Automated elementary kinetics for gas-phase reactions

Judit Zádor

Combustion Research Facility, Sandia National Laboratories, Livermore, CA 94550, USA
e-mail: jzador@sandia.gov

With the growing need to study the oxidation kinetics of new types of fuel molecules, theoretical chemical kinetics faces new problems. These new fuels, often derived from biological or other non-traditional sources, have two properties that cause difficulties: (*i*) they contain functional groups for which there are relatively few fundamental studies available, and (*ii*) the molecules of practical importance tend to be large. Although a significant portion of our knowledge could be transferred from small molecules to larger ones in the form of rate rules for instance, the reliability of these estimates for important quantities (rate coefficients, branching fractions, pressure dependence, etc.) is not known *per se*. It is naturally a much better approach to study these new molecules directly on the computer instead of using estimates, and while in principle we are in the position to do so, we do not know the limitations and scalability of the currently used methods for these larger systems.

In my talk I will show that larger molecules, even if of "traditional" kind, produce a combinatorial challenge when it comes to the exploration of the potential energy surface. I will partially review the currently available methods to overcome this problem, and show how the code I develop, called KinBot, enables such an exploration in an efficient way tailored for the needs of combustion and atmospheric chemistry. I will illustrate through several examples the capabilities of the code and present cases where it found new reaction types. I will make a case for the need and possibility of fully automated theoretical elementary reaction kinetics, and also describe its limitations.