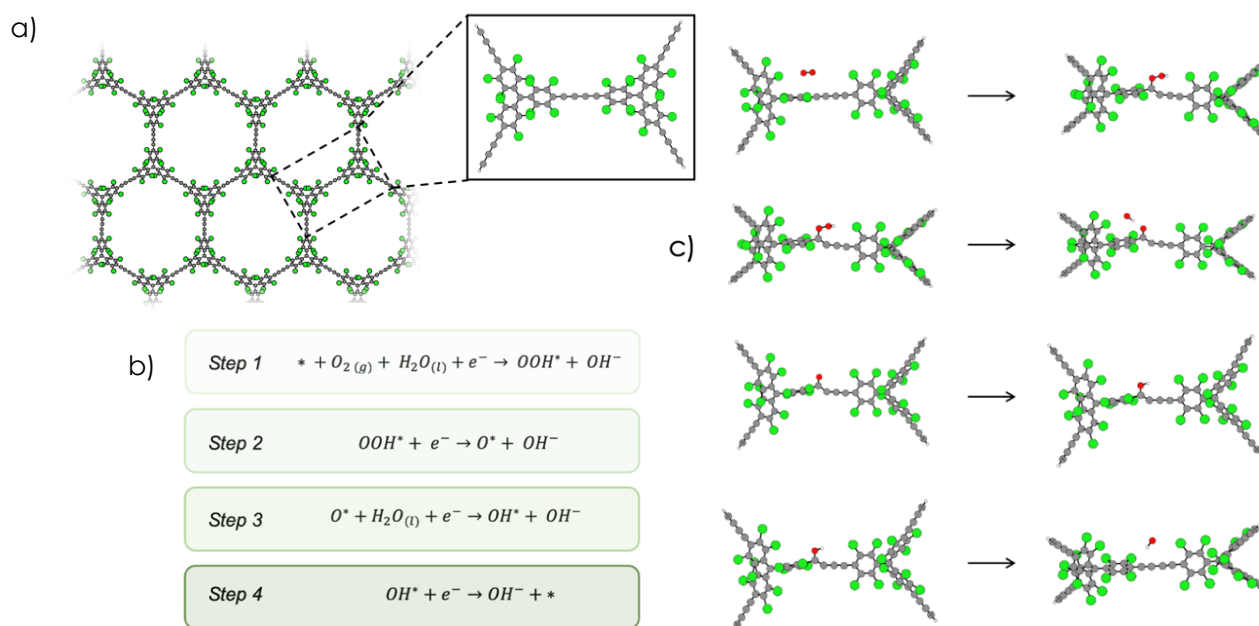


Figure 1: a) Model, b) Steps of ORR, c) ORR intermediates.