

# Scaling-relation-based kinetic Monte Carlo modelling of syngas reactions on stepped metals

Mie Andersen<sup>1</sup>

<sup>1</sup>Theoretical Chemistry, Technische Universität München,  
Lichtenbergstr. 4, 85747 Garching, Germany  
mie.andersen@ch.tum.de

The complexity of catalytic reactions on surfaces calls for efficient means of estimating adsorption energies and reaction barriers, which are required input for a microkinetic model. One commonly used approach is to employ scaling relations [1], which are linear relations between the adsorption energies of atomic and molecular species on various (typically metal) surfaces, in combination with simple rate equations for the catalytic activity based on the mean-field approximation (MFA). In my talk, I will demonstrate the prospects of instead combining scaling relations with kinetic Monte Carlo simulations, which allow for predicting the catalytic activity while taking into account spatial correlations in the distribution of the adsorbates on the surface. For the specific case of methane synthesis from syngas on stepped metal surfaces, I demonstrate that MFA models may overestimate the catalytic activity of metal catalysts by several orders of magnitude due to the neglect of spatial correlations [2] (see Fig. 1). Finally, I will discuss recent and ongoing work aimed at (i) overcoming challenges related to a large disparity in the timescales of the surface processes, (ii) accounting for lateral interactions between the adsorbates, and (iii) dealing with more complex reaction networks such as the synthesis of higher alcohols on metal catalysts.

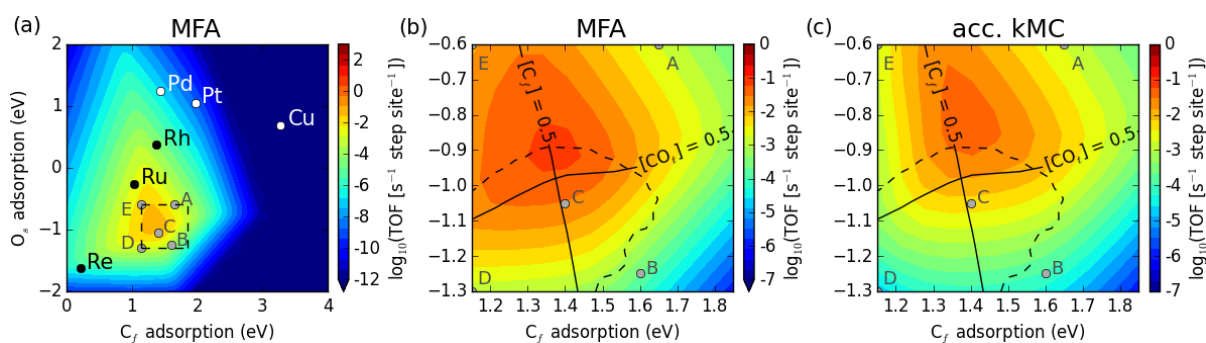


Figure 1. Theoretical activity maps [2] for the formation of methane as a function of the  $C_f$  and  $O_s$  adsorption energies to various stepped metal (211) surfaces as indicated in white or black in (a). The map in (b) is a zoom of the region marked with a dashed line in (a) calculated in the MFA, while the map in (c) is the corresponding region calculated using accelerated KMC. In (b) and (c), the solid lines indicate the contour line where the coverage of C on the f site (vertical line) or the coverage of CO on the t site (horizontal line) is equal to 0.5, while the dashed line indicates the contour line where the degree of rate control for the  $O_s$  hydrogenation step is equal to 0.08 as calculated in the MFA. Reaction conditions are 523 K and 1 bar with a gas composition of 1% CO, 97%  $H_2$ , 1%  $CH_4$  and 1%  $H_2O$ .

## References:

[1] F. Abild-Pedersen, Catal. Today **272**, 6 (2016).

[2] M. Andersen, C. Plaisance and K. Reuter, J. Chem. Phys. **147**, 152705 (2017).