Pt-Based Intermetallic Nanostructures: Activity Origin and Multifunctionality for Efficient Electrocatalysis

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Pt-based intermetallic nanostructures have demonstrated superior electrocatalytic performances compared to random alloy structures. However, the origin of their enhanced catalytic properties remains elusive, and a robust synthetic strategy for intermetallic nanostructures represents a challenge. Here, we reveal by combining theoretical and experimental results that the activity enhancement in intermetallic structures for oxygen reduction reaction (ORR) originates from the intensified ligand effect. We prepare well-defined model nanocatalysts *via* confined nanospace-directed synthesis using mesoporous silica templates, which allows control over the size, shape, and atomic ordering of nanostructures. The prepared ordered intermetallic Pt_3Co nanowires (*O*-PtCo NWs) benefitted from the intensified ligand effect, Pt-skin layer, show superior ORR activity and durability to disordered alloy Pt_3Co nanowires and Pt/C catalyst. The *O*-PtCo NWs further exhibited catalytic superiority in hydrogen evolution and methanol oxidation, and demonstrated excellent cell performance in proton exchange membrane fuel cells.