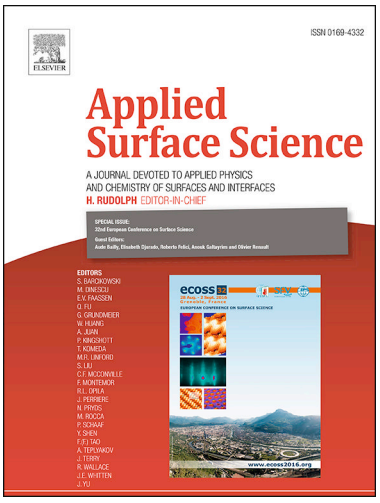

Detailed Mechanism of the NO + CO Reaction on Rh(100) and Rh(111): A First-Principles Study

Lu Tan, Liangliang Huang, Yingchun Liu, Qi Wang

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Detailed Mechanism of the NO + CO Reaction on Rh(100) and Rh(111): A First-Principles Study

Lu Tan[a], Liangliang Huang[b], Yingchun Liu[a,*], Qi Wang[a,*]

[a] *Department of Chemistry, Zhejiang University, Hangzhou 310027, People's Republic of China*

[b] *School of Chemical, Biological & Materials Engineering, University of Oklahoma, Norman, Oklahoma 73019, United States*

Abstract

Through DFT calculations, the detailed mechanism of the catalytic NO + CO reaction, a prototypical system with great practical applications especially in the automobile-exhaust aftertreatment, was determined on Rh(100) and Rh(111). The elementary steps and their energy evolution were revealed. These steps include NO dissociation, N_2 formation through N recombination, CO_2 formation, and N_2O formation, transformation, and dissociation. The reaction steps of NO_2 formation and direct reaction between NO and CO were also studied, and were verified to be relatively insignificant in this reaction system. Results shed light on the atomic-level origin why Rh(100) is more active for this reaction system and more selective for the production of N_2 versus N_2O compared with Rh(111). Meanwhile, the preference between the two routes for N_2 production, i.e., N atoms recombination and N_2O as intermediate, was found to be dependent on the distribution of surface species and the interaction among them intricately. This work provides a basis for further kinetic modeling to investigate the catalytic properties on a realistic scale.

Keywords: Nitrogen oxides, Carbon monoxide, Rhodium, Density functional theory, Reaction mechanism, Catalysis

1. Introduction

Heterogeneous catalysis plays an important role in the production of chemicals and the removal of pollutants in our modern society. Among them, the catalytic reduction of nitrogen oxides (NO_x) is a key reaction for air pollution control [1, 2]. In the automobile industry, the so-called three-way catalyst, which always contains Pt, Rh, and Pd as active ingredients, has been satisfactorily utilized for this purpose for decades. In this system, carbon monoxide

*Corresponding author at: Department of Chemistry, Zhejiang University, Hangzhou 310027, People's Republic of China

Email addresses: liuyingch@zju.edu.cn (Yingchun Liu), qiwang@zju.edu.cn (Qi Wang)