

Interaction Mediator Assisted Synthesis of Mesoporous Molybdenum Carbide: Mo-Valence State Adjustment for Optimizing Hydrogen Evolution

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To overcome inherent limitations of Mo_xC for hydrogen evolution reaction (HER), i.e., low density of active site and nonideal hydrogen binding strength, we report synthesis of valence-controlled mesoporous Mo_xC as a highly efficient HER electrocatalyst. The synthesis procedure uses an interaction mediator (IM), which significantly increases the density of active site by mediating interaction between PEO-*b*-PS template and Mo source. The valence state of Mo is tuned by systematic control of the environment around Mo, by controlled thermal activation under air. Theoretical calculations reveal that the hydrogen binding is strongly influenced by Mo valence. Consequently, Mo_xC achieves a significant increase in HER activity (exceeding that of Pt/C at high current density $\sim 35 \text{ mA/cm}^2$ in alkaline solution). In addition, a volcano-type correlation between HER activity and Mo valence is identified for the first time using experimental parameters which reflected the Mo valence state. The present strategies can be applied to various carbide and Mo-based catalysts, and the established Mo valence and HER relations can guide development of highly active HER electrocatalysts.