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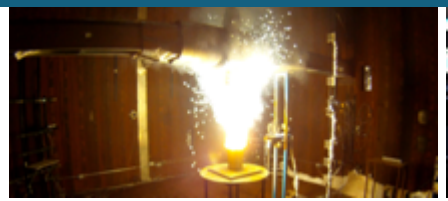

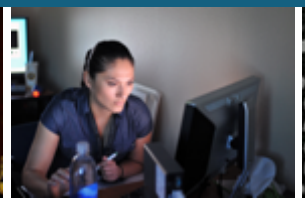

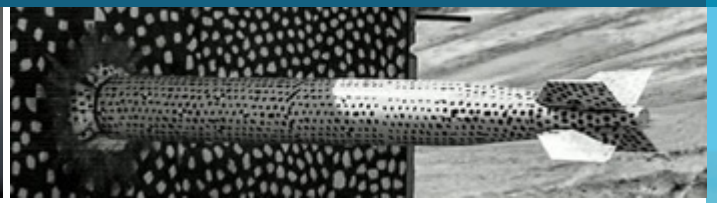

TChem – An Open Source Computational Chemistry Software Library for Heterogeneous Computing Platforms

Oscar Diaz-Ibarra, Kyungjoo Kim, Habib Najm, Cosmin Safta

U.S. DEPARTMENT OF ENERGY | NNSA National Nuclear Security Administration

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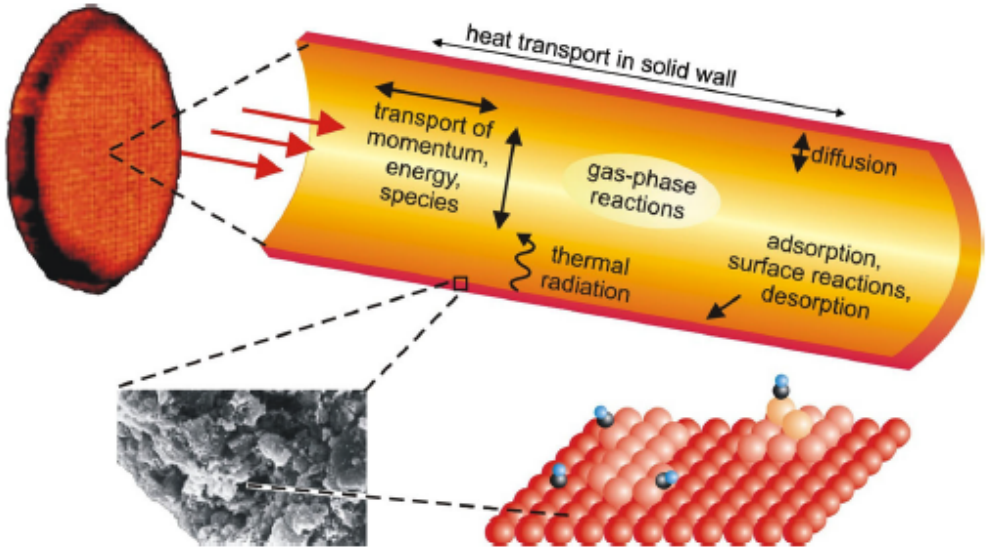
Motivating Example

- Chemkin/Cantera/TChem provide tools to compute thermo-chemical properties for detailed chemical reaction mechanism (gas-phase and surface species).
- Additionally, TChem offers…
  - numerical/analytical Jacobians (problem dependent) to analyze reaction mechanisms
  - portability across heterogeneous computing platforms via Kokkos.

https://melscience.com/AU-en/articles/interaction-methane-oxygen-combustion-reaction/

Provide Computational Tools for Gas-phase Chemistry and Catalysis Studies

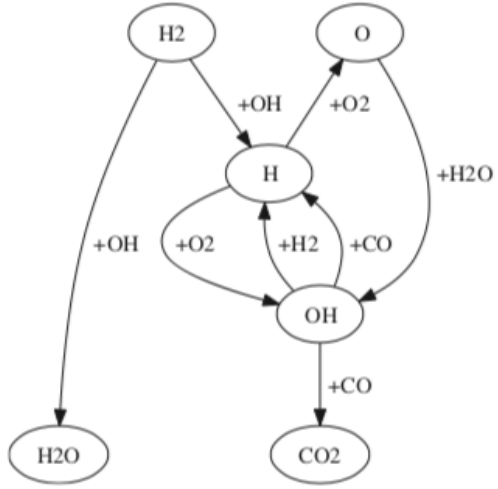


Olaf Deutschmann, Catal Lett (2015) 145:272–289

Surface and Gas-Phase Reactions in a Channel

$$\mathrm{H + O_2 + (M) \rightarrow HO_2 + (M)}$$
$$\mathrm{H + OH + (M) \rightarrow H_2O}$$
$$\mathrm{OH + H_2 \rightarrow H + H_2O}$$
$$\mathrm{CO + OH \rightarrow CO_2 + H}$$

Reaction Mechanism



Reaction Network

Fragment of hydrocarbon reaction mechanism from Prager and co-authors (Combustion and Flame 158 (2011) 2128–2144)

Kinetic Model Example

Overview of TChem

The TChem software is a toolkit for computing **thermodynamic properties**, **source term** and **Jacobian** for chemical kinetic models that involve gas and surface reactions.

TChem functionality:

- Thermodynamic properties based on NASA polynomials.
- Specific production/consumption rates for both gas and surface chemistry.

Chemical reactor models:

- Constant Pressure Gas-Phase Ignition (0d Ignition)
- Constant Volume Gas-Phase Ignition
- Continuously Stirred Tank Reactor (CSTR)
- Plug-Flow Reactor (PFR)

Jacobian of reactor models w.r.t the state vector:

- Analytic Jacobian using the automatic differentiation (AD) tool provided by the SACADO library
- Numerical Jacobian using three finite difference schemes (forward, central and Richardson's Extrapolation) and adaptive differencing size.
- Hand-derived Analytic Jacobian only for Constant Pressure Gas-Phase Ignition reactor

For example, a Plug Flow Reactor (PFR) with gas phase and surface chemistry solves the following system of Differential Algebraic Equations (DAEs).

- ***Species equation***

$$\frac{dY_k}{dz} = \frac{1}{\rho u}\dot{w}_k W_k + \frac{P_r^{'}}{\rho u A_c}\dot{s}_k W_k - \frac{P_r^{'}}{\rho u A_c} Y_k \sum_{k=1}^{Kg} \dot{s}_k W_k$$

- ***Energy equation***

$$\frac{dT}{dz} = -\frac{1}{\rho u c_p}\sum_{k=1}^{Kg}\dot{w}_k W_k h_k - \frac{P_r^{'}}{\rho u A_c c_p}\sum_{k=1}^{Kg}\dot{s}_k W_k h_k$$

- ***Momentum equation***

$$\frac{du}{dz} = -\gamma\frac{P_r^{'}}{\rho A_c}\sum_{k=1}^{Kg}\dot{s}_k W_k - \frac{R}{um}\left(\frac{1}{\bar{W}}\frac{dT}{dz} + T\sum_{k=1}^{Kg}\frac{dY_k}{dz}\frac{1}{W_k}\right)$$

Where $\gamma = \frac{1+\frac{p}{\rho u^2}}{1-\frac{p}{\rho u^2}}$ and $m = 1 - \frac{p}{\rho u^2}$.

- ***Continuity equation***

$$\frac{d\rho}{dz} = \frac{P_r^{'}}{u A_c}\sum_{k=1}^{Kg}\dot{s}_k W_k - \frac{\rho}{u}\frac{du}{dz}$$

- ***Algebraic constraint***

$$\dot{s}_k = 0 \quad k \text{ correspond to surfaces species}$$

TChem – v2.0

TChem was originally written in “C” and optimized for solving a set of problems sequentially.

New features in Version 2.0:

- Refactored in C++ and Kokkos to exploit modern heterogeneous computing platforms
  - Multi-level parallelism (coarse grain task parallelism using streams, batch parallelism and team parallelism)
- CMAKE build system
- Added functionalities for surface chemistry
- Numerical Jacobians and SACADO analytic Jacobians for all reactor models
- YAML input parser
- Python interface

TPLs in TChem:

- Kokkos, SACADO, Tines, YAML-CPP, pybind11, and OpenBLAS (only for host computations)

TChem Overview

Inputs

- Kinetic mechanism files: Chemkin format or Yaml-Cantera format
- Operating conditions: text file
- Modification Parameters: text file
- Numerical parameters: Command line arguments

Interface

C++ Batch Parallel Interface *for*
Source terms, Jacobians, rate of progress, S-matrix, net production rates, thermodynamic properties, and reactors (PFR, homogenous batch reactor , constant volume batch reactor, and CSTR)

PyTChem: *Python interface running TChem on heterogeneous computing platforms for* homogenous batch reactor and net production rates

Core functions

Thermodynamic Properties (NASA Polynomials):
Enthalpy, entropy, heat capacity, internal energy.

Chemical kinetics (gas and surface):
Production rates, rate of progress and rate constants.
Support for gas reaction types: Elemental, pressure-dependent (Lindeman, Troe, SRI), third-body, and PLOG.
Support for surface reaction type: Elemental, stick, and surface coverage modification.

TChem Overview

Inputs

```
T P SPECIES_NAME1 SPECIES_NAME2 ... SPECIES_NAMEN
T#1 P#1 Y1#1 Y2#1 ... YN#1 (sample #1)
T#2 P#2 Y1#2 Y2#2 ... YN#2 (sample #2)
...
...
...
T#N P#N Y1#N Y2#N ... YN#N (sample #N)
```

- Operating conditions: text file
- Modification Parameters: text file
- Numerical parameters: Command line arguments

UQ studies

Interface

C++ Batch Parallel Interface *for*
Source terms, Jacobians, rate of progress, S-matrix, net production rates, thermodynamic properties, and reactors (PFR, homogenous batch reactor , constant volume batch reactor, and CSTR)

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Reacting flows

Core functions

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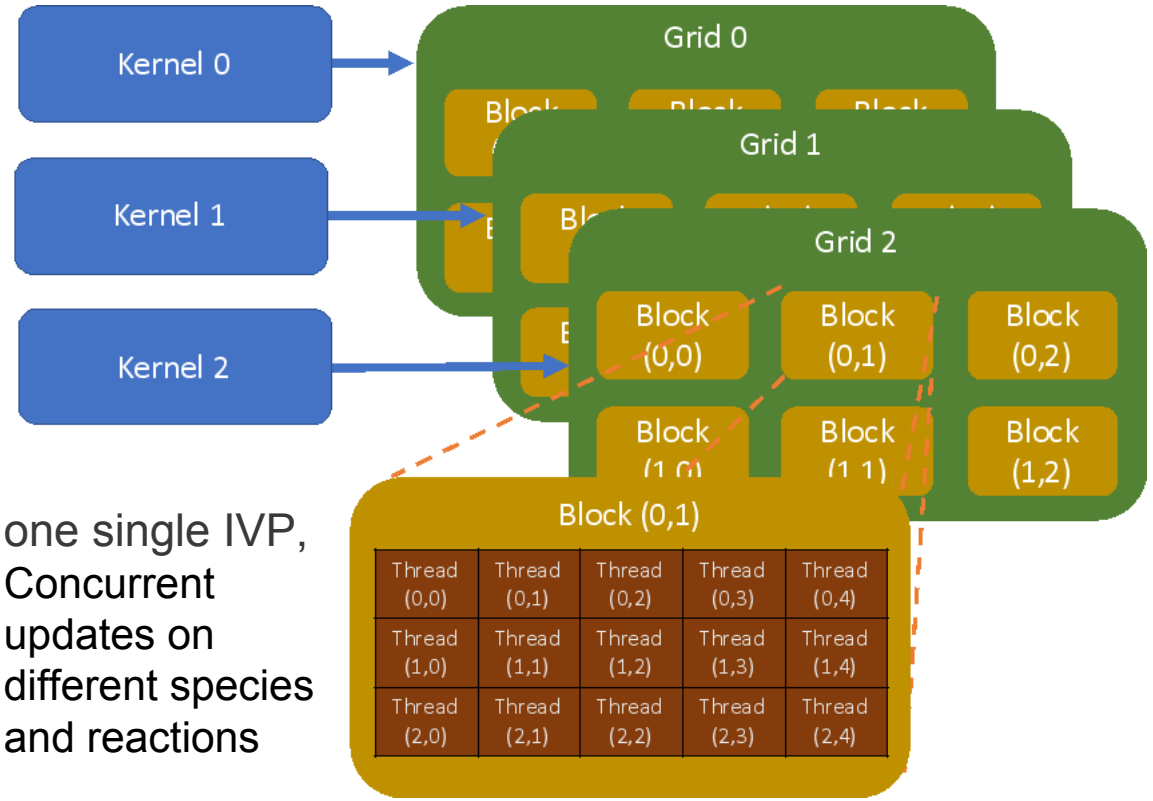
Parallelism in TChem

TChem is designed to exploit hierarchical parallelism to effectively map independent work items to massively concurrent compute units.

multiple kinetic models

multiple IVPs, varying model parameter and operating conditions



one single IVP, Concurrent updates on different species and reactions

Fig. CUDA hierarchically parallel execution scheme

Applications may want to evaluate…

- Sensitivity/UQ studies that need to solve many Initial Value Problems (IVPs) with different model parameters and initial conditions.
- A reactive flow solver needs to evaluate chemical source terms at each spatial grid point.

Numerical Experiments

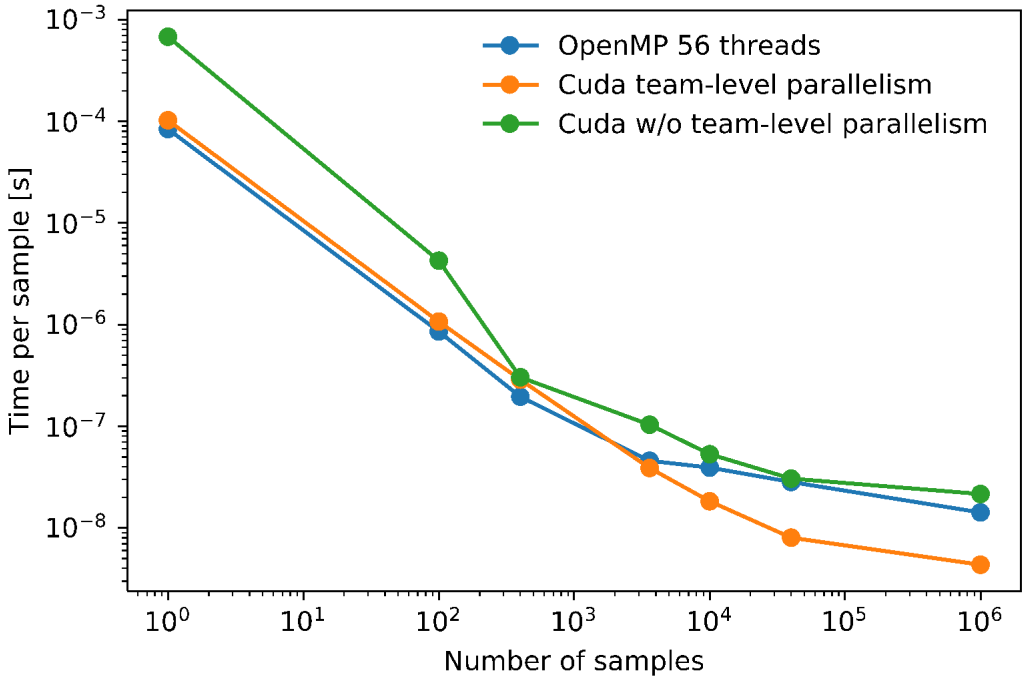
Testbed: Silver

- NVIDIA V100 SXM 32GB
- Intel Xeon Gold 6238R @2.2GHz, 2x28

Kinetic Model: GRIMech 3.0

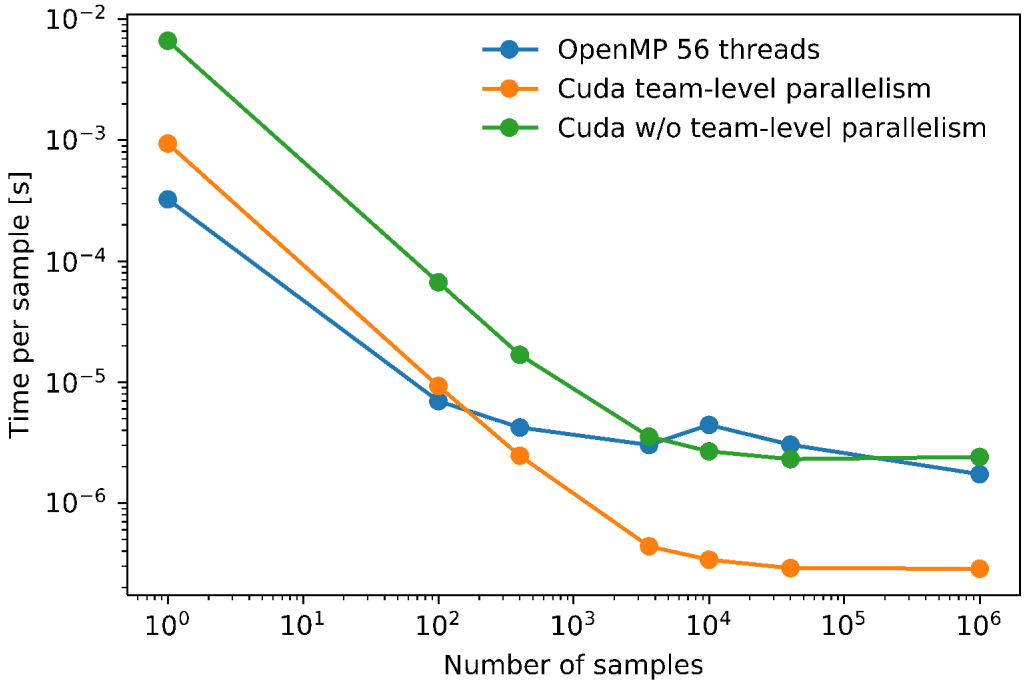
- Methane chemistry including NOx.
- 53 species with 325 reactions.
- A kinetic model can go up 100 species with 1000 reactions.

Parallel Performance with GRIMech 3.0 Model



Mixture Specific Heat Capacity

- **Light-**weight computations.
- 10k is cross-over point that GPU performs better.



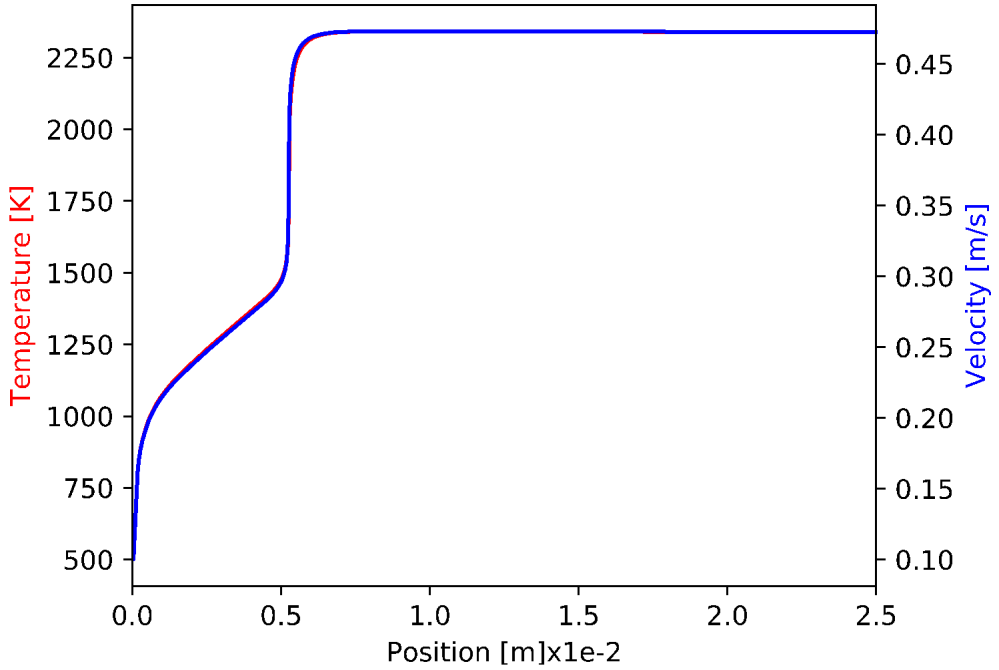
Net Production Rates

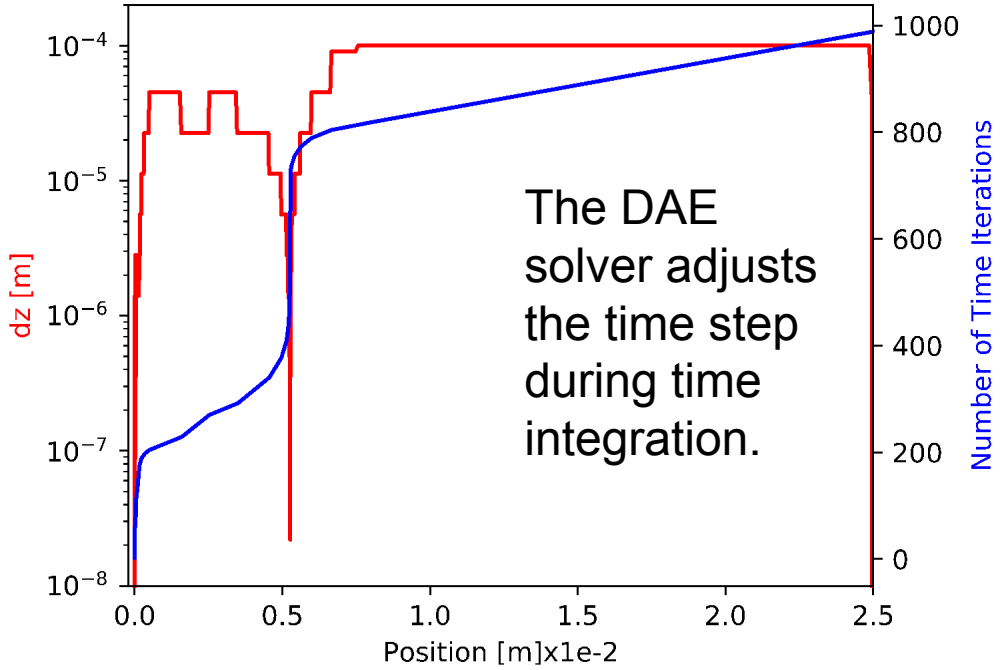
- **Heavy-**weight computation.
- GPU performs faster from 400 samples.

Plug Flow Reactor With Surface Reactions

Combustion of methane on platinum; gas kinetic model GRIMech 3.0, surface kinetic model by the research group of Olaf Deutschmann (Quiceno 2006).

- The DAEs is solved by Tines implicit 2nd order Trapezoidal BDF (TrBDF2)





Numerical setting used by the TChem simulation.

Time Step Settings		Time Step Adjustment		Newton Solver		
Min	Max	Relative tolerance	Absolute tolerance	Max iterations	Relative tolerance	Absolute tolerance
1e-20	1e-4	1e-6	1e-12	20	1e-18	1e-8

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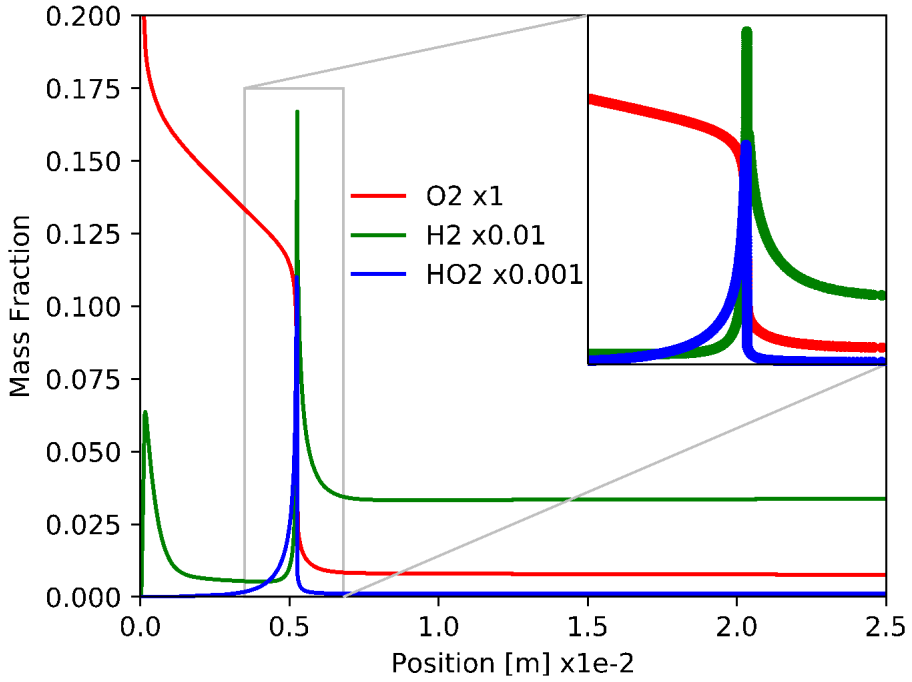
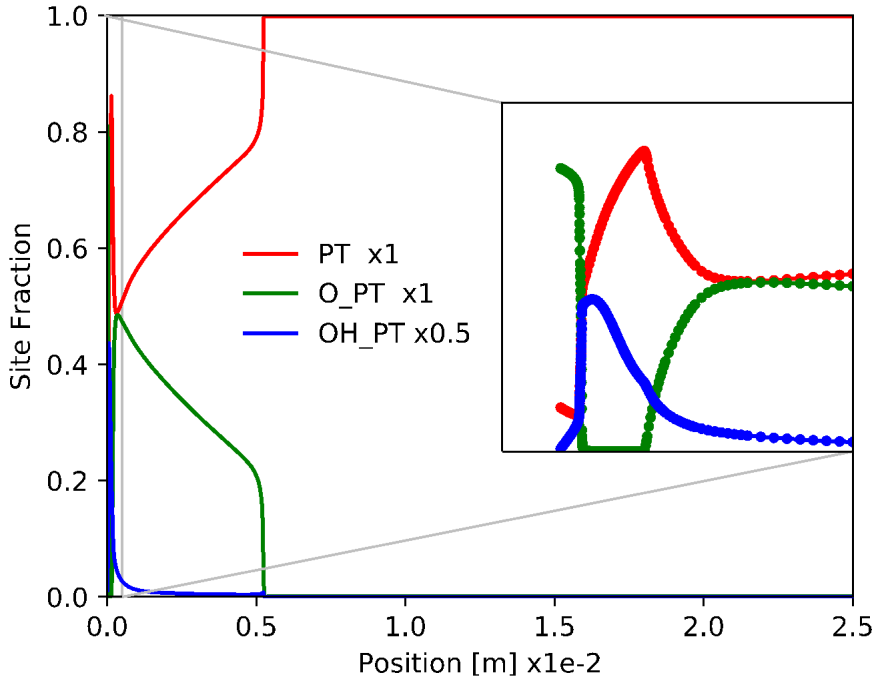
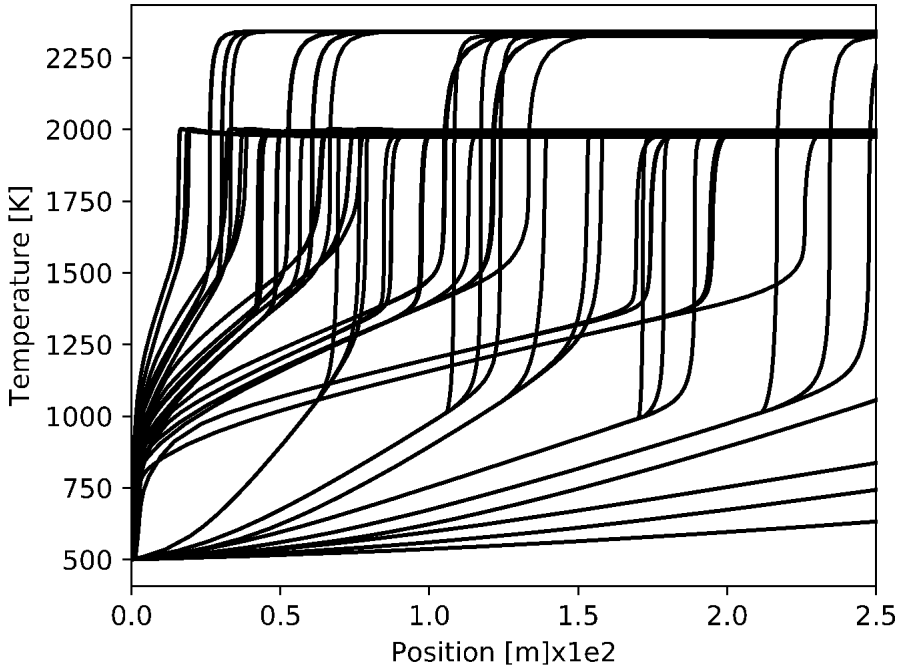


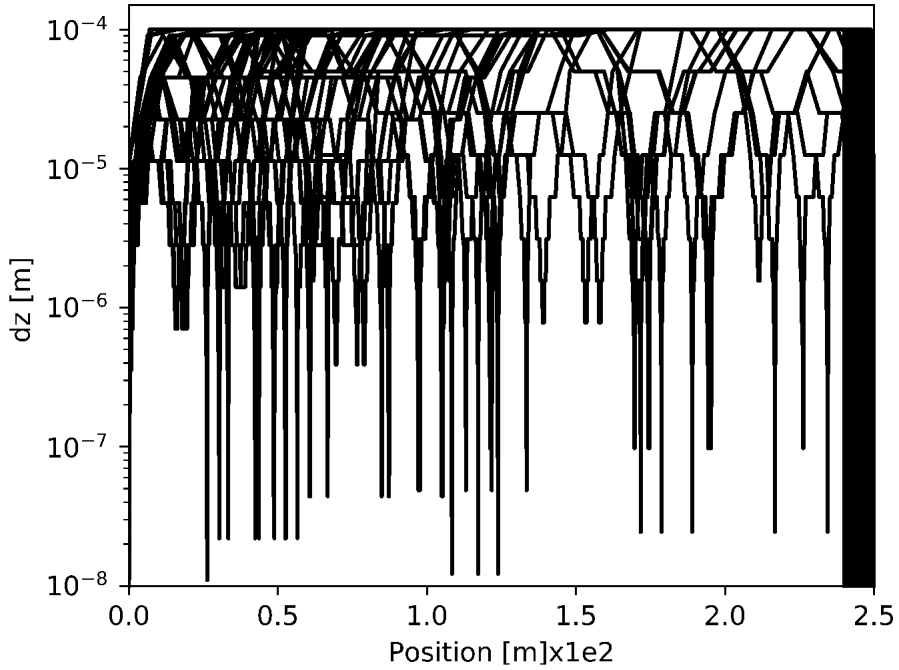


Plug Flow Reactor With Surface Reactions

We used the batched parallel interface available in TChem to run 81 samples for the PFR varying four variables.

Variable	Nominal value	Min value	Max value
Equivalence ratio(ϕ)	1	0.7	1.6
Velocity	0.1	0.05	0.2
$OH + H_2 \leftrightarrow H + H_2O$ ← Modifier gas reaction No 84	1	1e-4	1e4
$H_2 + 2Pt \rightarrow 2HPt$ ← Modifier surface reaction No 1	1	0	10





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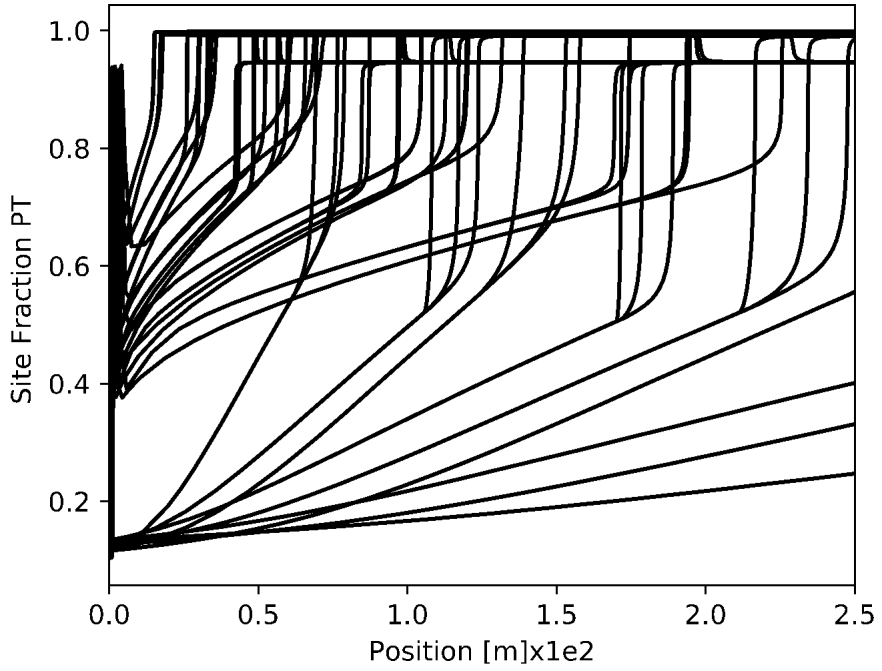
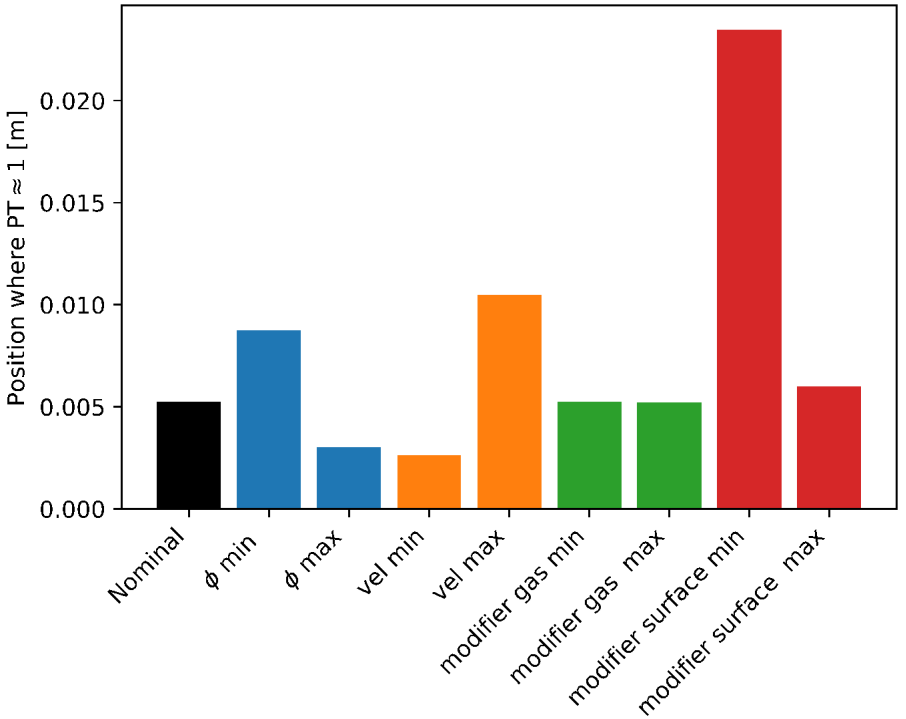
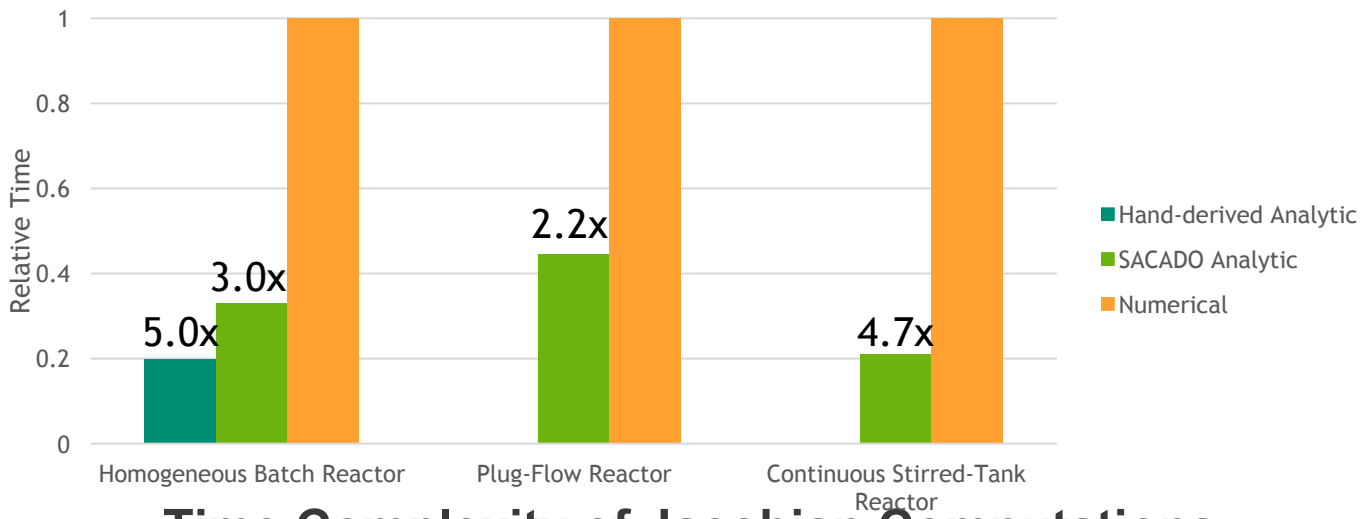




Numerical Jacobians vs Analytic Jacobians

- Numerical Jacobians are computed using adaptive step size to minimize truncation and round-off errors.
- SACADO analytic Jacobians are computed using the same source-term functions with SLFad type.
- Hand-derived analytic Jacobian is available for homogeneous batch reactor.



Time Complexity of Jacobian Computations

- Relative time complexity with respect to numerical Jacobian computation
- Numerical Jacobians are computationally expensive: require $O(N_{spec})$ source term evaluations.
- SACADO-based Jacobians are faster (as expected) – updates based on chain rules for related species only.
- Multiple SACADO SLFad versions are instantiated to efficiently work with different kinetic models.

Summary

- Present TChem, a software toolkit for solving complex chemistry kinetic models on heterogeneous computing platforms.
- Use Kokkos for future-proofing next-generation computing platforms.

On-going & Future Work

- GPU optimization for SACADO and many places
- Use SACADO for gradients w.r.t. kinetic model parameters for UQ

Point of Contact

- **github.com/sandialabs/Tchem**
- Cosmin Safta (csafta@sandia.gov), Kyungjoo Kim (kyukim@sandia.gov), Oscar Diaz-Ibarra (odiazib@sandia.gov)

Acknowledgement

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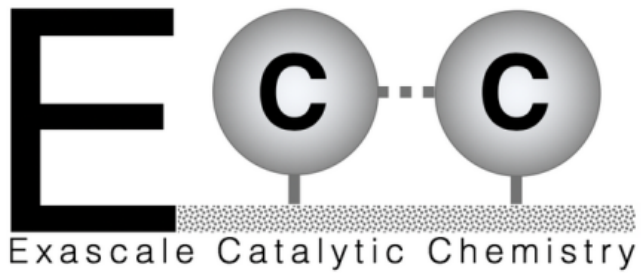
- Early contributions for the C to C++/Kokkos transition from Eric Phipps & Fazle Rob



Questions ?