

Progress towards Accurate Predictive Gas Kinetics

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It would be highly desirable to be able to build accurate kinetic models for technologically and societally important systems, so we could do predictive design of improved systems on the computer, or more reliably anticipate the consequences of changes in anthropogenic inputs to natural systems. Most technologically or societally important gas kinetics systems contain a multitude of reactive chemical species, and it is impossible to fully elucidate these processes and so develop accurate predictive models by experiments alone. Here we report recent progress toward predicting the time-evolving composition of these systems *a priori*: how unexpected reactions can be discovered on the computer, how reaction rates are computed from first principles, and how the many individual reactions are efficiently combined into a predictive simulation for the whole system. These model predictions can be quite accurate, see for example an example from the literature in Fig. 1 below. Some of the sources of error in these predictions, and associated unresolved challenges, are highlighted.

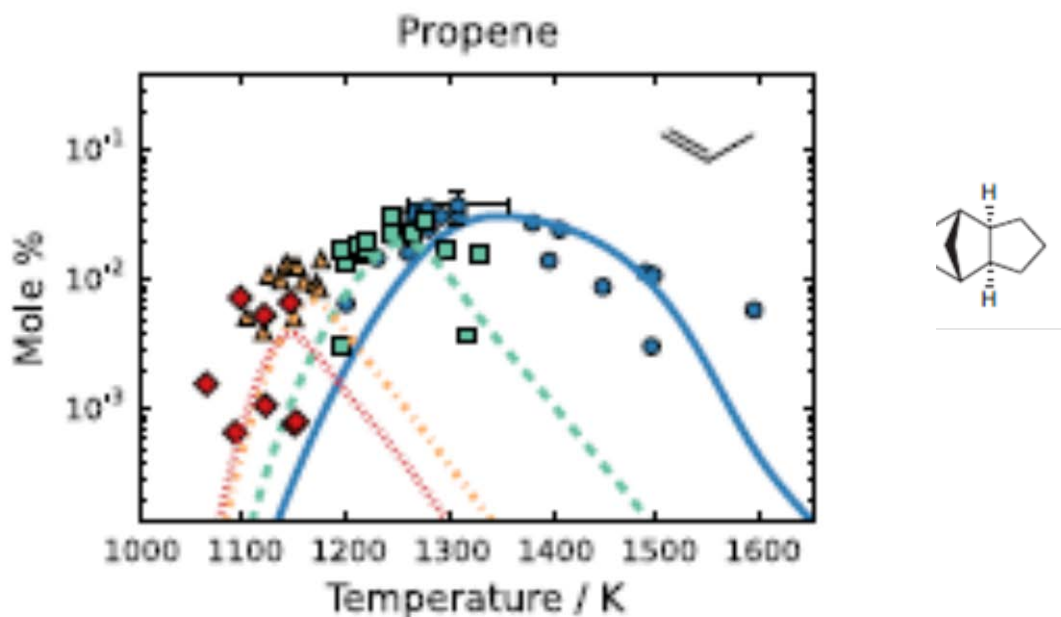


Fig. 1. Predicted vs. Experimental yield of propene from pyrolysis (solid line, circles) or partial oxidation (dashed lines and matching symbols) of *exo*-tetrahydrodicyclopentadiene (structure shown to right of the data plot). Experiments done in a single-pulse shock tube; kinetic model containing 691 species was built automatically by the Reaction Mechanism Generator (RMG) software, with parameters derived from quantum chemistry calculations. No parameters in the model were adjusted to fit the data, the curves are predictions not fits. After Gao *et al.*, *Combustion & Flame* **162**, 3115-3129 (2015).