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Vacancy Engineering of Nitrogen-Doped Graphene for Stabilizing Iron Single-Atom Catalysts: Unveiling the Balance Between CO_2 Reduction Activity and Stability

The development of efficient and sustainable catalysts is a cornerstone in the global effort to address pressing environmental challenges, particularly the conversion of carbon dioxide (CO_2) into value-added products. Among the emerging strategies, single-atom catalysts (SACs) have garnered significant attention due to their exceptional atom utilization, tunable electronic properties, and potential for high catalytic selectivity [1, 2, 3]. Precise defect engineering in nitrogen-doped graphene (NG) presents a powerful strategy for the design of advanced SACs [4]. Here, we employ spin-polarized density functional theory (DFT) calculations to systematically investigate the stability of graphene vacancies and their role in anchoring iron atoms for carbon dioxide reduction reaction (CO_2RR). Our work identifies key trends linking vacancy type, nitrogen doping patterns, and iron binding energies, revealing that vacancy stabilization is highly dependent on defect size and nitrogen configuration. Beyond thermodynamic considerations, we demonstrate that catalytic activity and stability are not necessarily correlated. Through detailed energy profile analysis of CO_2RR intermediates, we find that the most thermodynamically stable Fe–N arrangements do not always correspond to the lowest activation barriers. Instead, kinetically favorable, less stable Fe–N configurations provide superior catalytic performance. Furthermore, the oxidation state of iron, the local charge environment, and the solvent effect critically modulate both the stability and reactivity of Fe@NG sites. These findings highlight a crucial trade-off between stability and activity in SACs design and opens avenues for the rational development of highly efficient SACs for sustainable energy applications.

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Primary author(s) : LANGER, Rostislav (IT4Innovations, VSB – Technical University of Ostrava)

Presenter(s) : LANGER, Rostislav (IT4Innovations, VSB – Technical University of Ostrava)