Reaction and Catalysis Engineering – Back to Fundamentals

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We are already celebrating the 5th edition of *Perspective* articles for the journal's yearly special issue on *Reaction Engineering and Catalysis*. This issue contains outstanding contributions from academic and industrial researchers, which highlight progress on fundamentals and their importance in catalyst and reactor design. No model is perfect, but it needs to be an adequate representation of reality, and so it should include all relevant ingredients to be the basis for trustworthy process analysis and design. Without rigorous modelling, understanding is lacking and there is a risk to draw wrong conclusions by extrapolating from experimental data measured under conditions that differ from those relevant in practice. A return to fundamentals is essential to leverage the opportunities in materials and catalytic chemistry and analysis, which receive a lot of attention today, but will fail in their promise if they are not coupled with rigorous reaction/reactor engineering fundamentals.

As this issue exemplifies, there is a need presently for the study of fundamentals at all levels, starting from the microscopic (reaction mechanisms and kinetics, with understanding of the nature of the active sites and the reacting species), to the mesoscopic (hydrodynamics, heat and mass transfer), and the macroscopic (reactor) level.

Grabow and colleagues illustrate the opportunities offered by quantum chemical methods, in conjunction with experiments, to help develop selective, heterogeneous catalytic oxidation reactions in the presence of water as a co-catalyst. Water plays an important role in "green" partial oxidation routes, involving O_2 and H_2O, but the quantity of water matters and there is often an optimum amount. In their short review of recent literature, the authors discuss the oxidation of CO in the presence of small quantities of water, direct synthesis of H_2O_2, propene epoxidation, alcohol oxidation, and the conversion of methane into methanol. Water-assisted O_2 activation is especially important to Au catalysis, but also to Pt-group and other transition metal alloys, when O_2 dissociation is rate-determining and H_2O can assist and control this step.

Marin and colleagues note that today's CRE community is facing challenges as a result of environmental concerns and the new focus of the chemical process industry on using alternative feedstocks. Computer simulations already play a key role in facing these challenges, and are expected to play an even more important role in the future. One of the cornerstones of such efforts, which *Marin* and colleagues review in their paper, is automated kinetic model generation aimed at so-called "molecule-based management". They note that the CRE community is already working hard at improving the predictive capabilities of automated kinetic model construction, e.g., via on-the-fly quantum chemical calculations. Molecular reconstruction to accurately estimate the composition of a feedstock is a key enabling technology today towards molecule-based management of chemical processes. How a complex feed reacts is best described by intrinsic kinetics derived with detailed reaction networks. *Marin* and colleagues note that automated procedures exist to construct large kinetic models of

industrial relevance. When dealing with a large number of species and elementary reactions, on the other hand, determining accurate kinetics still remains a difficult task, as calculation methods cannot always deliver the required precision, and experimental work is too cumbersome and/or expensive. On-the-fly quantum chemical calculations are an important advance to address the challenge and in search for new reactions and missing pathways. Once completely developed, these modelling tools will be highly valuable to optimize process conditions, reduce environmental risks, and ensure economic process viability.

Vanden Bussche and Jeroro discuss designer-made catalysts and their performance, and describe how the CRE field is contributing to better and faster catalyst design. They note that, while we are still falling quite short of the ultimate goal of "computer-designed catalysts", several key innovations to date in feed and catalyst characterization tools are increasingly allowing us to develop breakthrough catalysts. CFD and cold-flow experimentation can be synergistically combined to largely eliminate the scale-up risk associated with developing new catalytic processes.

Boroun and Larachi take us beyond the intrinsic physicochemical properties of magnetic nanoparticles (MNPs), of interest, for example, to biomedical applications, by reviewing their collective behaviour in dense suspension, as "ferrofluids", which opens up unique applications to chemical reaction engineering. Hydrodynamics, mass and heat transfer are notably affected by changes in the magnetic field, and there are opportunities for process intensification when using oscillating and rotating magnetic fields that can curtail Brownian relaxation effects, and enhance the desired magnetic coherence. New work from *Larachi* and co-workers on controlling the RTD of ferrofluids is discussed, and a short review on magnetic stabilisation of fluidized beds is presented. The authors also review fundamental mechanisms (and open questions on these) to control mixing and mass transfer using MNPs, including interphase mass transfer, as well as heating and heat propagation, all of which are attractive when these same particles can also be used as catalysts. Their review mentions a lot of progress in this exciting field, but also emphasises the need for more rigorous formulations of untreated ferrohydrodynamic problems.

Li, *Ge* and colleagues from the Institute of Process Engineering in Beijing review 30 years of research on the energy-minimization multiscale (EMMS) model, which originated from their Institute. This method is rooted in non-equilibrium thermodynamics and variational calculus to solve a large range of problems targeted at bridging the "mesoscales" that are so hard to model in multiphase reaction engineering problems in particular. They focus mostly on gas-solid fluidization, where dynamic particle clusters present challenges to conventional modelling approaches, leading to widely different drag coefficients in different phases. The EMMS method has achieved a lot of success, and is now even applied for the design of industrial processes. *Li et al*. note that, generally speaking, chemical processes involve material, reactor and factory levels, whereas characterisation and design at each of these levels is a multiscale problem, with the mesoscale corresponding to, respectively, molecular assemblies, particle aggregates (e.g., in fluidized beds), and combinations of unit operations. The mesostructures at each of these scales correspond to material and interfacial structures, heterogeneous flow structures and process synthesis superstructures. For each of these mesoscale problems, the EMMS approach uses a variational criterion to seek a compromise in competition between dominant mechanisms that affect the system stability – for example, for fluidization, minimizing energy consumption for suspending and transporting particles, on the one hand, and minimizing voidage, on the other hand. This compromise leads to different regimes, subject to operating conditions, with the appearance of complex mesoscale phenomena and high dissipation in the intermediate regime. Apart from stressing the importance of mesoscience in

tackling complex problems, which can only be solved by interdisciplinary teams, virtual reality is emphasised as a promising research tool.

Hickman and Ribeiro discuss the right-sizing of laboratory fixed-bed reactors to enable the generation of quality data useful for catalyst screening and development, as well as reactor scale-up. The authors wish to provide a reminder of the importance in making adequate choices in designing and operating lab-scale reactors, and, for this, they return to the fundamentals while striving to break new ground. Their perspective highlights a public-domain, web-based tool that they developed to enable our community to quickly assess the thermal and concentration gradients in and around a single catalyst particle, as well as in a fixed-bed, providing guidance to enable users to design lab-scale fixed-bed reactors that generate data under isothermal and plug-flow conditions, while also checking for mass and heat transfer gradients at the particle scale. Going back to the foundations of CRE, it should serve as a reminder of the importance of performing such calculations.

This issue of *Current Opinion in Chemical Engineering* highlights the revived interest in expanding on the fundamentals of chemical reaction engineering, as key questions need to be resolved to tackle complex multi-scale problems and bring us one step closer to rational, model-based reactor design.