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Invited Paper

Ensemble catalyst design from a multi-scale simulation perspective

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ABSTRACT

Ensemble catalysts, composed of a single or a few atoms that make up the active center, have garnered widespread attention and application in the field of catalysis due to their potential for high-efficiency catalytic performance. This is attributed to the systemic exposure of their metal atoms, which allows them to break the linear scaling limit of single-atom catalysts. The coupling of first-principles calculations with micro-kinetics simulations represents an essential approach for multiscale research in materials science. However, the conventional micro-kinetics theory, based on mean-field approximation, struggles to handle surface site distribution and heterogeneity inherent to ensemble catalyst systems.

To tackle this challenge, we have developed a state-to-state micro-kinetics framework that goes beyond the average field. This framework is particularly suited for locally confined catalytic systems. By integrating the isolated compound reaction sites into a linear treatment, this method achieves an equivalent mapping with the higher-order nonlinear full reaction network, overcoming the limitations of traditional kinetics models in effectively recognizing similar surface states and tracking evolving catalyst surface states.

Utilizing this framework, we conducted multiscale analyses that couple Density Functional Theory (DFT) computations with microkinetic simulations for a series of reactions, such as CO oxidation catalyzed by Pdn (n = 1, 2, 3, 4) atom ensembles loaded on Au(111), CO preferential oxidation catalyzed by single/dual-atom transition metal catalysts loaded on CoOx/CeOx, and ammonia escape reactions catalyzed by transition metal alloy cluster catalysts. Not only were we able to elucidate the microscopic reaction mechanisms and obtain simulation results that compare favorably with the experimental Turnover Frequency (TOF) values, but this research also provides effective guidelines for the high-throughput rational design of ensemble catalysts.