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Selective Oxidation of Ammonia on RuO₂(110): a combined DFT and KMC study¹ SAMPYO HONG, Univ. of Central Florida, ALTAF KARIM, Brookhaven National Laboratory, SERGEY STOLBOV, TALAT RAHMAN, Univ. of Central Florida — Motivated by the experiments of Wang et al [1] on the selective oxidation of ammonia on RuO₂(110), we have performed first principles electronic structure calculations based on the density functional theory (DFT) and the pseudopotential method to calculate the activation energy for the associated reaction processes, and used the DFT results in kinetic Monte Carlo (KMC) simulations of the reaction rates. We find the overall energy barriers for $\text{NH}_3 + \text{O} \rightarrow \text{N} + \text{H}_2\text{O}$, $\text{N} + \text{N} \rightarrow \text{N}_2$, and $\text{N} + \text{O} \rightarrow \text{NO}$ to be about 1.0, 0.6, and 0.56 eV, respectively. We also find, in agreement with experiment, intermediate products such as NH to be short-lived (i.e. not detectable in experiment). For a simple model of the surface dominated by the two end reactions above, our KMC simulations show indeed almost 100% selectivity toward NO, in excellent agreement with experimental findings. For more realistic simulation, in which we include several intermediates and their reactions (over 20 processes), KMC simulations show about 60% selectivity toward NO within the experimental O₂ pressure range. We compare our calculated reaction rates to those obtained in experiment and rationalize our results through details of the surface electronic structure. [1] Y. Wang, K. Jacobi, W.-D. Schoene, and G. Ertl, J. Phys. Chem. B 2005, 109, 7883-7893.

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Sampyo Hong
likedew@physics.ucf.edu
University of Central Florida

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