Catalysis and Reaction Engineering: Sites, Pellets, and Reactors

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Abstract

The performance of catalysts during their industrial practice depends on processes occurring at time and length scales involved in making and breaking chemical bonds at active sites, in diffusing molecules and energy within porous pellets, and in interphase and intraphase contacting within reactor vessels. The understanding and control of these processes and of their inherent coupling have benefited from advances in experimental and theoretical methods designed to observe and interpret these phenomena with increasing accuracy, fidelity, and specificity. This presentation attempts to illustrate some of these advances using examples relevant to the refining, chemicals, and energy sectors within which many of the advances emerged and found their seminal application.

At the level of active sites within complex solids, we aim to describe how chemical reactions occur in response to local concentrations and temperatures, controlled in turn by the contacting imposed by pellets and reactors, and to structural and compositional features of catalytically-relevant inorganic structures. Structure-function relations with significant predictive rigor and the evolution of strategies for selecting and discriminating relevant elementary steps and species in complex reaction networks represent two of the essential enabling components in the remarkable progress evident within the last few decades. In particular, the structure and reactive properties of active materials are becoming more evident at the level of atomic connectivities and electronic configurations, as a result of spectroscopic and kinetic methods with greater specificity and time resolution and of theoretical tools essential for rigorous interpretations of the evidence from experimental methods. For complex mixtures and reactions, treatments have evolved from those requiring extreme lumping of species and intuitive selection of their most relevant pathways to modern methods capable of enumerating elementary reactions, assessing their contributions to overall rates and selectivities, and discriminating among mechanistic choices. Molecular speciation and theoretical methods with unprecedented accuracy and sensitivity and assessments of relevance using algorithms of increasing rigor and sophistication have been essential to recent progress.

Improvements in our descriptions of the structure of porous solids and of the dynamics of diffusion within them reflect concurrent advances in the fidelity of geometric descriptions, the accuracy of relevant molecule-surface interactions, and in the assessment of the chemical consequences of confinement for catalytic events at active sites from first-principles. Imaging of complex mesoporous structures and realistic models of their geometry, simulations of transport for interacting molecules within constrained micropores, transient methods for assessing the dynamics of transport in realistic or model solids, and theoretical treatments of the intrinsic coupling between constrained motion and chemical

reactivity in the context of shape selectivity will be used to illustrate the pace and consequences of progress in this area. Finally, descriptions of contacting in vessels with multiple phases and complex hydrodynamics will be briefly examined in the context of visualization and simulation methods that include increasingly specific details about contacting and mixing and of codes that incorporate these details while benefiting from algorithmic improvements and higher computational speed.