Process Informatics Model (PrIMe): A Systematic Approach to Building Combustion Chemistry Models

Investigators
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Introduction

Development of new energy-efficient low-emitting combustion technologies requires models for the combustion process that combine sub-models for combustion chemistry, heat and mass transfer, and fluid mechanics. A major limitation in developing these process models is the availability of accurate, validated, and computational tractable combustion chemistry models. This research is aimed at the development of a new approach for constructing and reducing models of complex chemical reactions. It has been termed *Process Informatics.* This endeavor is made possible by recent advances in computer science that allow assembly and manipulation of large amounts of data that may be scattered over different sources using Web-based computer networks. The goal is to convert combustion chemistry model building into science, to automate the methodology, and to make the information available in a prompt and convenient form on the Internet for researchers and designers of combustion equipment

Background

A good deal of the effort in this project is in essence political. That is, it involves community organization in the combustion research community and it requires the acceptance of a somewhat different paradigm than the standard in the community. External developments in the field in the past year involve several workshops and symposia in which the PrIMe idea has continued to gain currency. Presentations by the PI, Professor Golden, as well as American collaborators at UC Berkeley, MIT, SRI International and international collaborators from the UK and the Continent and other such developments are discussed below in the "Results" section of this report.

As previously reported, The National Institute of Standards and Technology (NIST) has agreed to be the permanent host for the PrIMe Library and eventually for the computer based tools required for model development. Some of these tools are being developed in a Sandia National laboratory led effort called, Collaboratory for Multi-scale Chemical Science (CMCS). Initially PrIMe will be available at Sandia as well. Professor Golden is a member of the Advisory Board of CMCS. Interest in industry is picking up with the attached endorsement from Caterpillar Inc. a major engine manufacturer.

The National Science Foundation has issued a solicitation entitled: Chemistry Research Instrumentation and Facilities: Cyberinfrastructure and Research Facilities. The program "will provide funding to build a foundation for cyber-enabled chemical research and education". As this Report is being written, a proposal encompassing PrIMe is being prepared as a collaboration between Stanford, (Golden), UC Berkeley (Frenklach andPacker) and MIT(McRae and Green). This solicitation grew, from among other events, a report on cyberinfrastructure discussed in the NSF solicitation.

"As described in the report, "Revolutionizing Science and Engineering Through Cyberinfrastructure: Report of the NSF Blue-Ribbon Advisory Panel on Cyberinfrastructure," the manner in which scientific and engineering research and education is conducted will be radically transformed by cyberinfrastructure. This report may be accessed at http://www.cise.nsf.gov/sci/reports/atkins.pdf. The NSF Division of Chemistry shares this vision and has held a workshop that has identified research and education frontiers that would be enabled by investments in cyberinfrastructure. The report from this workshop may be accessed at http://bioeng.berkeley.edu/faculty/cyber_workshop/. A National Research Council report, "Information and Communications," also identifies opportunities in cyberinfrastructure in the chemical sciences and is available at http://books.nap.edu/catalog/10831.html."

Results

The first goal of the PrIMe project is the creation of a database or library, sometimes referred to as a warehouse for combustion data. The goal is an agreed upon, evaluated by the community, updated in a timely fashion, data library. The library is open, accessible to all and contains, beside the agreed upon values, all data and all dissents from the consensus evaluation. There are some models for this type of activity, Professor Golden has been a member of the NASA/JPL Panel that evaluates rate data for atmospheric modeling, but the goal here is more far-reaching. All background information used in the evaluations will be available.

This program requires bringing a large community together and it has been somewhat slower than hoped. Professor Golden, Professor Michael Frenklach of the University of California, Berkeley and Professor Michael Pilling of Leeds University have acted as a "troika" in engaging the community. Prominent additions to the active team include Dr. Wing Tsang of NIST, Professors William Green and Gregory McRae of MIT and Dr. Jeremy Frey of The University of Southampton, UK. Many other workers have expressed their willingness to participate.

The first informal gathering of possible PrIMe participants took place during the International symposium on Combustion in Sapporo, Japan in August 2002.

Professor Frenklach of UC Berkeley made a presentation at the DOE Basic Energy Sciences Contractor's Meeting in Lake Tahoe, CA in May 2003 that outlined possibilities associated with this project. He and colleagues have published an article[1] in December 2003 that points out the large amount of information available from extant experiments that is not being accessed as result of lack of familiarity with many statistical procedures that can now be accomplished with relative ease. A second article[2] by the Berkeley group on mutual consistency of data appeared recently.

A meeting at NIST in September 2003 aimed at establishing a data base for "Real Fuels" served as a venue for discussions that resulted in NIST becoming the home for PrIMe. At that meeting Professor Golden explained the procedures used to develop the widely used Natural Gas combustion mechanism, known as GRI-Mech. He pointed out

all the ways this could be better accomplished once PrIMe is underway. Professor Frenklach, once again described the future of process modeling.

In March 2004 there was a symposium in the Fuel Section of the National American Chemical Society Meeting in Annaheim , CA. Two days of speakers discussed combustion modeling and PrIMe was a central focus in several presentations. (The agenda is attached and those presentations directly involving PrIMe related activities are highlighted.) A presentation based on collaboration with Professor William Lester, Professor Frenklach and students at UC Berkeley, includes Professor Golden. A paper based on this presentation has been accepted for publication in the International Journal of Chemical Kinetics, and is listed below.

In October 2004, there was a meeting at NIST during which the PrIMe concepts were briefed to a group of government Program managers. The briefing was conducted by Professors Golden, McRae and Frenklach and Dr. J. Manion of NIST. Program managers present were: Frank Tully, Dick Hilderbrandt and Eric Rohlfing all of DOE, Julian tishkoff of AFOSR, David Mann of ARO and Linda Blevins of DOE. A copy of the informal review of that meeting is attached.

At this time all the background data used in developing the GRI-Mech model has been cast into XML format and the very large amount of data available at Leeds University will be translated and made available for PrIMe. Extensive data searching tools are also being developed.

Dr. Gregory P. Smith of SRI International, Menlo Park, CA has together with Professor Frenklach and colleagues at Berkeley, applied the tools of process informatics to understanding a dichotomy in the understanding of OH chemistry in the upper stratosphere and the lower mesosphere. A paper[3] is being prepared.

Progress

In the instructions for this report we were asked to provide a discussion of the progress achieved to date toward the goal of developing the basis for technology options that could lead to substantial reductions in emissions of greenhouse gases that result from energy use. In other words, describe the potential impact of the research, if successful, on greenhouse gas emissions at a global scale.

This is a far-reaching goal. The best illustration of the future use of PrIMe for this purpose is found in the attached letter of support from Caterpillar Inc.. The goal is to make modeling and design of combustion driven devices easier and accurate, with the goal of energy efficiency and minimal pollution of all kinds.

Also attached is a letter from the University of Utah combined with Sandia National Laboratories that shows their enthusiasm for the PrIMe project.

Future Plans

This research project is really the tip of an iceberg. There will be ongoing work in codification of data in XML format and in putting together the evaluation teams. There will be an ongoing attempt to secure funding for a large scale collaborative effort. The work being put into the effort to secure NSF support will hopefully bear fruit. However, even if this attempt fails, we will have put together a team and documentation for further attempts to secure funding. Hopefully the modest support for Professor Golden will allow his continued participation.

Publications and Presentations

1. Evaluating and Codifying Data for Engineering Applications
   Presented by David M. Golden at NIST Workshop on Real Fuels, September 2003

2. Quantum Monte Carlo study of the thermochemistry of small hydrocarbons
   Kollias, A., Domin, D., Frenklach, M., Golden, D.M., and Lester, W. A., Jr.
   Accepted for publication International Journal of Chemical Kinetics

3. Presentation by David M. Golden at NIST briefing October 2004.

References

1. Frenklach, M. , Packard, A., Seiler, P., and Feeley, R.,; Collaborative data processing in developing predictive models of complex reaction systems, Int. J. Chem . Kinetics 2004, 36, 57-66

2. Feely, R., Seiler, P., Packard, A., and Frenklach, M.,; Consistency of a reaction dataset, J. Phys. Chem. A 2004, 108, 9573-9583.

3. Smith, G. P., Frenklach, M. , Packard, A., Seiler, P., and Feeley, R. A System Analysis Approach for Atmospheric Observations and Models: the Mesospheric HOx Dilemma,

Contacts

David M. Golden: david.golden@stanford.edu

Summary of PrIMe Meeting at NIST on October 12, 2004

Presentors: Greg Rosasco NIST; Grec McRae, MIT; David Golden, Stanford; Michael Frenklach, UC Berkeley; Jeff Manion, NIST
Attendees: Frank Tully, DOE; Dick Hilderbrandt, DOE; Eric Rohlfing, DOE; Linda Blevins, NSF; Julian Tishkoff, AFOSR; Dave Mann, ARO
Observers: Wing Tsang, Tom Allison, Jeff Hudgens, NIST

Motivation for PrIMe
Kinetic models are a vital ingredient in optimizing key industrial processes
Reduction in the elapsed time to obtain solutions to science/engineering problems
Converting model building into science rather than "art"
A formal framework for making resource allocation decisions
Creation of a larger community with synergistic interactions

Precursors to PrIMe
Data gathering-NIST Database
Evaluated data gathering-NASA Panel
Evaluated data gathering and optimization to produce "best current set"-GRI-Mech
All would be simpler and more reliable if tools were to be developed to aid in acquiring and processing data and assuring the data completeness and mutual consistency,

PrIMe Goal
Create *infrastructure and tools* for the development of predictive reaction models of combustion
--by extracting *all possible information* from all available data, and converting this knowledge into predictive models
--by establishing and demonstrating the *community approach* to kinetic-model development and, perhaps most importantly, establishing a means for reaching community consensus on the models and data

PrIMe Structure
Warehouse
--*Data Depository*-All Traceable Data (Good & Bad)
--*Library*-"Best Current Set" of values with uncertainties
Tools

Guiding Principles
Open membership to all qualified people
"Open-source" data sharing
Distributed management and shared scientific leadership
Data integration
Single copy of each data entry in the *Data Depository*
Periodic updates of "best current set" in the *Library*
On-the-fly model building

PrIMe Organization

PriMe team is organized into work groups
Each group has a responsibility for "monitoring" specific parts of *Data Depository* data
Work group activities include reviewing comments, recommendations, opinions, and new data, as they are submitted by the community at large, assessing their merits and completeness, and making a recommendation to PrIMe team
Membership in PrIMe work groups is open to all qualified individuals

PrIMe Customers

Data Provider

Experimenter (theorist) submits new measurement (calculations) to the *Data Depository*
Submission protocol assures completeness of the data
Submitted data are analyzed for consistency with the *Library* and the results are reported both to the *Data Provider* and to the PrIMe team
Upon consensus of PrIMe team, data is "deposited" and *Library* is modified (and periodically released to public) and/or feedback given to *Data Provider* on what may need to be checked, redone, done next, etc. (The possibility that new data at odds with the *Library* may be better than the extant values, will not be ignored!)

User

Customer needs a chemical model as input to a CFD code
This *User* specifies model requirements: type, size, accuracy, ranges of conditions to be covered, output format, etc.
PrIMe Tools generate such a model, if possible, and it is built on the fly
If not enough data are available, PrIMe provides choices to generate required data

Bringing-in the Community

Pulling rather than *pushing*
Moving via "lowest levels of mutual agreement"
Currently this means creating the Warehouse

Examples of Tools for PrIMe

Reaction mechanism generation/reduction
Estimation of thermochemistry data
Dataset consistency
Parameter/state estimation
Uncertainty propagation
Advanced numerics for stiff systems
Design optimization/parametric analysis

Major Benefits

Development of reaction models in a systematic, scientifically rigorous way
Better utilization of resources
Availability of best possible working model at any given time
Clear identification of future programmatic needs
Establishing of a new paradigm in the field of complex model development

Current Status of PrIMe

Building and launching of PrIMe (CMCS + NIST)

XML data formats have been developed (nearly completed)

Data are being prepared in those formats (nearly completed)

Data Depository is being populated with data (beginning)

Submission forms are being developed (~20% complete)

Working group collaborative environment (~90% completed)

Tool development – through individual efforts

Community support

US "major players", Utah–Sandia, Albuquerque,…

Companies: Caterpillar, (ExxonMobil, Volvo, …)

International: UK (Leeds), Germany, Sweden, …

Other fields

Atmospheric chemistry, astrophysics/astrochemistry,…

Tentative Timeline

PHASE I

-Building for the initial launch of PrIMe

--*Data Depository* is populated with the initial set of data based on the NIST library and the GRI-Mech release 3.0

--Data submission forms are developed and integrated through CMCS infrastructure

--operation of PrIMe working groups is enabled through CMCS portal

-Activation of the PrIMe group leaders and the "formulation" of the PrIMe groups

-Official launch of PrIMe: announcement to entire community and inviting submission of "individual" data

-Organizational meeting of the PrIMe team, and setting the objectives for the next year (i.e., what the PrIMe 1.0 relase should include; perhaps starting with natural gas system as an update and revision of GRI-Mech)

PHASE II

-Addition of data to *Data Depository* (Leeds, …)

-incorporation of simulation and analysis (sensitivity, optimization, …) tools available into the PrIMe infrastructure, through CMCS tools (ActiveTables, RecationLab, RIOT, …)

-Release of the first "best current data set" (i.e., the *PrIMe Library* 1.0)

-Annual PrIMe team meeting, and so on.

Points raised by audience

To what detail will experiments have to be entered?

Eventually it would be good to have all details. We will begin with some relatively obvious "standards" that we have developed over the past few years.

How do you support what is essentially an infrastructure with research funds?

This will require explaining how PrIMe will enhance research.

Is there an analogy with projects like the Advanced Light Source (ALS)?

This is an example of funds spent on infrastructure to enhance research.

How long before anything emerges?

We are working hard to launch the Data Depository, and expect it to happen within the current year

How do we get and demonstrate community support?

All aspects are open to the entire community. Nobody will be left out. The advantages of a single location for all data will be clear.

How do we interact with users?

Mainly through the internet --- using the CMCS Portal. Meetings will also be required from time to time, and we hope that they will occur under the umbrella of the Combustion

CATERPILLAR®

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June 3, 2004

Gregory J. Rosasco
Chief, Physical and Chemical Properties Division
National Institute of Standards and Technology
100 Bureau Drive, Mail Stop 8380
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Physics Building (221) Rm. A107

A project has been proposed by the National Institute of Standards and Technology (NIST), several universities and National Laboratories that will be of high importance to Caterpillar as we research technologies to comply with US 2010 Diesel Engine emissions regulations. The proposed effort is highly innovative, and must occur in a short time frame to have an impact on the ultra-low emissions levels required in 2010. Proposed is a multidisciplinary collaborative research initiative in combustion science enabling community-based development of highly optimized chemical kinetic models for the combustion of real, transportation fuels. This initiative is seen as a very high-impact application of advanced cyberinfrastucture in science and engineering research and allied education.

Caterpillar, like all manufacturers of heavy-duty engines, makes extensive use of computer simulations to predict in-cylinder combustion. To more accurately model the combustion process, industry and academia require better definition and documentation of the chemical processes that control the generation of the emission species. Key to this effort is a collaborative initiative to generate data libraries, analysis software, and the supporting information technology infrastructure that will enable a collaborative approach to development of predictive models of combustion. NIST has recently joined the PrIMe initiative. In its initial focus NIST will seek to enable achievement of PrIMe's first objective, i.e. the creation of the PrIMe Data Warehouse. This warehouse is visualized as a comprehensive repository of all data relevant to modeling combustion processes. Development of this library is a community project involving submission and evaluation of the data, often presented in the form of a "mechanism", describing various combustion scenarios, including reaction paths from diesel fuel to the products of combustion such as nitrogen oxides and soot.

Caterpillar technical representatives William Brown and Ken Erdman recently attended a meeting at NIST where the group discussed this problem, and were concerned with the level of support received by scientists working in this area. The assembled group was able to draft a program to address many of these concerns in a timeframe to positively impact the development of engines to meet 2010 emissions regulations. We now must ensure that this program receives the funding and support necessary to succeed. While it would have been beneficial to have this information in years past, it has become critical to future engine development programs for 2010 and beyond. We view NIST's recent association with the PrIMe effort as an important and essential first step in the process of developing accurate models.

We believe many of our European and Asian competitors are ahead of U.S. industry in this area of research. Caterpillar asks for your support in advancing this technology through this collaborative effort. The researchers we consider to be essential to this project include:

Michael Frenklach, U. C. Berkley and Lawerence Berkley
David Golden, Stanford University
Wing Tsang, NIST

They can best answer questions regarding the details of this effort. We at Caterpillar are prepared to cooperate with them in this collaboration.

Best Regards,

John K. Amdall
Director of Research
Caterpillar, Inc.

cc: Michael Frenklach
David Golden
Wing Tsang

THE UNIVERSITY OF UTAH

Adel F. Sarofim
Presidential Professor

September 13, 2004

Professor Michael Frenklach
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6161 Echeverry Hall
University of California, Berkeley
Berkeley, CA 94720-1740

Dear Michael:

This letter is to express the commitment of those involved in the C-SAFE program at the University of Utah and in the Fire Science and Technology Department at Sandia National Laboratories to assume a lead role in establishing and advancing a community-wide soot database and characterized repository for of soot formation models as part of the initiative on Process Informatics (PrIMe) you are leading with Mike Pilling and Dave Golden, and the associated initiates on CMCS by Larry Rahn and on data bases for practical fuels by Wing Tsang.

We would build on the experience we have had in organizing annual workshops as part of the pool-fire related activities at both institutions. The need for a quality database on soot formation and the use of this database to evaluate models was expressed during these workshops. We are committed to including all of the data and involving all of the national and international experts in soot modeling. The development, characterization (including applicability and uncertainty analysis), documentation, and archiving of soot models will meet a long-standing need in the fire science and combustion communities.

We would utilize the structure and format of the repository being developed by the PrIMe team and we would enjoy the opportunity to participate with the team in discussions on the evolution and implementation of the tools for model building. We see significant opportunities to advance our development of soot models for use in a variety of applications and are committed to the goal of the quantification and reduction in the uncertainty of model predictions, by thoughtfully defining the techniques, documentation, and analysis of results included in the database.

Sincerely yours,

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