Original Proposal (submitted January 2001)

# Realistic Uncertainty Bounds for Complex Dynamical Models

National Science Foundation, award CTS-0113985

09/01/01-08/31/05

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`http://jagger.me.berkeley.edu/~pack/nsfuncertainty`

# 1 Results from prior NSF support

A. Packard received an NSF PYI award, CTS-9057420, for the period 10/1/90–8/31/97 for the amount of $277,325, to study multivariable robust control, including robustness analysis, gain scheduling, and robust control design methodologies. During this time, improved algorithms for uncertainty modeling, analysis and design were developed, along with LFT and LPV gain-scheduling. The main results are described in the papers below:

1. Fen Wu, Xin Hua Yang, Andy Packard and Greg Becker, "Induced $\mathcal{L}_2$ norm control for LPV systems with bounded parameter variation rates," *International Journal of Nonlinear and Robust Control*, **6**, pp. 983-998, (1996).

2. Gregory Becker and Andy Packard, "Robust performance of linear, parametrically varying systems using parametrically-dependent linear dynamic feedback," *Systems and Control Letters*, **23**, pp. 205-215, (1994).

3. Andy Packard, "Gain scheduling via linear fractional transformations," *Systems and Control Letters*, **22-2**, pp. 79-92, (1994).

4. Andy Packard and Pradeep Pandey, "Continuity properties of the real/complex structured singular value," *IEEE Trans. Auto. Control*, **38-3**, pp. 415-428, (1993).

5. Andy Packard and John Doyle, "Complex Structured Singular Value," *Automatica*, **29-1**, pp. 71-109, Jan., 1993.

M. Frenklach received Grant CTS-9696085, entitled "Computational Study of Oxygen Interaction With Diamond Surfaces," for the period April 30, 1997 - May 31, 1999 (Extended). Award Amount: $200,000. Quantum-mechanical calculations at semi-empirical and ab initio levels were performed on a series of small and large size clusters, to explore various surface reconstruction patterns, and possible reaction steps responsible for diamond growth at conditions typical of chemical vapor deposition. The analysis was based on quantum-mechanical computations of potential energy barriers and vibrational frequencies, transition-state-theory evaluation of reaction rates, chemical-kinetic analysis of localized reaction networks, and idealized random-walk calculations of migration lengths. New reaction pathways were identified for the growth of diamond. A key factor which promotes incorporation of gaseous hydrocarbon precursor into the diamond lattice was identified to be the surface migration of hydrogen atoms. Isolated dimers on the diamond surface were found to be kinetically unstable. A phenomenon of migration on diamond surfaces was discovered. Surface migration of lone $CH_2$ and $CCH_2$ groups was computed to be fast and nearly isotropic in both directions, along dimer chains and along dimer rows. While in a general sense, this migration is analogous to surface diffusion of metals, it has a very distinctive property: the diamond migration is conditional to the formation of surface biradicals. The most distinguishing feature is migration of surface radicals. It is highly anisotropic and was found to reach one to two orders of magnitude larger migration distances than those of lone $CH_2$ and $CCH_2$ groups. Based on the above, a new paradigm for diamond growth mechanism was suggested. Starting with a reconstructed 100-$(2 \times 1)$ surface, dimer sites are converted at random into $CH_2$ and $CCH_2$ bridge sites. Both types continuously migrate on the surface, some being

etched, some $CCH_2$ being etched partially with the formation of $CH_2$ bridges, and some combining into next-layer dimers. Those dimers formed in isolation transform into $CCH_2$ bridges, but those formed right next to the existing dimer rows extend them irreversibly, thereby creating the next layer of the 100-$(2 \times 1)$ surface, and so on. This model thus establishes the governing role of surface migration in chemical vapor deposition of diamond. The critical role of the interplay between surface growth, surface etching, and surface migration was further examined and confirmed in detailed, sterically-resolved, time-dependent Monte Carlo simulations.

Publications resulting from this grant:


1. B. Weiner, S. Skokov, and M. Frenklach, "A Theoretical Analysis of a Diamond (100)-$(2 \times 1)$ Dimer Bond," J. Chem. Phys. 102, 5486-5491 (1995).

2. S. Skokov, B. Weiner, and M. Frenklach, "Chemistry of Acetylene on Diamond (100) Surfaces," J. Phys. Chem. 99, 5616-5625 (1995).

3. S. Skokov, B. Weiner, M. Frenklach, Th. Frauenheim, and M. Sternberg, "Dimer-Row Pattern Formation in Diamond (100) Growth," Phys. Rev. B 52, 5426-5432 (1995).

4. M. Frenklach, S. Skokov, and B. Weiner, "On the Role of Surface Diffusion in Diamond Growth," Proc. Fourth Int. Symp. Diamond Materials, edited by Ravi and Dismukes, Electrochemical Society, Pennigton, N.J., 1995, pp. 1-12.

5. S. Skokov, B. Weiner, and M. Frenklach, "Transformation of Monoatomic Steps on (100) Diamond Surfaces," Proc. Fourth Int. Symp. Diamond Materials, edited by Ravi and Dismukes, Electrochemical Society, Pennigton, N.J., 1995, pp. 546-551.

6. S. Skokov, B. Weiner, and M. Frenklach, A Theoretical Study of the Energetics and Vibrational Spectra of Oxygenated (100) Diamond Surfaces, in Diamond for Electronic Applications, edited by D. L. Dreifus, A. Collins, T. Humphreys, K. Das, and P. E. Pehrsson, Materials Research Society, Pittsburgh, 1996, pp. 281-286.

7. M. Frenklach, in Evolution of Epitaxial Structure and Morphology, A One-Dimensional Stochastic Model of Diamond Growth, edited by A. Zangwell, D. Jesson, D. Chambliss, and R. Clarke, Materials Research Society, Pittsburgh, 1996, pp. 83-88.

8. S. Skokov, B. Weiner, and M. Frenklach, Theoretical Study of Oxygenated (100) Diamond Surfaces in the Presence of Hydrogen, Phys. Rev. B 55, 1895-902 (1997).

9. M. Frenklach and S. Skokov, Surface Migration in Diamond Growth, J. Phys. Chem. 101, 3025-3036 (1997).

10. M. Frenklach, Simulation of Surface Reactions, Proceedings of the Ninth International Conference on High Temperature Materials Chemistry (K. E. Spear, Ed.), Electrochemical Society, Pennington, N.J., 1997, pp. 511-522.

11. M. Frenklach, Pure Appl. Chem. Simulation of Surface Reactions, 70, 477-484 (1998).

12. A. Netto and M. Frenklach, MRS Proceedinds, Kinetic Monte Carlo Simulation of Diamond Film Growth with the Inclusion of Surface Migration, in Diffusion Mechanisms in Crystalline Materials, edited by Y. Mishin, G. Vogl, N. Cowern, R. Catlow, and D. Farkas, Materials Research Society, Warrendale, PA, 1998, pp. 383-388.

13. M. Frenklach, Numerical Modeling of Surface Reactions, in Materials Science of Carbide, Nitrides and Borides (Y. G. Gogotsi and R. A. Andrievski, Eds.), Kluwer, Dordrecht, Netherlands, 1999, pp. 119-132.

14. M. Frenklach and Craig S. Carmer, Molecular Dynamics using Combined Quantum and Empirical Forces: Application to Surface Reactions, in Molecular Dynamics of Clusters, Surfaces, Liquids, and Interfaces (W. L. Hase, Ed.), Advances in Classical Trajectory Methods, Vol. IV, JAI Press, Stamford, CT, 1999, pp. 27-63.

## 2 Introduction

Conclusions from a scientific study almost always involve lossy information compression. In processing the data and results for others to use some information is often lost. With proper data structures and computational algorithms in place, it will be possible to avoid unnecessary processing of the results. Our work re-examines the concept of a mathematical model associated with complex, physical systems. We explore the issues that arise when one considers collaborative scientific work involving experimentation on isolated aspects of the system, along with the need to make accurate predictions on the behavior of entire system. We propose an approach to "reverse-reductionism" – namely, how to structure local models so that reliable global predictions can be constructed from combinations of uncertain local models and experiments.

We are motivated by the current state of model-based and experimental research in chemical reaction networks. This field is characterized by several interrelated, relevant facts:

- Processes are complex, though physics-based reasoning leading to governing equations is widely accepted as viable;
- Uncertainty in process behavior exists, but much is known regarding "where" the uncertainty lies in the governing equations;
- Experiments on semi-isolated aspects of processes are effectively carried out;
- Numerical simulations of processes, with uncertain parameters "fixed" to certain values, may be performed reliably, but no closed-form solution is possible.

Each experimental result implicitly contains information about the uncertainty in the process model. Yet, there is no systematic manner in which to combine this information, and no coordination among researchers as to the form by which information and data should be shared. Most of the times, the information transfer occurs at a highly processed level, in the familar "read my paper" mode. As a result of individual data processing, the critical correlations within the data and compound uncertainty of the overall model are simply lost. To our knowledge, the first attempt to address this problem was the GRI-Mech project [26] (a brief description is given in section 4). This project was the first to use IT to systematically collect, consistently combine, rigorously process, and conveniently retrieve " raw" experimental and theoretical information – all inter-related, often in different patterns, through their common dependence on a single multivariable model. The essence of the IT process and its final product was a paradigm of a "living model": at any point in time one has the "best current model", not the ultimately right one — possibly an utopic goal of the present science — but the best possible *today*, individually

consistent with separate research results, and quantified against *all* available data. This proposal goes a step further, extending the ideas and underlying numerical procedures to address an even broader issue: modeling realistic uncertainty in prediction of a diversely-compound model. Explaining our ideas requires reexamination of the concept "model"; and we begin just with such. We then describe the numerical procedures we propose to employ, present some very preliminary results, and end with the proposed plan of action. We emphasize that although the case study we focus on in this proposal is in the area of chemical reaction networks, the methods we develop will apply equally well to any scientific field characterized by the bulleted list above and will benefit from the paradigm we put forth.

### 2.1 Nature of a Predictive Model

Development of predictive models marks human civilization. Ancient models were philosophical in nature and relied entirely on logical deduction; one example of this can be the concept of corpuscular (i.e., atomic) nature of matter arrived to by the Greeks two milleniums ago. Invention of geometry and algebra in the Middle East was motivated by and applied to agricultural problems. Early scientific discoveries were expressed in terms of simple phenomenological laws, like Fick's Law of diffusion and Boyle's Law of gases. Development of calculus expressed these laws in terms of differential equations and formulated solutions to their linearizations. The advent of computer provided means to solve these differential equations in their complete nonlinear formulation, such as Navier Stokes equation of fluid mechanics or Schrodinger equation of quantum mechanics.

Thus, the concept of a model and the "physical" form it assumes have been changing with time: from conceptual statements to simple algebraic relationships to differential equations to numerical algorithms to computer programs and files. The present problems facing our society—such as global warming, earthquake preparedness, safety of transport of nuclear waste, and pollutant emission from automobile engines—call for integration of a variety of computer programs, each solving numerical problems in a different discipline. The overriding concern for such complex models is their reliability: predictability, authenticity, uncertainty, etc. In other words, one has to have a certain degree of confidence to apply these models for political decisions, economic forecast, or design and manufacturing of automotive engines.

The primary difficulty, common to essentially all fields of science and engineering, is the fact that more authentic models introduce larger numbers of parameters. The general expectation is that advancement in science should provide means to establish these parameters with required accuracy. The experience both supports and disproves this expectation. Indeed, the unprecedented advance in scientific instrumentation (e.g., laser spectroscopy) and computer technology (the increase in speed and memory and the reduction in cost) provides powerful means to determine the parameter values on sound experimental and theoretical grounds. At the same time, however, this often (if not always) involves another model, either instrumental or/and theoretical, which in turn introduces additional parameters. The end result is that even most accurately determined model parameters have uncertainties.

The current practice of model development/use is based on the following paradigm: (a) A model is postulated introducing a set of parameters; (b) A "unique" set of parameter values are

determined from experiment or/and theory, and (ideally) supplied along with a corresponding set of individual uncertainties; and (c) the model is then applied to conditions of interest employing the unique set of parameter values. The natural uncertainties of the underlying experiment and theory must somehow be transferred into the final prediction uncertainty using the uncertainties that were assigned to the model parameters. Experience shows that this conventional paradigm does not lead to a desirable quality of prediction. Each model parameter has associated with it an interval of uncertainty. Taken together, the uncertainties of all parameters form a hypercube in the parameter space. Each point of this hypercube does not violate accepted experimental results (which are highly processed) since each coordinate (parameter) individually belongs to it's corresponding interval of uncertainty. But, some parts of the hypercube fit the experimental data base better than others. Typically, the central point of the hypercube, whose coordinates correspond to the individual best-fit values of the individual parameters, is not necessarily the best-fit point for the combined set of parameters. Unknown correlations imply the center is not even a point of maximum-likelihood, although each individual parameter was thought to have maximum likelihood at the center. So, the better-fit parts form a low-dimensional manifold, the result of correlation among the model parameters that were introduced. A methodology that implicitly or explicitly samples preferentially the manifold and not the entire hypercube volume should provide a more realistic estimate of model uncertainty than superposition of individual parameter errors.

### 2.2 New Perspective on Predictive Model

The specific focus of this proposal is representation and propagation of uncertainty of dynamic models. While the suggested approach is applicable to any model, our emphasis is on the most general case: a large-scale dynamic model which does not possess a closed-form solution. The underlying notion of the approach is a shift in the current paradigm of a model: from considering model parameters as a "unique", predetermined values with individual, uncorrelated uncertainties to considering the actual experimental data and theoretical constraints as an integral part of the model, with model parameters playing role of "internal" variables. In this way, the uncertainties of the experimental and theoretical foundation are transferred "directly" into uncertainties of predictions. This direct relationship allows one also to address the reverse problem: to identify which specific data contribute the most to the prediction uncertainty; to determine the required accuracy of an experiment to bring the prediction uncertainty to a given level; or to assess whether a planned experiment will be able to improve the prediction uncertainty.

Here we suggest that the combination of solution mapping and polynomial optimization provide a foundation for evaluation of realistic prediction uncertainties of large-scale dynamic models, and we propose to investigate this approach fully. Our suggested approach lends itself naturally towards a more data-centric approach to model updating.

# 3 Formulation

Throughout this section, we will refer to physical processes, models, experiments, data, etc. Clarification of meanings and interpretation, and notational choices are crucial for understanding. Associated with a physical process $P$, consider several other entities, listed below.

| Symbol | Meaning |
|---|---|
| $Y_P$ | Outcome variable (scalar) of the process $P$ |
| $M_P$ | Mathematical model of $Y_P$ (usually physics-based) |
| $x_P$ | Uncertain parameters in $M_P$ |
| $S_P$ | Surrogate model of $Y_P$ |
| $E_P$ | Experimental realization of $P$ |
| $D_P$ | Experimental data (ie., *measured* outcome) from $E_P$ |
| $\epsilon_P$ | Error tolerance associated with $D_P$ |

Together, the set $\{Y_P, M_P, x_P, S_P; E_P, D_P, \epsilon_P\}$ constitutes a *model of process* $P$. For a given process, some of these items may be missing. As an example, for a given process, we may have a mathematical model, but no available experimental data. When there are many processes under consideration, we usually index them not by name (ie., $P$, $Q$, etc.) but by an integer index (process $1, 2, \ldots, n$).

Our prediction uncertainty concepts involve *composite processes*, which consist of $n+1$ processes, ($P_1, P_2, \ldots, P_n$ and $P_0$) coupled together by the common dependence that their mathematical models have on $m$ uncertain parameters $\{x_1, x_2, \ldots, x_m\}$. Of course, any of the mathematical models may actually depend only on a subset of these parameters. The processes $\{P_i\}_{i=1}^n$ do have experimental data $D_i$, and are considered ancillary, usually associated with specialized laboratory experiments. The *surrogate models* $S_i$ are algebraic functions, reduced from the mathematical model $M_i$ (surrogate models, and the reductions which map differential equations into algebraic functions are described in Section 3.1). $P_0$ is a more complex scenario, without experimental data, and our goal is to reliably predict its outcome. $P_0$ is referred to as "the predicted process," and the $P_i$ are "experiment processes." As with $S_i$, $S_0$ is the reduction of $M_0$ into a surrogate, algebraic function.

Each of the uncertain quantities $x_j$ are known a priori to lie in sets $X_j$, reflecting prior information about the uncertainties. In many cases, the prior information (compactly written as $x \in X$) is not rich enough to bound the possible values of $P_0(x)$ to a range accurately reflecting the compound uncertainty of the entire knowledge base. The many experimental results usually supply correlation, which must be used. Through the surrogate model $S_i$, information about $x$ is obtained implicitly via the constraint

$$\|S_i(x) - D_i\| \leq \epsilon_i \tag{1}$$

A parametric optimization solves for what might be called the "best-fit" parameter, for instance

$$x^* := \arg\min_{x \in X} \sum_{i=1}^{n} \frac{1}{\epsilon_i} \|S_i(x) - D_i\| \tag{2}$$

The minimizer, referred to as "the solution to the inverse problem," yields a forecast $S_0(x^*)$ of the outcome of the process $P_0$. The question is how to obtain realistic bounds of $x^*$.

The viewpoint we take is as follows. Any $x \in X$ which is consistent with **all** of the experiments (ie., satisfying the constraint in (1) for all $1 \leq i \leq n$) yields a possible outcome of $P_0$, namely $S_0(x)$. All such possible outcomes constitute the predicted outcome set of $P_0$. Our goal is to understand the "extremes" of this predicted outcome set.

An appropriate analysis problem is to compute hard bounds, $\bar{H}$ and $\underline{L}$ satisfying

$$\underline{L} \leq L(d,\epsilon) := \min_{\substack{x \in X \\ |S_i(x) - D_i| \leq \epsilon_i}} S_0(x) \qquad \leq \qquad \max_{\substack{x \in X \\ |S_i(x) - D_i| \leq \epsilon_i}} S_0(x) =: H(d,\epsilon) \leq \bar{H} \tag{3}$$

The uncertainty in the model's forecasting quality is

$$A(d,\epsilon) := H(d,\epsilon) - L(d,\epsilon) \leq \bar{H} - \underline{L}$$

These computations answer the question "What are the extreme values of the prediction model which are consistent with results from collections of related experiments?" That one question is the heart of our proposal. Nevertheless, there are additional interesting queries: Will a particular experiment (either new, or the refinement of an existing one) add to the accuracy of the predictive model?, Which subset of experiments (if redone) offer the most potential for improving the predictive model?, How well does an experiment need to be carried out to improve the accuracy of the predictive model by a fixed percent? The strength of our approach is that it can handle all of these questions.

For instance, the latter question could be attacked as follows: suppose experiment $I$ (a particular value of $i$) can be rerun with a guaranteed accuracy of $\tilde{\epsilon}_I < \epsilon_I$. Then the new size of the range of the prediction is guaranteed to be bounded by

$$\begin{aligned} A(d,\epsilon;I,\tilde{\epsilon}_I) \quad := \max_{\substack{x_H, x_L \in X, \tilde{D}_I \\ |S_i(x_H) - D_i| \leq \epsilon_i \\ |S_i(x_L) - D_i| \leq \epsilon_i \\ \left|S_I(x_H) - \tilde{D}_I\right| \leq \tilde{\epsilon}_I \\ \left|S_I(x_L) - \tilde{D}_I\right| \leq \tilde{\epsilon}_I \\ \left|\tilde{D}_I - D_I\right| \leq \epsilon_I}} S_0(x_H) - S_0(x_L) \end{aligned}$$

which is the possible range of $P_0$, if experiment $I$ is rerun, with a new tolerance of $\tilde{\epsilon}_I$, knowing that the outcome will be consistent with the old data, but not knowing the outcome itself. This may be computed before rerunning experiment $I$, and indicate what level of accuracy improvement can be expected. Note that these expressions are as before – constrained maximums and minimums of the prediction model.

Our proposed work centers on how these various functions ($S_i, S_0$ and $H, L$) can be estimated from the underlying uncertain differential equation models ($M_i$ and $M_0$) and data ($D_i, \epsilon_i$). Optimization issues that arise in bounding $H$ and $L$ are outlined in Section 3.2 and will require specific forms of the $S_i$. Transforming the large scale, complex differential equation models $M_i$ and $M_0$ into the reduced functions $S_i$ and $S_0$ is discussed in section 3.1.

### 3.1 Building Surrogate Models via Solution Mapping

The general optimizations posed in Section 3 are computationally intensive even for well-behaved, well-parametrized algebraic functions $S_i$ and $S_0$. The proposed work is further complicated by the fact these functions are not immediately obtained from the governing equations $M_i$ and $M_0$. Consider a dynamic model, $M$, described by a set of first-order nonlinear ODEs,

$$\dot{y}(t) = f(y(t), t, x), \qquad y(0) = y_0 \tag{4}$$

where $x \in \mathbf{R}^m$ is a parameter vector. For most functions $f$, this equation does not possess a closed-form solution, and expressing some property of the solution, $\phi_y$, called *a response* (e.g., the steady-state value, $\bar{y}$, the peak excursion from $y_0, \max_{t\geq 0} \|y(t) - y_0\|$) as a function of the parameter vector $x$ is only possible in a tabular/numerical sense.

The essence of the *solution mapping* technique, [11], [17], [13], is approximation of responses $\phi_y$ in equation (4) by simple algebraic expressions, i.e., $S_y(x) \approx \phi_y(x)$ within a subset $X$ of parameter space $\mathbf{R}^m$, referred to hereafter as *active* parameter subset. The approximating functions $S_y$ are obtained using the general methodology of the response surface technique [3], [4], by means of a relatively small number of computer simulations, referred to as *computer experiments*. They are performed at preselected combinations of the parameter values and the entire set of these combinations is called a *design* of computer experiments. The computer experiments are performed using the complete dynamic model (4) and the functions $S_y$ obtained in this manner are referred to as *surrogate* model.

Once developed, the surrogate model $S_y$ replaces $\phi_y$, the solution of the original dynamic model, whenever evaluation of $\phi_y$ at a given $x \in X$ is required. There is in principle no restriction on the mathematical form of the surrogate model. In our work we have used quadratic functions for $S_y$, with coefficients determined via computer experiments arranged in a special order, called *factorial design*. These designs originate from rigorous analysis of variance, with the objective of minimizing the number of computer experiments to be performed to gain the information required [3], [4]. Factorial designs have found extensive use in experimental and process development work, and have begun recently being applied to computer experiments (e.g., [24] and [18]). The use of a polynomial form for the surrogate model also ties in closely with our optimization formalism put forth next in section 3.2.

### 3.2 Optimization Techniques/Main computational sub-problem

If all of the surrogate models are affine, then the quanitites defined in section 3 are linear programs, easily solved for large dimensions. However, it is often the case that an affine function is not adequate to represent the dependence. Hence, a natural, nontrivial, and useful extension is to consider quadratic surrogate models. These enlarge the applicable domain of a surrogate model (in chemical reaction networks, recent experience indicates that quadratic surrogate functions are suitable), but introduce nonconvexity and computational complexity into subsequent predictions. In this section, we outline the research directions we will pursue regarding prediction uncertainty estimation from these types of models.

The initial computational subproblem we focus on is an indefinite quadratic program: for $x \in \mathbf{R}^n$,

$$\text{bound } \begin{bmatrix} 1 \\ x \end{bmatrix}^T M_0 \begin{bmatrix} 1 \\ x \end{bmatrix} \quad \text{subject to } \begin{bmatrix} 1 \\ x \end{bmatrix}^T M_i \begin{bmatrix} 1 \\ x \end{bmatrix} \in [\alpha_i, \beta_i] \tag{5}$$

for $i = 1, 2, \ldots, N$. Here, all $M_i$ are symmetric $(1+n) \times (1+n)$ real matrices. There are two convex relaxations (different optimizations, with known relation to original problem, e.g. an upper bound, and better computational properties, namely convex) of this problem that we will begin our work with. They are referred to as the $\mathcal{S}$-procedure (separating hyperplane) and the SOS (sum of squares) procedure. Although they apply to the original problem in (5), these methods are more easily described for a modified feasability problem: *Given $n \times n$ symmetric matrices $M_0, M_1, \ldots, M_N$, determine if the constraint*

$$\bigcap_{i=1}^{N} \left\{ x : x^T M_i x \geq 0 \right\} \subset \left\{ x : x^T M_0 x \geq 0 \right\}$$

*holds.*

In the $\mathcal{S}$-procedure, the computation to ascertain the containment is: If there exist nonnegative real numbers $\{\lambda_i\}_{i=1}^N$ with

$$M_0 - \sum_{i=1}^{N} \lambda_i M_i \succeq 0 \tag{6}$$

then the containment is guaranteed. Here, $A \succeq 0$ means $A$ is a symmetric, positive semi-definite matrix. The new feasability problem, referred to as the "relaxation," is to search over the $\{\lambda_i\}$ to satisfy (6). While only a sufficient condition (hence, a relaxation) for containment, the feasable set of $\{\lambda_i\}$ is convex, and there is a great deal of current research (refered to as Semi-Definite Programming (SDP) or Linear Matrix Inequalities (LMI)) in developing and improving polynomial-time algorithms to find feasable points, or prove infeasability, [2], [9], [5].

In the SOS-procedure, the computation to ascertain the containment is: If there exist symmetric matrices $\{Q_i\}_{i=1}^N$ and scalars $\{r_{ij}\}_{i,j=1}^N$, with $Q_i \succeq 0$ and $r_{ij} \geq 0$, such that the quartic

$$x^T M_0 x - \sum_{i=1}^{N} x^T Q_i x x^T M_i x - \sum_{i=1}^{N} \sum_{j=1}^{N} r_{ij} x^T M_i x x^T M_j x$$

is a sum-of-squares, then the containment is guaranteed. Moreover, the search for such $Q_i$ and $r_{ij}$ is a convex feasability problem, specifically an SDP/LMI, of the form in (6), but with different data, and more (than $N$) variables. Mathematically, the SOS procedure easily extends to higher order polynomial constraints and objectives than the quadratic forms in (5). The SOS methodology can be traced back to [1], [29] for nonquadratic Lyapunov function generation, and [22] for feasability calculations involving polynomial constraints. In the case of quadratic polynomials, the SOS procedure has been shown to often be superior to the simple-minded $\mathcal{S}$-procedure.

We recognize that the problem in (5) includes (as a special case) maximization of an indefinite quadratic function subject to indefinite quadratic constraints, and as such is computationally complex, [27]. We will not fully address or attempt to solve this complexity issue. Rather,

we will simply aim for algorithms which do well bounding the optimizations for problems we encounter. In section 5, we outline specific technical approaches that we will pursue.

# 4 Natural Gas Example

The viability of our proposal is evident from a small, initial study we have conducted on the natural-gas combustion models which form the *GRI-Mech Data Set*.

The term *GRI-Mech*, by itself, refers to a mathematical model for pollutant formation in combustion of natural gas [14], [26]. This mathematical model, in its latest form, is the detailed chemical kinetics describing 325 reaction steps of 53 chemical species. The dynamics is governed by 53 coupled ordinary differential equations, collectively containing 102 uncertain parameters.

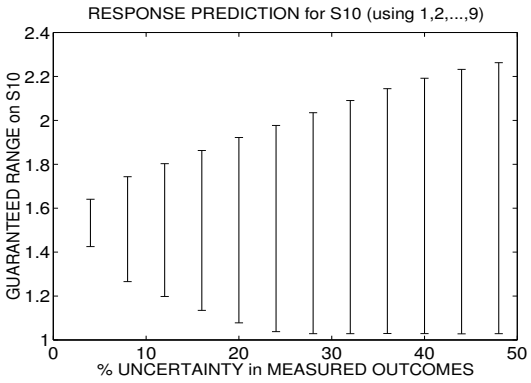
The GRI-Mech Data Set consists of a wide variety of 77 process/experiments, all linked through the GRI-Mech mathematical model. Some of the experimental targets included laboratory flames, whose numerical simulation required coupling of chemical kinetics with mass and heat transfer, in which case evaluation of an individual target was on the order of 20 to 60 mins. Using solution mapping/factorial design, and numerical "virtual" experiments, quadratic surrogate models have been developed for each of the 77 processes, representing the effect that the 102 uncertain parameters have on the (scalar-valued) outcome of each process. The uncertain parameters have been normalized, and are each "known" a-priori to lie in the unit interval $[-1\ 1]$. Each of the 77 processes also have associated experimental results – a measured outcome and corresponding error tolerance – performed by experts throughout the world. This wealth of information about several inter-related complex processes provides an excellent opportunity to test and validate the bounding techniques described in section 3.2. Here we describe a modest, preliminary study we have carried out in this vein. Our specific goal in this section is to predict the possible range of process $P_{10}$ using the information implicit in the models and experimental results associated with processes $P_1, P_2, \ldots, P_9$.

Surrogate models $S_1, S_2, \ldots, S_9$ together involve 17 of the 102 uncertain parameters (ie., 85 of the uncertain parameters play no appreciable role in the outcome of processes $1, 2, \ldots, 9$). Surrogate model $S_{10}$ depends on 11 of these 17 parameters, and none of the other 85.

Using *only* the unit-interval bound on the uncertain parameters, the $\mathcal{S}$-procedure bounds the values taken on by surrogate model $S_{10}$ to lie in $[1.03\ 2.26]$. Interestingly, the accepted experimental value is about 1.5, consistent with this simple prediction on the range.

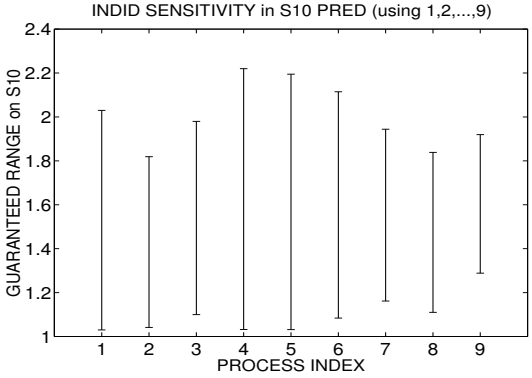
On the other hand, if we use the information known to us regarding processes 1-9, distilled into the corresponding surrogate models along with the experimental results and experimental tolerances, we can refine the range of surrogate model $S_{10}$ by bringing into consideration parameter correlation implicit in the GRI-Mech mathematical model. As a simple illustration, we assumed a common percentage error tolerance on all experimental results of processes 1-9, and used a single $\mathcal{S}$-procedure calculation to yield a hard upper bound on the value that could be taken on by $S_{10}$, consistent with the information regarding 1-9. A nearly identical calculation yields a hard lower bound. The results for percentage uncertainty in measured outcome ranging from 4 to 48% are discussed next.

At nearly 50% uncertainty level, the experimental data from processes 1-9 offers little additional information in bounding the response of $P_{10}$ beyond the apriori [1.03 2.26]. At 4% though, the possible range of $P_{10}$ has been narrowed to [1.43 1.64]. Recall, it is not known whether these are the true possible ranges, or if more sophisticated analysis techniques (e.g., the SOS formulation) could further narrow the guaranteed intervals of containment.

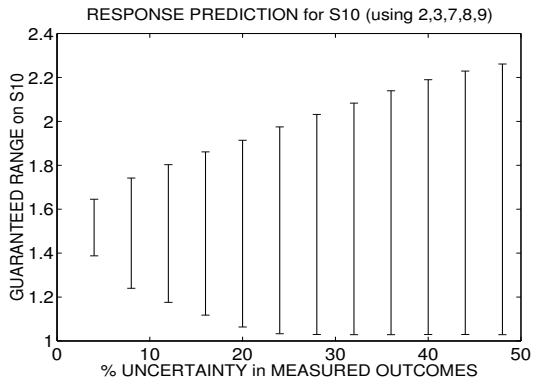


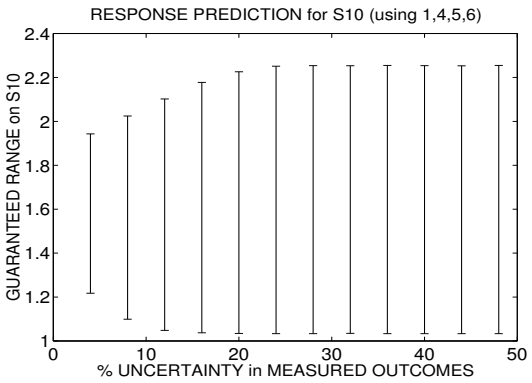

At 3% (not shown), the $\mathcal{S}$-procedure detects an inconsistency between the surrogate models 1-9, experimental data, and experimental tolerance, meaning that no parameter set in the unit-hypercube exists that can explain the data.

By considering one experimental process at a time, we can crudely estimate the sensitivity of our range prediction for $S_{10}$ using each of the separate experimental data from processes $1-9$. The bounds are shown, assuming 5% experimental tolerance in the data. At that uncertainty level, experimental results 2, 3, 7, 8 and 9 **individually** reduce the size of the prediction interval for $S_{10}$ more effectively than 1, 4, 5, and 6.




With this in mind, we repeat the original $S_{10}$ prediction, using two separate groups of information: processes $\{2,3,7,8,9\}$ and processes $\{1,4,5,6\}$. These are shown below, effectively tying the individual sensitivity results to calculations involving multiple sources of information. As can be seen, the results support the previous finding. Of course, the opposite could have been true for a different set of circumstances – although 1, 4, 5 and 6 provided little information individually, it would have been possible that together, they provided more than 2, 3, 7, 8 and 9 provided together. That type of situation is very interesting, and we hope to develop effective coupled sensitivity calculations to identify which information will be most relevant in the prediction task.



This small example already demonstrates the power of the ideas underlying our approach and the feasability of its practical implementation.

# 5 Work Plan

We will spend a great deal of time researching computational issues associated with these methods. Specific to the ideas in section 3.2:

1. Often times, the $M_i$ are low rank, and sparse. This is what Box, [4], refers to as "effect sparsity." We will develop an SDP/LMI solver for the $\mathcal{S}$-procedure relaxation of CSP that exploits sparse and low rank $M_i$.

2. As mentioned, the SOS relaxation is usually more powerful than the simple $\mathcal{S}$ procedure. We will study the structure of SDP/LMI for SOS formulation of CSP for sparse and low rank $M_i$.

3. Study methods for effective problem decomposition. An obvious approach is to divide a constraint interval into a union of two intervals

$$[\alpha_I, \beta_I] = [\alpha_I, m_I] \bigcup [m_I, \beta_I]$$

   which creates 2 problems, from a single original problem. The main question is how to pick $I$, and $m_I$. A generalization divides the domain by appending to the original problem a constraint of the form $f(x) \leq 0$, as well separately appending to the original problem the constraint $f(x) \geq 0$. The union of the ranges that the separate optimizations yield is guaranteed to contain the desired range.

Many aspects of our proposed methods are parallelizable in the most obvious manner. Thus, our methods are highly scalable, and suitable for an Internet-based, distributed computing environment.

1. The solution mapping technique (reducing $M_i$ into $S_i$ for $(i = 1, 2, \ldots, m)$ must be applied to several differential equations models. Moreover, for each model, numerous runs are required to explore the parameter space and complete the solution mapping.

2. Divide-and-Conquer to estimate $H$ (and $L$ and variants): The divide & conquer algorithms, by their very nature, split problems up into (essentially) independent subproblems.

We will develop scheduling and distribution rules appropriate for these computations, in order to gain maximum benefit from the simultaneous processing capability.

Case studies play an important role in validating the ideas. We will continue to use the GRI-Mech data to study small (as in the previous example) to medium-scale (the whole GRI data set) problems. In performing these case studies, we will develop, and make available to all, prototype algorithms and documentation for single processor and multiprocessor machines. We will develop interfaces for multiuser operation over the Internet, thus expanding and testing the on-line optimization capabilities of the GRI-Mech `www` site.

# 6 Goals and Future Expectations

Our goal, within the scope of this proposal, is to investigate the concepts and ideas we have described. Our success will establish the main impetus for the creation of a complete infrastructure for IT in support of the overall goal – linking researchers, software developers, engineers, and policy makers with a science-centric information management system that we envision is necessary to move science and engineering forward.

Based on the framework presented, one can envision a public database for scientific knowledge. Much like a handbook of physical constants, it would be a repository of related processes, their mathematical models, and (if available) experimental data. Yet, unlike a handbook of constants or compilation of "recommended" data, the proposed database framework will be dynamic in its very nature: the "stored" information will be the result of processing (exhaustive and rigorous optimization) of all data available, and repeated every time a new data is contributed.

The ideas outlined in sections 2-3.2 suggest an infrastructure whereby a user would *contribute* or *query* the database in the following manner:

- Build numerical simulation model, $M_P$, of process, $P$. Simulation model should have, as adjustable parameters, those parameters $x$ for which uncertainty remains.
- Perform extensive numerical experiments, over accepted ranges of parameters. Use factorial design to sample the parameter space. Create surrogate model $S_P$ using solution mapping method, generating a (quadratic, say) function representation of the modeled outcome as a function of the uncertain parameters.
- Depending on whether we are considering a *contribution* or *query* the tasks differ:
  - For a *contribution*: perform experiment $E_P$, measure outcome, $D_P$, and assess potential error, $\epsilon_P$. Submit *surrogate model*, *measured outcome*, and *error bound* to database.
  - For a *query*: request `PossibleRange` of surrogate model $S_P$, based on available related surrogate models, experimental data and error bounds.

The prototype algorithms, mentioned in section 5, will be wrapped into a working database of this envisioned form, allowing us to illustrate to others the potential impact the combination of our ideas may have.

# 7 Incorporating Research into Undergraduate Education

We (Frenklach and Packard) have developed and are now beginning to teach a new course for upper division undergraduates, E177, entitled "Advanced Programming with Matlab." It is targeted at junior and senior engineering students. We will include a 1-week case study based on the proposed work, and also cover the architecture of our prototype code. By the end of the project, we will have a small undergraduate research course where students carry out a full analysis on a problem area of their choice, using our prototype algorithms.

## 8 Integrating Diversity

The College of Engineering's CUES (Center for Underrepresented Engineering Students) has a close alliance with Mechanical and Electrical Engineering. We will work with Director Carla Trujillo to identify interested students associated with the Center. The center also connects us to the Julia Morgan Engineering Program (aimed at increasing and maintaining female students in engineering) and Graduate Academic Diversity Program (GrAD), which is aimed at increasing and maintaining underrepresented graduate students in engineering.

## 9 Collaborative Nature

This proposal is the first product of collaboration initiated recently between the PIs. Although both PIs hold faculty appointments in the same Department, their expertise and research activities span very different disciplines: Control (Packard) and Combustion Chemistry (Frenklach). The proposed work merges the convex relaxations from control and system theory, [8], [25], [10], [16], [19], [7], [21], for robustness analysis with the technique of solution mapping developed and applied to numerical modeling of chemical kinetics typical of fossil-fuel combustion, [11-14, 17, 26]. Its promise and ultimate success will heavily rely on the cross-fertilization of these two different yet apparently complementary fields.

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