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Preface to special issue of catalysis today on modeling catalysis

Catalysis remains central to almost every aspect of the modern chemical processing industry. Despite the thousands of man and woman years that have been devoted by our society to the development and optimization of practical catalysts, improvements in well-known processes and the creation of entirely new catalytic processes remain possible and highly desirable. One key ingredient in continuing efforts to pursue these improvements is progress in modeling of catalysis. The purpose of this special issue is to provide a snapshot of the state of the art in modeling catalysis, particularly heterogeneous catalysis.

Continued improvement in computational hardware and, perhaps more importantly, widespread access to powerful quantum chemistry software packages has led to what might be termed a renaissance in mechanistic modeling of heterogeneous catalysis. The first two papers in this issue provide overviews of current thrusts in these areas. The paper by Stampfl reviews the rapidly developing area of ab initio thermodynamics, while the paper by Illas et al. reviews the capabilities of modern Density Functional Theory methods and prospects for future developments. The papers by Nelson et al., Sorescu, Kitchin et al., James et al., Hwang et al., and Willock et al. provide examples of applying these Density Functional Theory methods to surface-related processes that control many practical catalysts.

Not all heterogeneous catalysis takes place on what can loosely be thought of as two-dimensional surfaces. Zeolites receive very wide use as industrial catalysts and in these materials catalysis takes place in extremely confined geometries. Using detailed modeling methods to understand the complex relationships between zeolite structure and catalysis is a challenging area in which significant advances have been made. The two contributions from Trout et al. and Thomson et al. describe recent efforts in this arena.

Detailed computational modeling is being pursued not only of problems motivated by heterogeneous catalysis but by issues stemming from homogeneous catalysis. The papers by Larsen et al. and Neurock et al. represent two contributions from this broad area, providing quantum chemistry based analysis of EPR spectroscopy of transition metal complexes and the acidity of Keggin-type heteropolyacids, respectively.

Quantum chemistry approaches to catalysis still face severe limitations, simply because the number of atoms that can be accurately treated is miniscule by macroscopic standards. Even a cursory consideration of a practical catalytic process soon leads to the conclusion that many factors other than the highly localized information on structure and mechanism available through quantum chemistry are of vital importance. The remaining contributions to this issue address some of the issues that arise in this context. The paper by Balbuena et al. examines at an atomic-scale the structure of metal nanoclusters deposited on a graphite support, a problem where full scale quantum chemistry approaches are not feasible yet atomic detail is necessary. The paper by Gellman examines what is really measured by a common surface science technique, Temperature Programmed Desorption, when it is applied to molecules with many degrees of freedom. Vlachos et al. give an example of how well-constructed modeling methods can greatly simplify the task of understanding complex multistep catalytic processes. The final paper in the issue, by Krishna and Baur, shows that by building meaningful models of the combined effects of molecular transport and catalysis, one can begin to explore new modes of effective operation for well-known materials.

It is a pleasure to acknowledge the time and energy devoted by the authors and reviewers who have contributed to this special issue. It is my hope that this issue will be useful to the experimental community as an overview of current trends in modeling catalysis and to the modeling community as an invitation to explore the full breadth of topics that catalysis continues to provide.

David S. Sholl*
Department of Chemical Engineering, Carnegie Mellon University, Pittsburgh, PA 15213, USA
*Tel.: +1 412 268 4207; fax: +1 412 268 7139
E-mail address: sholl@andrew.cmu.edu

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