41st Annual Robert S. Mulliken Lecture

Chemical Kinetics in Multiphase Chemical Transformation

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(Room 1218)

Abstract

The first part of the talk describes experimental studies of molecular-weight growth pathways leading to PAH formation under combustion conditions, focusing on resonantly stabilized radicals, strained aromatic intermediates, and radical–radical chain-propagating ("well-skipping") chemistry. Three complementary studies highlight the importance of five-membered-ring motifs in curved aromatic growth. We show that cyclopentadienyl (C_5H_5) + propargyl (C_3H_3) predominantly forms dihydropentalenes (DHPs) rather than styrene (C_8H_8) via ring-enlargement/cyclization pathways; that o-benzyne (C_6H_4) + propargyl (C_3H_3) forms the indenyl radical (C_9H_7) through a barrierless route; and that phenyl (C_6H_5) + propargyl efficiently yields indenyl-type products through well-skipping propagation channels at low pressure and high temperature. These investigations combine mass-selected threshold photoelectron spectroscopy (ms-TPES), molecular-beam mass spectrometry, microreactor pyrolysis, and theory to resolve pathways and product branching.

The second part focuses on complex heterogeneous catalytic reaction networks. We highlight (i) near-surface molecular-beam mass spectrometry studies of methanol partial oxidation near atmospheric pressure, including detection of methoxymethanol (CH_3OCH_2OH) above Pd, AuxPdy , and oxide-supported Pd with clear temperature and feed-ratio dependencies, and (ii) thermocatalytic CO_2 reduction on structurally distinct Ru/TiO2 catalysts that exhibit different selectivities (methanation versus CO formation via reverse water–gas shift). We also describe a newly developed time-resolved pulsed-reactant approach that tracks the full effluent composition and, when applied to ethane dehydroaromatization over Pt/HZSM-5, captures transient intermediates (e.g., 1,3-butadiene and cyclopentadiene), identifies distinct methane/benzene formation pathways, and reveals pathway-specific catalyst deactivation.

The last part highlights how non-equilibrium plasma initiates chemical conversion via charged species, radicals, and excited states. We discuss examples from plasma-assisted methane dry reforming and siloxane decomposition and clustering.

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Nils Hansen is a physical chemist at the Combustion Research Facility of the Sandia National Laboratories in Livermore, California, United States. He received his Ph.D. in Physical Chemistry under the supervision of Prof. F. Temps at the Christian-Albrechts-Universität Kiel, Germany (2000). Before joining Sandia in 2004, he worked with Prof. A. M. Wodtke as a postdoctoral researcher at UC Santa Barbara and as a staff member at the BASF AG, Ludwigshafen (Germany). Hansen's research focuses on gas-phase chemistry that is relevant for energy conversion processes. In his work, he unravels the fundamental chemistry of low-temperature oxidation and of aromatics formation in incomplete combustion processes, of gas-surface interactions in heterogeneous catalysis, and of plasma-assisted chemistry. He was elected a "Fellow of the Combustion Institute" in 2019, a "Helmholtz International Fellow" in 2020, and he received the "Wilhelm Jost Memorial Medal" in 2024.