Electrochemical Realism in Single-Atom Catalysis

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Single-atom catalysts (SACs) represent a powerful and adaptable class of materials that hold great promise across a broad spectrum of catalytic applications, particularly in electrochemistry. By virtue of their atomically dispersed active sites, SACs offer a unique platform for maximizing metal efficiency and precisely tuning catalytic behavior. Furthermore, the structural simplicity of these active sites makes them highly amenable to theoretical modeling.

Yet, one of the fundamental challenges in this field lies in the dynamic nature of the active site under real electrochemical operating conditions. Variations in parameters such as pH and electrode potential can significantly alter the structure and reactivity of these sites, effects that are too often overlooked in theoretical treatments.

In this lecture, we introduce a conceptual and computational framework designed to bridge this gap. By extending the concept of surface Pourbaix diagrams to the realm of SACs, we provide a strategy for assessing their electrochemical stability and predicting their active configurations under working conditions. This approach facilitates a deeper understanding of catalytic function and supports the rational design of SACs with enhanced durability and performance.

While the methodology is broadly applicable to various SAC systems, including those based on metal oxides, carbon materials, or ceramic supports, we will focus on illustrative examples involving two-dimensional materials. Ultimately, incorporating electrochemical stability into the design paradigm offers a crucial step forward in the development of next-generation SACs for energy conversion and other technologically relevant processes.