

Effective strategies for collectively improving the robustness of chemical kinetic models

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Detailed chemical kinetic models are now being used to describe an increasingly wide range of thermochemical processes such as in combustion, atmospheric science, chemical engineering, pharmacokinetics, and metabolic pathways. The development of such models can provide us with improved understanding of complex couplings between chemical and physical processes that can assist in developing design strategies. Examples of such design processes include the development of low carbon fuels and high efficiency engines, mitigation strategies for atmospheric pollutants, the design of new drugs etc. Some of the world's greatest thinkers offer differing advice about how to deal with such complexity. According to Ernst Schumacher "Any intelligent fool can make things bigger, more complex, and more violent. It takes a touch of genius — and a lot of courage to move in the opposite direction." The advice of Schumacher is therefore to simplify. On the other hand Fisker states that "Increasing complexity always increases capability and adaptability." So according to this line of thought, larger more detailed models will allow us greater flexibility in describing complex processes. However, if we develop larger and more detailed models which increase our adaptability, are we also overcomplicating things? Clearly by developing bigger and bigger kinetic mechanisms we are creating need for a larger number of detailed kinetics and other studies required to populate the models with data. Despite significant advances in ab initio theoretical kinetics calculations which have reduced the uncertainty of their predictions, we are not at the stage where detailed experiments can or should be abandoned. Therefore to further improve models still requires extensive collective experimental and modelling effort.

The question arises as to whether as a community we are tackling the development of kinetic models in the most effective way. Do we need careful quantification of all the model data or are some parts of the model more important than others? What we would like to be able to do is to focus in on particular aspects of the models that determine our ability to predict important targets (e.g. fuel efficiency, engine knock potential, pollutant concentrations and exposure, disease outcomes). If we are able to identify the key processes then we could potentially design experiments or theoretical studies that accurately quantify important rate parameters or thermodynamic data in the most effective way for these key steps. Other parts of the model may require less accuracy, or may in certain circumstances even be redundant, thus providing the opportunity to simplify. However, deciding which parts of the mechanism are redundant in the absence of any data would perhaps be foolhardy. Therefore a process of model refinement may be necessary, which is a large task for a single research group.

The paper will discuss a proposed framework for tackling such issues and therefore for the development of chemical models with improved robustness that can be used in engineering and strategic design. Topics covered will include mechanism development, global uncertainty and sensitivity analysis methods for assessing model parameter importance, collaborative working tools for the effective gathering and sharing of data, model optimization techniques and the use of multi-scale informatics, the information content of experiments, and experimental design strategies.