

First-principles prediction of oxide surface structure and properties in aqueous electrochemical environments

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The design of efficient, stable, and inexpensive catalysts for oxygen evolution and reduction is crucial for the development of electrochemical energy conversion devices such as fuel cells and metal-air batteries. However, such design is often limited by challenges in atomic-scale experimental characterization and computational modeling of solid-liquid interfaces. To address these challenges, we recently proposed a general framework, based on a combination of electronic structure computations and electrochemical principles, for the prediction of oxide surface structure, stoichiometry, stability, and catalytic activity as a function of pH and applied potential. In this talk, we use this approach to demonstrate the critical role of solvated cation species in governing oxide surface composition, and, thereby, surface electronic structure and catalytic activity, in perovskite oxides. We develop environment-structure-property relationships to predict trends in catalyst stability and activity, and show that these relationships can be used to understand experimental observations of surface amorphization and oxygen evolution/reduction activity in perovskite oxide-based catalysts. In addition to providing guidance for catalyst design, our work may lead to new electrochemical approaches for tailoring the properties of complex oxide heterostructures.