Towards Predictive and Operando Computational Catalysis – Recent Advancements for Transition-Metal Chemistry

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The ultimate goal in computational catalysis is to quantify the factors that govern reactivity and selectivity, enabling the prediction and rational design of superior catalysts. However, the success of such endeavours relies heavily on the accuracy of the chosen computational methodology. Apart from the selection of the electronic structure method, correctly identifying the most stable conformer and considering explicit solute-solvent interactions are crucial for accurately predicting molecular structures, reactivities, and reaction mechanisms.

Yet many current computational studies consider only a single specific conformation, typically derived from experimental data, such as X-ray crystal structure analysis and describe only implicit solvation. This approach starkly contrasts the reality of experimental reaction conditions, where more than one (experimentally derived) conformation can be relevant, and specific solute-solvent interactions can significantly impact reactivity. This discrepancy is particularly problematic when studying transition-metal catalysts, as they can adjust their geometries in response to environmental perturbations.

In my presentation, I will share examples that highlight techniques to enhance the accuracy of computational modelling of transition-metal catalysts. These approaches aim to address explicit solute-solvent interactions [1] and incorporate factors such as conformational diversity,[2-3] counter ions,[4] and reaction dynamics.[5] By integrating these methods, we can significantly improve our ability to predict reactivity and selectivity of reactions catalysed by transition-metal complexes. Ultimately, these techniques contribute to the development of efficient and selective catalysts, a necessity for sustainable processes.

References

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