Nitrogen-doped carbon nanomaterials as a potential metal-free catalyst for CO oxidation

I-Hsiang Lin, Yu-Huan Lu, and Hsin-Tsung Chen*

Department of Chemistry, Chung Yuan Christian University, Chungli District, Taoyuan City, 32023, Taiwan

We elucidate the possibility of nitrogen-doped carbon nanomaterials as a robust catalyst for CO oxidation. We carry out first-principles calculation with considering the spin-polarization effect to demonstrate the reaction of CO oxidation catalyzed by nitrogen-doped carbon nanomaterials. The calculations show that O_2 species can be partially reduced with charge transfer from the nitrogen-doped carbon nanomaterials and directly chemisorbed on the C–N sites of the nitrogen-doped carbon nanomaterials. The partially reduced O_2 species at the C–N sites can further directly react with CO molecule via the Eley-Rideal mechanism with the barriers of 0.20 ~ 0.58 eV. The ab initio molecular dynamics (AIMD) simulation is performed and evidences that the oxidation of CO takes place by CO reacting directly with the partially reduced O_2 species on the nitrogen-doped carbon nanomaterials via the Eley-Rideal mechanism. The relationship between the curvature and reactivity of the nitrogen doped carbon nanomaterials are also unraveled. It appears that the barriers height of the rate-limiting step depends on the curvature of the nitrogen doped carbon nanomaterials in the trend of N-doped C_{60} < (3, 3)-NCNT < (4, 4)-NCNT < (5, 5)-NCNT < N-doped graphene (decreases with the increased curvature). Using this relationship, we can predict the barriers for other NCNTs with different tube diameter. Our results reveal that the nitrogen doped carbon nanomaterials can be a good, low-cost, and metal-free catalyst for CO oxidation.

References:

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Email: htchen@cycu.edu.tw