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Transforming data into knowledge—Process Informatics for combustion chemistry

Michael Frenklach *

Department of Mechanical Engineering, University of California, and Environmental Energy Technologies Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA

Abstract

The present frontier of combustion chemistry is the development of *predictive* reaction models, namely, chemical kinetics models capable of accurate numerical predictions with quantifiable uncertainties. While the usual factors like deficient knowledge of reaction pathways and insufficient accuracy of individual measurements and/or theoretical calculations impede progress, the key obstacle is the inconsistency of accumulating data and proliferating reaction mechanisms. Process Informatics introduces a new paradigm. It relies on three major components: proper organization of scientific data, availability of scientific tools for analysis and processing of these data, and engagement of the entire scientific community in the data collection and analysis. The proper infrastructure will enable a new form of scientific method by considering the entire content of information available, assessing and assuring mutual scientific consistency of the data, rigorously assessing data uncertainty, identifying problems with the available data, evaluating model predictability, suggesting new experimental and theoretical work with the highest possible impact, reaching community consensus, and merging the assembled data into new knowledge.

Keywords: Kinetics; Modeling; Optimization; Consistency; Informatics; PrIMe

1. Introduction

1.1. Progress in combustion depends on reliable reaction kinetics

Research is motivated by the human desire to explain phenomena surrounding us and the passion for new discoveries. Driven by individual satisfaction such activities aim at improving the quality of human experience. One can generalize the stimulus for research as developing the ability of *making predictions*. Indeed, predicting properties of a material before it is synthesized, predicting performance of a device before it is built, or predicting the magnitude of a natural disaster ahead of time all have obvious benefits to society.

In the area of combustion, we have all the above elements: the fascination with fire from ancient times, its broad application to numerous aspects of everyday life, and the definite need for reliable predictions (see Fig. 1). In recent decades the combustion research was largely driven by the desired increase in energy efficiency, reduction in pollutant formation, and material synthesis. The need for tying the research to practical applications was further emphasized in the Hottel address of the 28th Symposium by Professor Glassman [1].

* Fax: +1 510 643 5599.
E-mail address: myf@me.berkeley.edu

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Sometime in the year 2008 …

Chemist to PrIMe: I have an idea of how to measure the elusive reaction between $C_{14}H_7$ and C_3H_3 forming $C_{16}H_8$ and CH_2. What impact would such a measurement have on present knowledge concerning the nucleation of interstellar dust?

PrIMe to Chemist: If the rate coefficient is established to within 3% accuracy, I will be able to discriminate between two competing hypothesis, A and B.

Chemist to PrIMe: I do not think my experiment can attain better than 10% accuracy. What is the next best thing can I do experimentally to advance knowledge of this subject?

PrIMe to Chemist: Measure the reaction between $C_{10}H_7$ and C_3H_2; I can then discriminate between hypotheses B and C.

… in the year 2010 …

Engineer to PrIMe: What fueling rate produces peak output power while holding NO_x yields within the EPA prescribed limits in a HCCI engine running GTL prescribed fuel #22 with the following design and operating parameters: xx,yy, …

PrIMe to Engineer: How well do you want to know this?

Engineer to PrIMe: I need 5% accuracy!

PrIMe to Engineer: This accuracy is not achievable. The uncertainty range on the fueling rate runs from 1.22 to 1.35 g/s.

Engineer to PrIMe: What is the dominant source of this uncertainty?

PrIMe to Engineer: 80% of the uncertainty in the predicted fuel consumption rate is caused by the uncertainty in the estimated rate constant for the reaction $(CH_3)_2CCHCH_2+O_2 \rightarrow (CH_3)_2CCCH_2 + HO_2$. There are no literature data on this reaction, only indirect estimates. The needed data can be obtained via quantum chemistry calculations (time 2 days, cost $$) or by performing a series of experiments (time 2 years, cost $$$$); what is your choice?

Engineer to PrIMe: The quantum chemistry calculations right away and start experiments as well.

PrIMe to Engineer (2 days later): Optimal fueling rate needed is 1.31 g/s, based on computed rate constant of 5×10^9 cc/mol s. Prediction uncertainty range is 1.29 – 1.33 g/s.

PrIMe to Chemists: Perform measurements to determine the rate constant for the reaction $(CH_3)_2CCHCH_2+O_2 \rightarrow (CH_3)_2CCCH_2 + HO_2$.

… and in the year 2020 …

Policymaker to PrIMe: How much longer will there be an Antarctic ozone hole?

PrIMe to Policymaker: 50 to 150 years.

Policymaker to PrIMe: Can I get the answer more accurately, within at worst a 5 year interval, and what would it take?

PrIMe to Policymaker: My result is the best current estimate based on all available data and the scientific community consensus. To get to the requested level of certainty the heat of formation of Cl_2O_2 has to be known to within 1 kJ/mol and the rate coefficient for the reaction between two ClO radicals to within 10%.

Policymaker to Chemists (via funding agency): Please (re)measure/(re)calculate the heat of formation of Cl_2O_2 and the rate coefficient for the reaction between two ClO radicals to the above accuracy.

Fig. 1. The Prolog from NSF proposal "Collaborative Research: Cyberinfrastructure and Research Facilities: Process Informatics for Chemical Reaction Systems" by M. Frenklach, A. Packard, C.T. Bowman, D. Golden, W.H. Green, G. McRae, 2005.

One of the key areas of present and future research in combustion, emphasized in essentially all Plenary Lectures of the 30th Symposium [2–5], is chemical kinetics. The combustion community has realized from very early on that dynamics of reaction systems plays a critical role in combustion