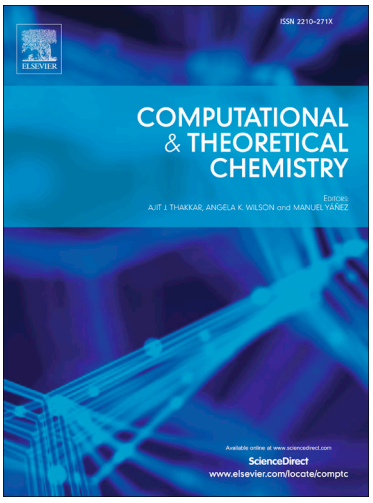

Novel nitrogen-doped Au-embedded graphene single-atom catalysts for acetylene hydrochlorination: a density functional theory study

Wanqi Gong, Fei Zhao, Lihua Kang

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Novel Nitrogen-doped Au-embedded Graphene Single-Atom Catalysts for Acetylene Hydrochlorination: A Density Functional Theory Study

Wanqi Gong [1], Fei Zhao [1], Lihua Kang [1,2,*]

[1] School of Chemistry and Chemical Engineering of Shihezi University, Shihezi 832000, Xinjiang, China

[2] Key Laboratory for Green Processing of Chemical Engineering of Xinjiang Bingtuan, Shihezi 832000, Xinjiang, China

*Corresponding author E-mail: kanglihua@shzu.edu.cn;

Tel:+86-0993-2057213; Fax: +86-0993-2057270

Abstract: The mechanisms of an Au-embedded graphene (AuG) single-atom catalyst (SAC) and AuG-SACs doped with nitrogen at different locations for acetylene hydrochlorination were investigated through density functional theory (DFT). The density functional dispersion correction was calculated with the DFT-D3 method. We studied the adsorption characteristics of HCl and C_2H_2 on these SACs and simulated the corresponding reaction mechanism. We also found that adding the heteroatom N to AuG-SACs can reduce the reaction activation energy. Furthermore, we found the optimal location of the N atom of the N–substituted SAC to reduce the activation energy. The results suggest that N-doped AuG-SACs could find potential in catalyzing acetylene hydrochlorination to vinyl chloride and reduce the amount of noble metal used in mercury-free catalysts.

Keywords: Au-graphene SACs, DFT-D3 calculation, Acetylene hydrochlorination, Nitrogen-doped graphene

1. Introduction

Single-atom catalysts (SACs) can take full advantage of every metal atom in order to maximize their catalytic efficiency. Supported single metal atoms, as the active sites of a chemical reaction, can enhance the selectivity, activity, and stability of heterogeneous catalysts and thus have potential for application in a great variety of industrial chemical reactions. Kwak et al. [1] coordinated unsaturated Al^{3+} ions in anchoring a catalytically active phase (Pt) to aγ-Al_2O_3 support and obtained a noble metal single catalyst. Qiao et al. [2] synthesized a Pt SAC consisting of only isolated single Pt atoms anchored to the surfaces of iron oxide nanocrystallites. Experiments demonstrate that SACs can promote the catalytic performance in a variety of heterogeneous reactions. [3-12] So far, there is no theoretical study on the catalytic mechanism of a SAC in the hydroxylation of acetylene.