

1 Introduction

In the context of chemical kinetics, model reduction typically focuses on the following general problem. Consider a detailed model defining a set of chemical reaction processes occurring with transport in a spatial region $\Omega \subset \mathbb{R}^N$, where $N = 1, 2$, or 3 . Let N_S be the number of chemical species present in this model and let N_R be the number of chemical reactions present in this model. In its most general form, we are trying to solve

$$\begin{bmatrix} \dot{T} \\ \dot{\mathbf{y}} \end{bmatrix} = \Theta(T, \mathbf{y}) + \Gamma(T, \mathbf{y}) \quad (1a)$$

where dots denote time derivatives, the temperature is defined by the function $T : \Omega \times (0, t_f) \rightarrow \mathbb{R}$, $T \in L^2(\Omega \times (0, t_f))$, and the species mass fractions are defined by the function $\mathbf{y} : \Omega \times (0, t_f) \rightarrow \mathbb{R}^{N_S}$, $\mathbf{y} \in (L^2(\Omega \times (0, t_f)))^{N_S}$. Both Θ and Γ are maps on function spaces, where in this case, $\Theta, \Gamma : (L^2(\Omega \times (0, t_f)))^{N_S} \rightarrow (L^2(\Omega \times (0, t_f)))^{N_S}$. Here, Θ is a map describing transport processes (such as convection and diffusion), and Γ is a map describing the changes in state variables due to chemical reaction processes.

Currently, the form of Θ is not important; model reduction techniques applied to chemical kinetics do not handle transport processes well. Few of them treat transport directly, and the rest sidestep the issue by assuming that when the system (1) (possibly supplemented by other PDEs, algebraic equations, appropriate boundary conditions, and appropriate initial conditions) is solved, the PDEs are solved using Strang (operator) splitting. Strang splitting separates the PDE integration problem into integration of the transport problem without reaction (that is, assuming that $\Gamma = 0$), and integration of the stiff chemistry/reaction problem without transport (that is, assuming that $\Theta = 0$). Typically, the procedure is to decompose a full PDE integration time step into a half time step of transport integration, a full time step of chemistry integration, and then a half time step of time step of transport integration. When carrying out this Strang splitting procedure, the chemistry integration time steps are most costly (empirically, they account for up to 90% of the CPU time), and it is these integrations to which model reduction is applied.

During a chemistry integration step, (1) reduces to a large number of instances (equal to the number of grid points in the spatial discretization of (1)) of the ODE

$$\begin{bmatrix} \dot{T} \\ \dot{\mathbf{y}} \end{bmatrix} = \Gamma(T, \mathbf{y}). \quad (2)$$

Typically, during a chemistry integration step, we assume that chemical reactions at each point in Ω occur under adiabatic-isobaric conditions (because we are neglecting transport processes), in which case the model equations are

$$\dot{T} = \frac{\sum_{j=1}^{N_S} M_j h_j(T) \sum_{i=1}^{N_R} \nu_{ji} r_i(\mathbf{y}, T)}{\rho(T) \widehat{C}_P(\mathbf{y}, T)}, \quad (3a)$$

$$\dot{y}_j = \frac{M_j \sum_{i=1}^{N_R} \nu_{ji} r_i(\mathbf{y}, T)}{\rho(T)}, \quad j = 1, \dots, N_S. \quad (3b)$$

The terms in (3) are as follows: M_j is the molar mass of species j , h_j is the ideal gas enthalpy of species j , ν_{ji} is the stoichiometric coefficient of species j in reaction i , ρ is the mass density of the mixture, \widehat{C}_P is the ideal gas mixture heat capacity, and r_i is the molar rate of production of reaction i . The terms \widehat{C}_P , r_i , and h_j are generally nonlinear functions of their arguments, and the main source of nonlinearity is r_i , which typically takes the form

$$r_i(\mathbf{y}, T) = k_i(\mathbf{y}, T) \prod_j C_j^{\nu_{ji}}, \quad (4)$$

where C_j is the concentration of species j (obtained by converting from the mass fraction of species j using available state variables), and k_i is a rate coefficient that is an exponential function of temperature (and possibly also species concentrations, depending on the reaction).

2 Current Work

My work has focused primarily on simultaneous reaction and species elimination and affine lumping (which is a projection-based method). Since projection-based methods have been discussed in previous MRG summaries, I'll focus primarily on reaction and species elimination.

Simultaneous reaction and species elimination determines a reduced model from a detailed model by selecting a subset of species and reactions from the detailed model and eliminating all other species and reactions, such that the right-hand side of the detailed model differs from the right-hand side of the reduced model by less than a specified error tolerance at a set of user-supplied reference points. To determine the subset of species and reactions that are retained in the reduced model, simultaneous reaction and species elimination uses the solution $(\mathbf{z}^*, \mathbf{w}^*) \in \{0, 1\}^{N_R} \times \{0, 1\}^{N_S}$ of an integer linear program (ILP). If $w_j = 1$, species j is retained in the reduced model; if $w_j = 0$, species j is excluded from the reduced model. Similarly, if $z_i = 1$, reaction i is retained in the reduced model; if $z_i = 0$, reaction i is excluded from the reduced model.

Constraints on the ILP result in the following reduced model approximating (3):

$$\dot{T} = \frac{\sum_{j=1}^{N_S} M_j h_j(T) \sum_{i=1}^{N_R} \nu_{ji} z_i^* r_i(\mathbf{y}, T)}{\rho(T) \hat{C}_P(\mathbf{y}, T)}, \quad (5a)$$

$$\dot{y}_j = \frac{M_j \sum_{i=1}^{N_R} \nu_{ji} z_i^* r_i(\mathbf{y}, T)}{\rho(T)}, \quad j \in \{j : w_j^* = 1\}, \quad (5b)$$

$$\dot{y}_j = 0, \quad j \in \{j : w_j^* = 0\}. \quad (5c)$$

We can solve (5), replacing the equations $\{\dot{y}_j = 0 : j \in \mathbb{N}, w_j^* = 0\}$ with the constants $\{y_j = y_{j,0} : j \in \mathbb{N}, w_j^* = 0\}$. We could either take $y_{j,0}$ from the initial conditions accompanying (5), or we could set $y_{j,0} = 0$ (provided the ILP has appropriate constraints). These replacements decrease the number of ODEs that need to be solved during a chemistry integration, decreasing the CPU cost of a chemistry integration step.

Reaction elimination alone is similar to simultaneous reaction and species elimination; it is like setting $w_j^* = 1$ for all $j \in \mathbb{N}$, but a detailed description of the method is beyond the scope of this document.

3 Challenges

Some of the main challenges of model reduction in chemical kinetics are:

- Developing model reduction techniques that generate reduced models that properly reproduce the results of the full PDE in (1) to within user-supplied error tolerances
- Developing methods that propagate any error bounds on time derivatives in a model reduction method (projection-based methods and species and reaction elimination are two examples) through a numerical method to determine bounds on the difference between the solution of the reduced model and the solution of the original detailed model
- Develop model reduction techniques that more aggressively reduce the system of ODEs in (3)
- Develop theory for model reduction techniques so that many techniques share a common theoretical basis so that they can be more easily compared; there are lots of model reduction techniques out there, but it's not necessarily clear which techniques work best for different problem instances