Perovskite Electrocatalysts: Oxygen Intercalation, Surface Reactivity and Structural Degradation

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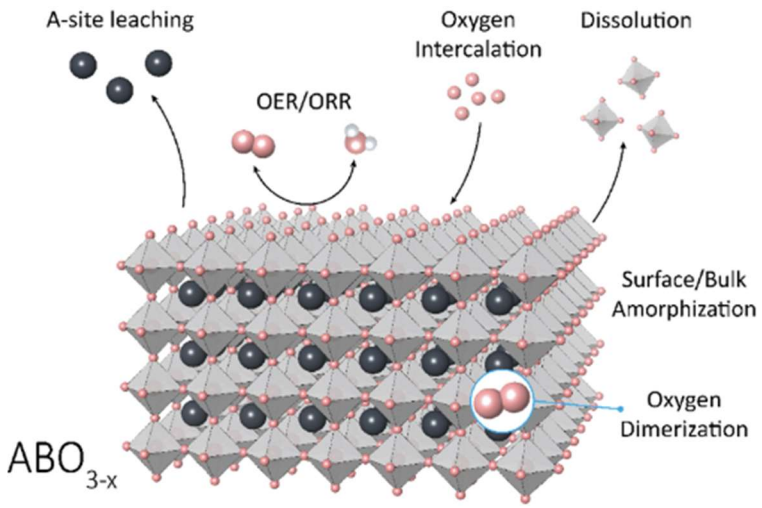

Oxygen evolution and reduction (OER/ORR) are essential reactions in electrocatalysis. Perovskite oxides have emerged as efficient electrocatalysts for these reactions, benefitting from their widely tunable electronic structure. However, structural and chemical transformations of perovskites during electrocatalysis can be rather complex. Such materials undergo A-site cation leaching, surface amorphization, oxygen (de)intercalation, and dissolution. A poor understanding of these processes limits the design of more active and stable electrocatalysts.

In my PhD, I studied oxygen intercalation, surface reactivity and structural degradation for highly active $SrFeO_{3-x}$ and $SrCoO_{3-x}$ electrocatalysts using epitaxial thin films as model materials. First, I will introduce the topotactic phase transformation that takes place in oxygen-deficient perovskites upon oxidation. Second, I will discuss the anomalous ORR reactivity of $SrFeO_{3-x}$ that does not require electrochemistry and the critical role of aqueous media in this reaction. Finally, I will demonstrate how $SrCoO_{3-x}$ forms oxygen molecules inside its crystalline bulk under OER conditions and how this is connected to lattice instability and amorphization in this material. These results can be generalized to other perovskite oxides and provide fundamental insights into their surface reactivity and structural degradation.