

Atomic Regulation of PGM Electrocatalysts for the Oxygen Reduction Reaction

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Perspective

The Oxygen Reduction Reaction (ORR) occurs at the solid/electrolyte interface in a heterogeneous regime during operation. At a particular voltage, the electron can deplete the oxygen in the electrolyte as well as other species. Each intermediate species from the appropriate sub-reaction step has a different sensitivity to the catalytic structure, which affects the micro-kinetics and the mechanism that results.

The binding energy of oxygen species in the adsorption-desorption process plays a central role and affects the ORR kinetics at the most fundamental level. The Sabatier principle shows that the ORR activity connects to the oxygen binding energy following a volcano pattern, implying that the Pt has nearly perfect oxygen binding energy toward the ORR.

To get the best activity, it is usual or rational to optimise the binding energy toward the top of the volcano plot. Modulation of adsorption energy relies on adjusting the catalyst surface's surface electronic structure, or more precisely, tweaking the localised surface atomic structure, and hence increases ORR kinetics. As a result, the local atomic structure and the consequent electronic structure serve as a link between the energy of oxygen binding and catalytic activity. The in-plane atomic arrangement and inter-plane atomic stacking over 1-3 atomic layers at the top-surface and sub-surfaces are usually required to complete a catalytic process. As a result of their surface-sensitive properties, catalysts should not be limited in their rational design to certain nano-morphology or nanostructures.

Through the aforementioned link, the surface structure of the catalyst has a significant impact on ORR kinetics. In particular, the atomic arrangement at the surface and near-surface, in addition to the atomic composition, has a significant impact on the surface electrical structure. Modulating the atomic distance and atomic Coordination Number (CN) are thus theoretically feasible approaches. To induce phase change in most bulk metal materials, a certain temperature or pressure is usually required. At atmospheric pressure, the temperature of the bulk Fe's bcc-fcc phase change is 1183 K. Because surface energy may dominate total energy in nanoscale materials, their crystal structure may differ from that of bulk materials. As a result, phases that may not be stable in bulk materials can exist in nanostructures at the same temperature and pressure. In nature, Au has a fcc structure, however nanocrystal equivalents with 2H, 4H, and 8H phases have been found and synthesised.