UQ-Predictive Modeling of Chemical Reaction Systems

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The presentation will focus on methodology of developing chemical reaction models to attain the desired level of predictability. The knowledge gained through increasingly sophisticated experimentation and theory forms the basis for the development of models. But what makes such models truly predictive?

The theoretical dream is to build models entirely from first principles. However, even most fundamental of the present models include uncertainties. There are many sources of model uncertainties: incomplete knowledge of the physical phenomena, truncated expansions of numerical methods, numerical diffusion, and the like. One view of model predictiveness is to gain higher and higher veracity for all parts of the model and by this virtue alone acquire trust in predicted results. Even in this possibly utopian view, the question of how to judge that the model predictions are sufficiently accurate needs to be answered. Furthermore, one would like to have a direction for advancing the model predictiveness. Not all model parts and not all of their uncertainties contribute equally to the accuracy of model predictions, especially when one is interested in a specific set of conditions. The usually complex, nonlinear nature of models of physical phenomena prevents one from identifying the extent to which individual uncertainties influence model predictions without analysis.

A broad field of study and techniques related to the numerical characterization of uncertainty has been termed *uncertainty quantification* (UQ). Its main objective is the propagation of model's uncertainties to the model's prediction, and hence "uncertainty quantification" has become synonymous with "predictive modeling". We here define "predictive" to mean that the numerical result of a model is accompanied by its rigorously determined uncertainty bounds and a more *predictive* model is the one with more narrowly bounded interval. Hence, we will term such models *UQ-predictive* and the associated process *UQ-predictive modeling*.

This presentation will outline specific characteristics of chemical reaction systems and difficulties associated with development of predictive models, past and contemporary directions to predictive-model building, and the UQ framework we termed Bound-to-Bound Data Collaboration (B2B-DC). The latter is an integrating, system approach that is built on an underlying physical process and associated model, a collection of experimental observations with specified uncertainties, algebraic surrogate models (response surfaces) representing parametric dependence of the physical-model predictions of the experimental observables on the uncertain parameters, and specialized constrained-optimization algorithms. The examples will be drawn from the field of combustion chemistry, demonstrating the methodology and its capability. The presentation will outline emerging new concepts and future needs in the field of UQ-predictive modeling.