RESEARCH ARTICLE

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Enriched Oxygen Coverage Localized within Ir Atomic Grids for Enhanced Oxygen Evolution Electrocatalysis

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Inefficient active site utilization of oxygen evolution reaction (OER) catalysts have limited the energy efficiency of proton exchange membrane (PEM) water electrolysis. Here, an atomic grid structure is demonstrated composed of high-density Ir sites ($\approx$10 atoms per nm^2) on reactive MnO$_{2-x}$ support which mediates oxygen coverage-enhanced OER process. Experimental characterizations verify the low-valent Mn species with decreased oxygen coordination in MnO$_{2-x}$ exert a pivotal impact in the enriched oxygen coverage on the surface during OER process, and the distributed Ir atomic grids, where highly electrophilic Ir—O$^{(1\cdot\delta)-}$ bonds proceed rapidly, render intense nucleophilic attack of oxygen radicals. Thereby, this metal-support cooperation achieves ultra-low overpotentials of 166 mV at 10 mA cm^{-2} and 283 mV at 500 mA cm^{-2}, together with a striking mass activity which is 380 times higher than commercial IrO$_2$ at 1.53 V. Moreover, its high OER performance also markedly surpasses the commercial Ir black catalyst in PEM electrolyzers with long-term stability.

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1. Introduction

The proton exchange membrane (PEM) water electrolysis offers broad promise for renewable energy conversion and storage.[1–3] However, it remains challenging to meet the ever-increasing demand for high energy efficiency and long-term operation when concerned about the undersupplied precious metals, particularly the benchmark Ir-oxide-based catalysts.[4] It is therefore highly expected to increase catalyst utilization and reduce the precious metal consumption.[5,6]

To develop the activity and durability of the oxygen evolution reaction (OER) catalyst for PEM water electrolysis with minimized precious metal usage, numerous efforts have been dedicated to modifying the interface configuration of precious metal active centers on acid-stable supports.[7–10] It is widely investigated that the geometric structures of the precious metal from highly dispersed atoms to regularly arranged ensembles can result in distinctive electronic structures and a tunable coordination environment.[11–14] Hence, the profound modulation of the interfacial precious metal and the supported materials enables the interaction of multiple active centers with regulable reaction pathways.[15–17] Ideally, the co-catalytic metal-support interactions, which involve the support sites to cooperate with the precious metal traditional active centers, can contribute to even more sufficient site utilization of the catalyst.[18] Actually, very less effective strategies are put forward to achieve the precious metal-support cooperation on an acid-stable OER catalyst. The main challenge is the considerable discrepancy in the adsorption structures of precious metal and support sites due to the unbalanced binding strength of oxygen-related species, which usually leads to separated reactions at each site and remains limited by linear scaling relation between intermediates.[14] In this regard, it can be advantageous to select reactive supports like manganese- and cobalt-based oxides which exhibit intrinsic OER activity among the non-precious metal-based host materials.[19–21]

Recently, MnO$_x$ support-based OER catalysts have received increasing attention. Li et al. reported the significant role of MnO$_2$ support to form hexavalent iridium oxide (IrO$_3$) which was predicted to have the highest activity and stability in acid.[6] Lattice