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Efficient O–O Coupling at Catalytic Interface to Assist Kinetics Optimization on Concerted and Sequential Proton–Electron Transfer for Water Oxidation

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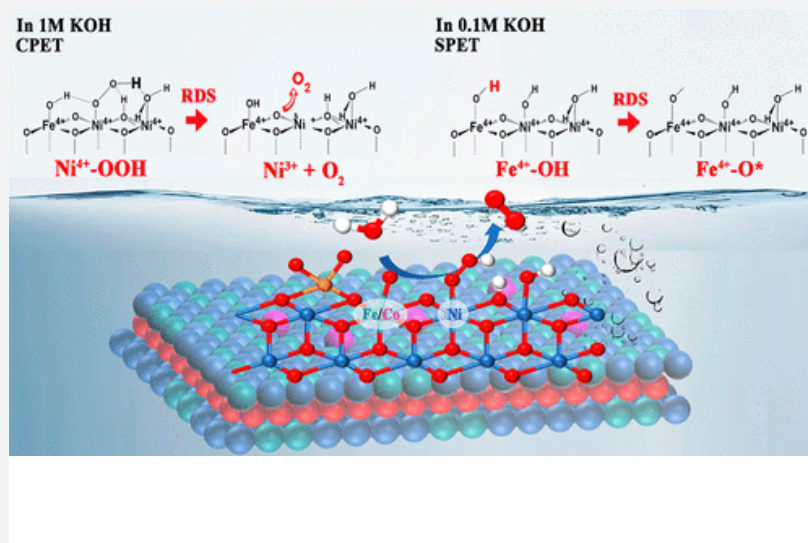
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Abstract



A catalyst kinetics optimization strategy based on tuning active site intermediates adsorption is proposed. Construction of the M-OOH on the catalytic site before the rate-determining step (RDS) is considered a central issue in the strategy, which can optimize the overall catalytic kinetics by avoiding competition from other reaction intermediates on the active site. Herein, the kinetic energy barrier of the O–O coupling for as-prepared sulfated Co-NiFe-LDH nanosheets is significantly reduced, resulting in the formation of M-OOH on the active site at low overpotential, which is directly confirmed by in situ Raman and charge transfer fitting results. Moreover, catalysts constructed from active sites of highly efficient intermediates make a reliable model for studying the mechanism of the OER in proton transfer restriction. In weakly alkaline environments, a sequential proton–electron transfer (SPET) mechanism replaces the concerted proton–electron transfer (CPET) mechanism, and the proton transfer step becomes the RDS; high-speed consumption of reaction intermediates (M-OOH) induces sulfated Co-NiFe-LDH to exhibit excellent kinetics.

KEYWORDS: O–O coupling, intermediates adsorption, concerted proton–electron transfer ▾

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