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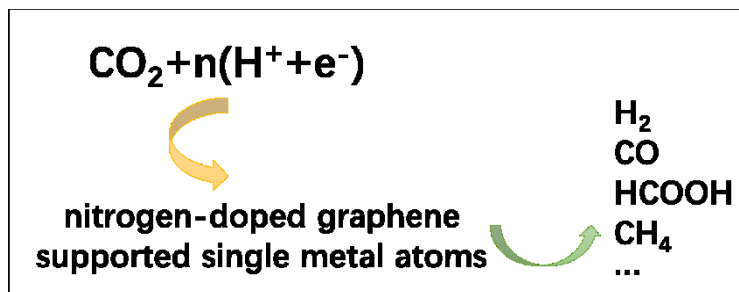
Theoretical investigation of the selectivity of CO₂ electroreduction on different single atom catalysts (SACs)

Aim

To understand of the origin of the selectivity of CO₂ electroreduction on different single metal atoms supported by nitrogen doped graphene using density functional theory (DFT) calculations.

Justification

The conversion of CO₂ into valuable chemical products via electroreduction processes at mild condition is of great significance to global environmental protection and sustainable development. Single atom catalysts have shown potential for CO₂ electroreduction at relatively low potentials with high CO₂ reduction selectivity. The main competitive reaction is the hydrogen evolution reaction (HER). It is suggested that for Fe, Co, Ni, Pd, Bi based SACs, CO₂ is selectively reduced to CO, while on Sn, Sb, In based SACs, CO₂ is mainly reduced to HCOOH and CH₄ is preferred on Zn based SACs. Nitrogen doped carbon materials such as graphene are an important substrate to anchor single metal atoms. To better understand the factors that determine the product selectivity, we will use DFT to systematically investigate the CO₂ electroreduction mechanisms of different SACs.



Program

1. Literature study: (i) investigate CO₂ reduction products such as CO, HCOOH and CH₄. (ii) review structural information of SACs characterized by experiments, (iii) learn how to simulate electroreduction processes via DFT calculations and how to evaluate the accuracy of the calculations.
2. Construct a series of SAC configurations by considering the local coordination environments including the coordination number, coordination atoms and possible long-distance interactions.
3. Calculate the adsorption of reactants (H₂O, CO₂, H₂O etc.) and possible intermediates (*COOH, *HCOO, *CO, *CH_x etc.) on single metal atom sites. Furthermore, carbon sites will be also considered as possible active sites for CO₂RR.
4. Build potential diagrams of CO₂RR and explain product selectivity from a thermodynamic point of view. If the trends do not match experimental observations, more possible SAC configurations, more accurate simulation methods or reaction kinetics should be considered.
5. The origin of the product selectivity will be unravelled from the electronic structure analysis.