**Enhancement of Electrocatalytic Activity by Surface Dopant**

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Heterogeneous electrochemical processes within energy conversion devices have been one of the intense focuses. The surface atomic structures of the electrocatalysts (noble metal materials as current bench mark) play determining roles in energy conversion efficiency. To enhance the catalytic activities multiple modification strategies, such as facet engineering, defect introduction, doping have been proposed. Atomic doping has been widely used for optimal performance however bulk doping approach has been generally adopted which might not able to utilize the accessible surface active sites. Also, the actual roles the dopant element play in the electrochemical processes has not been fully explored.

Herein a robustness of a vapor-phase hydrothermal (VPH) method is introduced as a generic surface doping method to unlock the electrochemical activities of metal (hydro)oxide materials, one of the most abundant group of materials on earth, towards some important electrochemical reactions such as triiodide reduction reaction, hydrogen evolution reaction. The performance of the devices (dye-sensitized solar cells, water electrolyzer) equipped with the modified metal (hydro)oxide materials as catalysts demonstrates excellent performance similar to the one with bench marking noble metal catalysts. Through the combination of ex-situ, in-situ characterizations and theoretical calculation, we acquire the mechanistic information of the interfacial electrochemical reactions and rationalize the enhanced electrochemical performance.