A Simple Stochastic Approach to Probe Reaction Dynamics on Individual Nanocatalysts

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Abstract: Recent single-molecule experiments that investigate catalytic properties of individual nanoparticles have provided a large amount of quantitative information. These techniques have allowed researchers to uncover the molecular properties of catalytic systems, which are hidden in the ensemble-averaged bulk measurements.[1-2] The observations stimulated significant theoretical efforts, but the underlying molecular mechanisms are still not properly understood.[3-4] We introduce a simple theoretical method to provide a comprehensive description of the reaction dynamics on catalysts with multiple active sites based on a discrete-state stochastic description.[5] To establish a link between observed experiments and the theory, we explicitly determine the dependency of the number of active sites, number of intermediate species and the topology of underlying chemical reactions on the dynamics of catalyzed chemical reactions.[6] Our theory provides quantitative bounds for realistic dynamic properties of catalytic processes that can be directly applied to analyze the experimental observations. This stochastic approach can also be utilized to probe the dynamics of chemical reactions on individual catalysts that are heterogeneous in nature. Our analytical and numerical calculations are supported by Monte Carlo computer simulations and the proposed theoretical approach successfully clarifies several important aspects of the molecular mechanisms of chemical reactions on catalysts.

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