Manipulating dynamic surface transformation of electrocatalysts for superior water oxidation

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Rationally manipulating the in-situ formed catalytically-active surface of catalysts remains a tremendous challenge for highly-efficient water electrolysis. Here we present a cationic redox-tuning method to redirect dynamic surface restructuring from the same parent structure, for electrochemical oxygen evolution reaction (OER). Partially replacing oxygen of the layered transition metal oxides with halide modified redox transitions during the OER, which was monitored by operando X-ray absorption spectroscopy. Surface-restructured transition metal oxides showed an OER activity approximately two orders of magnitude higher than that of benchmark RuO_2 and sustained this high activity for 500 h without degradation, making it among the best OER electrocatalysts ever reported. This work makes a stride in modulating surface restructuring and opens up new opportunities to design superior OER electrocatalysts via tuning cationic redox.