

# Mechanism Reduction as a Preconditioning Strategy for Integrating Large Chemical Kinetic Systems

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**Abstract**— Mechanism reduction techniques are used to develop a class of adaptive preconditioners for integrating the ordinary differential equations (ODE) associated with large chemical kinetic mechanisms. The new adaptive preconditioners reduce the computational cost by several orders of magnitude relative to traditional dense solution methods for a detailed 2-methylnonadecane mechanism ( $C_{20}H_{42}$  with 7172 species).

## I. INTRODUCTION

Preconditioners are sought to increase the convergence rate of the iterative linear system solver embedded within the implicit ODE solver. In particular, the linear system solver is used for every iteration of the Newton-Raphson nonlinear solver, which computes the chemical state at future timesteps in the implicit discretization of the ODEs [1]. A good preconditioning matrix for ODE solvers is one that captures the dominant behavior of the system evolution, but is significantly easier to solve than the complete system [2].

The Jacobian matrices associated with the ODEs of reduced mechanisms are thus ideal candidates for use as preconditioners. A preconditioner need only serve as an approximation to the true ODE system, which is then improved by the iterative linear and non-linear solvers to achieve the same solution as direct methods (to within a user-specified accuracy). This means that the reduced mechanism selected for the preconditioner can be smaller and lower in fidelity than the standard approach to global mechanism reduction. Further, the reduced mechanism must only be accurate for the chemical state over the duration of an individual timestep, which means even greater reduction is possible by allowing the preconditioning mechanism to adapt. This feature, referred to as adaptive preconditioning, allows the integration of detailed chemical kinetic mechanisms to occur at a cost

comparable to methods using on-the-fly mechanism reduction without any loss of accuracy.

## II. APPROACH

A number of adaptive preconditioners are tested using several different reduction heuristics. The simplest reduction technique is based on removing reactions that have a slow characteristic timescale relative to the integrator timestep. To filter out the slow reactions in the preconditioner matrix  $P$ , a threshold operation is applied to the Jacobian matrix  $B$  of the backward differentiation formulas (BDF) for the stiff ODE integrator. Specifically,

$$P_{ij} = \begin{cases} B_{ij} & \text{if } i = j \text{ or } |B_{ij}| > \eta \\ 0 & \text{otherwise,} \end{cases} \quad (1)$$

where  $\eta$  is a nondimensional, user-specified threshold. A more complete description of the approach to reducing the mechanism in this preconditioner is found in [3].

Other preconditioners are also tested as part of this investigation, but none have been found so far to produce a measurably faster method than the simple reaction filtering approach in (1). As a consequence, the computational timings for these other preconditioners are not discussed in detail. The filter metrics for these other preconditioners include: (i) the species coupling frequency normalized by the destruction rate (i.e., row-normalization); (ii) the species coupling frequency normalized by the perturbation species destruction rate (i.e., column normalization); (iii) the maximum and minimum of metrics (i) and (ii); and (iv) the species coupling metric used in the technique in [4] for mechanism reduction via directed relational graph with error propagation (DRG-EP) analysis. It is interesting to note that filtering by the DRG-EP metric offers large reductions in the preconditioner size near equilibrium. However, the performance is not consistent over the range of conditions tested due to convergence problems. Further investigation is necessary to apply this more sophisticated preconditioner in a manner that is able to systematically avoid the non-linear convergence errors currently observed.

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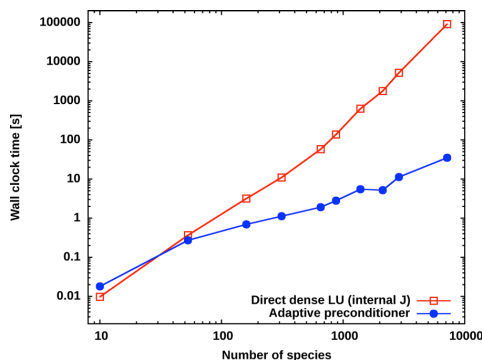


Fig. 1. Comparison of the computational cost between the new adaptive preconditioner approach (blue solid circles) and the traditional ODE solver using dense matrix methods (red empty squares).

### III. TEST CASES

The average computation times are compared between two approaches for integrating a constant volume, homogenous reactor model. The first approach is based on the traditional stiff-ODE solver methods first used in chemical kinetics. This approach is still found in some multi-dimensional computational fluid dynamics codes (e.g. KIVA-MZ [5]). The second approach is based on the adaptive preconditioner (1) using the optimized integrator and sparse matrix settings developed in [6].

A total of ten different mechanisms are tested ranging in size from hydrogen (10 species [7]) to 2-methylnonadecane (7172 species [8]). The initial compositions are a stoichiometric mixture of fuel and idealized air (79%  $N_2$  and 21%  $O_2$ ) at an initial pressure of 20 atmospheres. Between eight and twenty-five initial temperatures are tested ranging from 650 to 1450 Kelvin for each mechanism. The evolution of each mixture composition is simulated for one second of physical time, and only those initial conditions leading to auto-ignition are included in the average computation time reported. A more complete description of the test cases is found in [3] along with a full reference list and optimum threshold values  $\eta$  found for each mechanism.

### IV. RESULTS

The average computation time is obtained for each approach using a single thread of an Intel Xeon E5620 processor (2.4 GHz clock speed with 1.33 GHz DDR3 ECC RAM). The timing results are shown in Fig. 1. For the largest mechanism tested (2-methylnonadecane, 7172 species), the adaptive preconditioner has an average solution time of 32 seconds compared to  $10^5$  seconds (more

than one day) for the traditional approach, which represents three orders of magnitude of computational speedup. It is important to stress that while the mechanism is effectively reduced at each timestep by the filtering operation for the preconditioner (1), the mechanism governing the ODE system is unchanged. Consequently, the ignition delay times, temperatures and major species mass fractions (greater than  $10^{-6}$ ) obtained through the two approaches agree to a minimum of six decimal places for the user-specified relative tolerance of  $10^{-8}$ . The mass fractions of trace species (between  $10^{-12}$  and  $10^{-6}$ ) are found to agree to within four decimal places. The computational speedup shown in Fig. 1 therefore occurs with no practical loss in accuracy.

For the smaller mechanisms, hydrogen (10 species) and methane (53 species) the Jacobian matrix is sufficiently dense that the adaptive preconditioner is comparable to the traditional approach. In fact, for the hydrogen mechanism the dense approach is approximately two times faster. To avoid this penalty, a metastrategy can be developed that combines the two approaches and automatically selects the best method based on mechanism size or through automatic tuning when the mechanism information is first read into the simulation.

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