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Boosting acetylene hydrochlorination over high-valent Cu(δ+) sites via tuning axial oxygen coordination

Abstract:

Transition metal sites with high valence states can accelerate the reaction kinetics to offer highly intrinsic activity but suffer from thermodynamic formation barrier. Herein, we present a general two-step annealing strategy to fabricate a series of hollow carbon-based high-valent Cu(δ+) single atom sites featuring Cu-N4 moieties simultaneously modulated with axial O coordination (designated as Cu-NOC). Systematic analyses demonstrate that the synergetic effects between the -N4 species in the first coordination shell and the O atoms in axial coordination led to robust discrete Cu(δ+) sites with the optimal electronic structure for the hydrochlorination of acetylene. Remarkably, the high-valent Cu-N4 moiety with axial O coordination possesses a strong interaction with HCl and partial dissociative chemisorption C2H2 that endow their superior catalytic performance compared to Cu(I)/Cu(II) counterparts, which favors the formation and desorption of VCM on Cu sites. This is the first study demonstrating that high-valent Cu(δ+) used in heterogeneous catalysis, which enlightens us to design stable, and high-performance metal catalysts.

Biography

Jia Zhao is received his PhD in Industrial Catalysis in 2015 from the Zhejiang University of Technology (ZJUT), where his thesis focused on the development of metal-based catalysts for acetylene hydrochlorination. After a Postdoc at the ZJUT (2015–2017), he accepted a position at the ZJUT and now he was a professor at ZJUT. His current research field is catalysis, covering areas of synthesis, characterisation and reactivity in hydrogenation and oxidation reactions. He has (co)authored over 90 publications in peer reviewed journals.