Automated Predictive Chemical Kinetics: Triumphs and Challenges

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A long-term goal of the chemical kinetics community is to be able to quantitatively predict the time evolution of a reacting mixture. The ability to do this quickly and reliably would completely transform many areas of technology which currently rely on rough estimates and trial-and-error experimentation. To be fast/efficient enough to be practically useful, the whole process of making the predictions needs to be automated.

Advances in rate theory, quantum chemistry, and kinetic modeling algorithms, coupled with the constantly increasing number of accurate experimental rate coefficients and thermochemical parameters, are beginning to make it possible to quantitatively predict chemical kinetics even in the absence of experiments in some cases. However, these predictions are not yet reliably accurate, so experimental validation is highly advisable. Some of the triumphs and failures of predictive chemical kinetics using the open-source RMG software are highlighted, using as examples the combustion of butanol isomers, the pyrolysis of the synthetic jet fuel JP-10, and reactions of organic sulfides in supercritical water. The reasons for the failures are examined. We outline several challenges that the kinetics community must overcome before we will achieve our long-term goal of reliable quantitative predictions for reacting mixtures.