Uncertainty Quantification and Data Discrimination in Combustion Kinetic Modeling

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Chemical mechanisms in combustion consist of large numbers of elementary reactions. Models to describe such mechanisms must therefore include many rate parameters in order to describe all of these reactions. The rate parameters cannot be determined from first principles and so must be measured. Measuring individual rate parameters can be difficult, however, and estimates from theory are known to be highly uncertain.

As computational power has increased, it has become easier to simulate complex combustion systems with large fuels. Indeed, our ability to describe the combustion mechanisms of these fuels has outstripped our ability to precisely measure individual rate parameters, which has left us with large numbers of rate parameters with large uncertainty. This has led to a tendency to tune chemical reaction models to match experimental measurements, adjusting the model rate parameters until the model can reproduce the observations. In principle, at least, such adjustments result in a model with a set of rate parameters that are closer to reality and have tighter uncertainties.

Tuning models against experimental data has two significant pitfalls, however. The experimental measurements (the training set) and the parameters for adjustment are often chosen on an ad-hoc basis. This ensures a set of model parameters that will reliably reproduce the training set. Without an estimate of the uncertainty, however, it is not possible to reliably describe the model's performance outside the training set. Furthermore, the training set may include many experiments are chemically similar, meaning that perhaps only one condition need have been measured. This has resulted in a proliferation of chemical kinetic models published by different groups.

This talk will discuss recent development of experimental and modeling methodologies, in particular the Method of Uncertainty Minimization using Polynomial Chaos Expansions. These methodologies use data analytics, uncertainty analysis, and experimental design techniques to produce a rigorously optimized model with quantified prediction uncertainty, while at the same time minimizing experimental measurement duplication. Rigorous optimization ensures that the recommended rate parameter values are indeed the best, based on the data provided. A quantified prediction uncertainty provides that the behavior of the model is known outside the training set. Experimental design ensures that the recommended model is based on best set of experimental measurements available. Examples of the application of the method will include estimating rates of radical attack on small hydrocarbons using detailed kinetics measurements in shock tubes and optimization of combustion reaction models using laminar flame speeds and ignition delay times.