Editorial

The hardest challenge facing humanity in the years to come is to build a sustainable society. That is to say, we must find ways to be able to meet our current needs without consuming the resources that will be needed by our descendents in years to come. Currently, we do not have the technology or insight to do this, but crucially, we know we have only a limited span of time in which to find a solution. One major task is to find energy-efficient and selective methods for producing the wide array of molecules and materials that are needed for industry, health, agriculture, etc., and for research in these areas. We must learn to utilize renewable resources in an economic manner, while at the same time minimizing the extent to which we pollute our environment. This is the concept of 'sustainable chemical production' and catalysis and thus Chemists and Chemical Engineers will play key roles in its realization.

By their very nature, catalysts reduce the energy cost for specific transformation. An ideal catalyst will also allow a selective transformation that gives only the desired materials without production of waste. In the future sustainable society, we will design catalytic reactions for production of most of our needed substances. However, our knowledge of catalysis is not yet sufficiently advanced to allow *a priori* design of desired reactivity. This is where computational modeling and prediction comes in to play. This '*Computational Catalysis*' Special Issue of the Journal focuses on the application of computer modeling to problems in catalysis. As already alluded to, the development of our understanding of catalysis may be a key component to the survival of the human race in the future.

The basis for our knowledge about catalysis, or even about chemical reactions in general, is the mechanism. By knowing how each step of the reaction responds to changes in the reaction conditions, we can predict the result and thereby design the process for a desired outcome. Traditionally, the elucidation of reaction mechanisms has been achieved by experimental techniques, and in particular by kinetic and spectroscopic measurements. The resulting data is always fitted to a model. A century ago, this was exclusively done by hand, by setting up mathematical expressions for how the reaction should behave and fitting the observed data to this relationship. In recent years, the analysis has been strongly augmented by computer modeling, so much so that we can talk about a qualitative change in how we analyze chemical reactions. Today, we can follow the detailed movement of each molecular fragment through each step of a reaction, with an accuracy measured in tenths or even hundredths of an Angstrom. We can evaluate alternative paths, determine which fits best to the available observations, and predict the response to changes in reaction conditions. We can also predict spectroscopic fingerprints of fleeting intermediates, giving unprecedented possibilities for accurate analysis of reaction mixtures. Modeling is without a doubt a very impor-

tant tool for understanding reactions, and thus a key for building the knowledge base we need to design our future sustainable processes.

Catalysis comes in three main flavors: heterogeneous, homogeneous, and enzymatic, each with its own special requirements and techniques. From a computational viewpoint, homogeneous catalysis offers certain advantages. The active catalyst is a single, rather small molecular species, either a metal complex or an organic molecule, allowing application of fairly high levels of theory. The computational methods can be validated, either by spectroscopic studies of relevant structures in the reaction mixture, or by rationalizing the effect of small and well-defined structural changes in the catalyst. In the current issue, the field of computational homogeneous catalysis is illustrated by applications and reviews from leaders in the field.

For more than a decade, the workhorse in computational catalysis has been density functional theory, DFT. This accurate yet relatively fast method allows reaction paths to be determined for systems with more than a hundred atoms. More than half of the papers in the current issue concern DFT studies of reaction mechanisms, ranging over many of the most important reaction types in catalysis. Oxidations and reductions are ubiquitous in organic synthesis. Ahlquist has looked at Ir-catalyzed hydrogenation of CO_2, a process with potential for recycling of carbon dioxide into energy carrier materials; Wiest and coworkers have studied the application of Rh-catalyzed hydrogenation to acrylamide substrates; and Hall and coworkers have investigated oxygen atom transfer catalyzed by Mo.

For complexity-building in organic synthesis, the most important steps are formation of new C–C or C–N bonds. Several examples of investigations of this type of processes can be found here. Rh-catalyzed addition of aryl moieties to aldehydes is studied by de Bruin and coworkers. Houk and coworkers provide revision and extension of earlier studies into proline-catalyzed aldol reactions. Ligand effects in the Suzuki-Miyaura cross-coupling are elucidated by Harvey and coworkers. The effect of a bulky tBu_3P ligand on a Buchwald–Hartwig coupling has been studied by Fey and coworkers. Szabo investigates Pd^{IV} intermediates in couplings of diaryl iodonium salts catalyzed by pincer complexes, whereas Jensen and coworkers have studied Ru-catalyzed polymerization. Poater and Cavallo follow up on earlier studies of metathesis, and have identified a potentially problematic catalyst deactivation.

A critical component of the continuous development of computational tools is evaluation of existing methods in different contexts. Truhlar and coworkers studied the performance of several computational methods in describing the bonding of alkenes to Pd and Pt. Various models for treating the solvent in investigations of Pt-catalyzed hydroamination have been evaluated by Dub and

Poli, whereas Nyhlén and Privalov have assessed the performance of several computational methods for imine hydrogenation.

For many types of problems, it is necessary to augment DFT by methods that are faster, or better at treating nonbonded interactions. One possible extension is to combine quantum mechanical calculations (like DFT) with rapid molecular mechanics, allowing more extensive searching of the available conformational space. Ananikov, Musaev, and Morokuma have reviewed several hybrid QM/MM approaches for transition metal complexes and larger systems, whereas Kozuch and Shaik combine QM/MM calculations with the energetic span model to optimize ligands for a model C–S coupling reaction.

Stereoselectivity poses special problems for computational techniques. The selectivities are largely determined by non-bonded interaction, which are not always well represented in DFT, and frequently a multitude of reaction pathways must be evaluated. On the other hand, the high similarity between competing pathways can give rise to beneficial error cancellation. Maseras and coworkers utilized QM/MM methods in the study the asymmetric Pauson–Khand reaction, whereas Ohno and Maeda, using a quite different approach, studied the asymmetric Ru-catalyzed hydrogenation reaction with their new global method for identification of alternative reaction channels. Going away from DFT, Kozlowski and Ianni show how asymmetric selectivity can be predicted through QSSR, whereas Corbeil and Moitessier present a focused review on methods applicable to high throughput prediction in asymmetric catalysis, covering also approaches based on molecular mechanics force fields.

To conclude, this Special Issue of The Journal showcases the breadth and utility of computational methods in the field of homogeneous catalysis. Application of methods like these or their successors in years to come will eventually increase our knowledge of catalytic systems to the point where true design of new and selective reactivity becomes possible. With the increased understanding of chemical processes, we will be able to achieve a sustainable production, a key factor in the future sustainable society. We hope that you enjoy reading this special issue and that it will encourage you to contribute to the evolution of The Journal in this important and rapidly expanding area.

Per-Ola Norrby [*]
Department of Chemistry, University of Gothenburg,
Kemigården 4, 412 96 Gothenburg, Sweden

Guy C. Lloyd-Jones
School of Chemistry, University of Bristol Cantock's
Close, Bristol, BS8 1TS, UK

[*] Corresponding author. Tel.: +46 31 7869034; fax:
+46 31 7723840.
E-mail address: pon@chem.gu.se (P.-O. Norrby)

Available online 10 April 2010