



Vortragsankündigung

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Es spricht: **Prof. Dr. S. Ted Oyama**, Virginia Polytechnic Institute and State University, Department of Chemical Engineering, Blacksburg, USA

Zeit: **Mittwoch, 13. Mai 2009 17:15 Uhr**

Ort: **TU Berlin
Institut für Chemie, Altes Chemiegebäude
Straße des 17. Juni 115, 10623 Berlin
Raum C 243**

Thema: **A General Method for Determining the Role of Spectroscopically Observed Species in Reaction Mechanisms:
Analysis of Coverage Transients (ACT)**

Abstract: A kinetic method is described to determine the role of adsorbed intermediates in heterogeneous reaction mechanisms. The method, denoted as analysis of coverage transients (ACT), involves comparing the time response of a spectroscopically-observed species in an inert gas and a reactive gas to differentiate between adsorption-desorption processes and reaction. The TCA method has potential as it can be used with any type of spectroscopy, as long as the surface coverage can be calibrated. The method is applied in kinetic studies of pyridine hydrodenitrogenation (HDN) and thiophene hydrodesulfurization (HDS) on supported nickel phosphide (Ni_2P) catalysts. Nickel phosphide has emerged as a promising catalyst for hydroprocessing with activity higher than commercial catalysts. The ACT method is applied using Fourier transform infrared (FTIR) spectroscopy and x-ray absorption near edge structure (XANES), both at in situ conditions.

For the pyridine reaction, FTIR revealed the formation of a pyridinium surface intermediate. The coverage of the intermediate increased with pyridine partial pressure and decreased in the presence of hydrogen, suggesting that it was a reaction intermediate. However, transient and steady-state kinetic measurements showed that the rate of reaction of the species did not correspond to the overall reaction rate, and it is concluded that the pyridinium intermediate is not directly involved in the HDN reaction. The studies demonstrate that mere observation of an adsorbed surface species at reaction conditions is not sufficient to prove that it is a reaction intermediate.

In the reaction of thiophene, FTIR showed the presence of a tetrahydrothiophene surface intermediate. XANES measurements showed corresponding growth of a signal attributed to Ni-S interactions. Transient XANES measurements showed that the tetrahydrothiophene species responded dynamically at the same rate as the steady-state rate, indicating that it was an intermediate in the reaction. Thus, the ACT method has the ability of distinguishing spectators from true reactive species observed spectroscopically at reaction conditions.

Organisator: Prof. Dr. H.-J. Freund (Fritz-Haber-Institut der MPG Berlin)

Gäste sind herzlich willkommen!

Prof. Dr. Matthias Driß
Sprecher des Exzellenz-Clusters UniCat