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Error propagation in first-principles kinetic Monte Carlo simulation

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Abstract

First-principles kinetic Monte Carlo models allow for the modeling of catalytic surfaces with predictive quality. This comes at the price of non-negligible errors induced by the underlying approximate density functional calculation. On the example of CO oxidation on $RuO_2(110)$, we demonstrate a novel, efficient approach to global sensitivity analysis, with which we address the error propagation in these multiscale models. We find, that we can still derive the most important atomistic factors for reactivity, albeit the errors in the simulation results are sizable. The presented approach might also be applied in the hierarchical model construction or computational catalyst screening.

Keywords: stochastic simulation, sensitivity and uncertainty analysis, Quasi Monte Carlo, heterogeneous catalysis, first-principles kinetic Monte Carlo

2010 MSC: 65C05, 65C40, 82C99, 62-07

First-principles kinetic Monte Carlo (1p-kMC) have become popular for modeling the chemical kinetics in heterogeneous catalysis[1]. This popularity results from the predictive quality of the approach, enabling an understanding of the detailed surface chemistry without the possible masking by the fitting of parameters. In 1p-kMC, the mechanism and corresponding energy barriers are instead obtained from first-principles electronic structure methods and kinetic Monte

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