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Validating quantum chemical simulations of Oxygen Evolution on Single-atom catalysts

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Single Atom Catalysts (SACs) consist of isolated metal atoms stabilized on a support, maximizing the exposed active sites and showing behaviours that are reminiscent of coordination compounds[1,2] and are now widely investigated for catalytic approaches for the energy transition. Among them, Dual Atom Catalysts (DACs) are emerging as a promising frontier due to the possibility to use two catalytic centres. Starting from experimental results of the dual atom FeCo-N3O3@C[3], of which the structure is known, we investigate the reaction pathways for Oxygen Evolution and Oxygen Reduction Reactions (OER and ORR) with DFT calculations, providing evidence of the actual reaction mechanism which involves unconventional intermediates, that can form on single atoms. Previous studies reported that on single atom catalysts OER does not follow the classical four-electrons step mechanism,[4] as alternative pathways are possible due to the formation of unconventional intermediates *OH*OH, *OH*O and O2*.[5] In the case of DACs the picture is rather more complex due the very high number of possible adsorption configurations. Finally, the ab initio thermodynamic approach was combined with the generalized Butler-Volmer model, in order to include kinetic effects;[6] this permit to select which reaction pathway explains better the experimental data. Figure 1 reports a comparison between calculated and experimental polarization curves and the corresponding ORR pathway (panel b, c). This study provides an example of the potential combination of the structural information with atomistic details, quantum chemical calculations, and activity experiments to understand the complexity of the reactivity of Dual Atom Catalysts.

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