Recent Development of Thermochimica for Simulations of Nuclear Materials

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NuFuel – MMSNF 2019 Workshop
Paul Scherrer Institute
November 5, 2019

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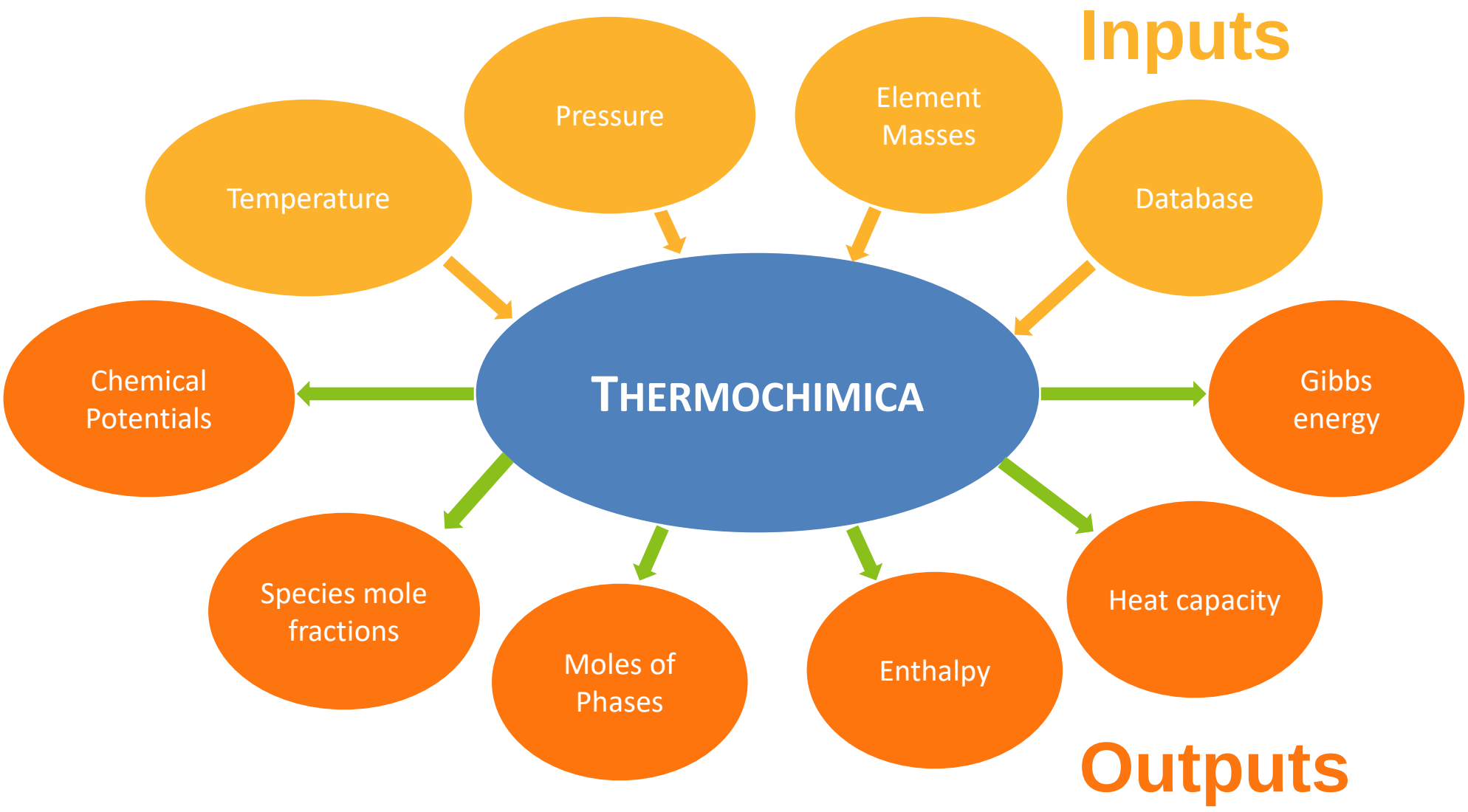
Acknowledgements

- Research was sponsored by the U.S. Department of Energy, Office of Nuclear Energy, Nuclear Energy Advanced Modeling and Simulation Program and Fuel Cycle R&D Program, under contract DE-AC05-00OR22725 with UT-Battelle, LLC.

- This research was undertaken, in part, thanks to funding from the Canada Research Chairs program (950-231328) of the Natural Sciences and Engineering Research Council of Canada.

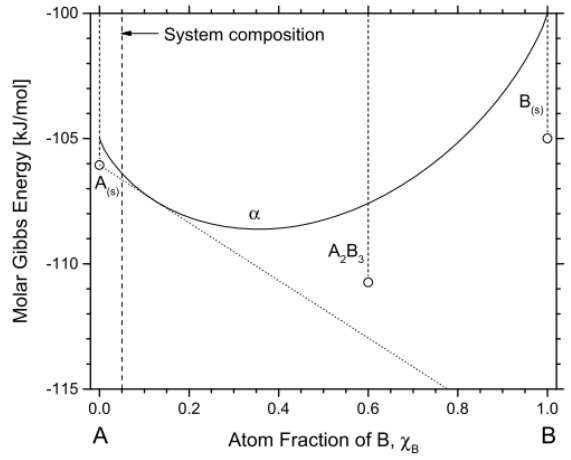
Brief Overview of Thermochimica

- Gibbs energy minimizer designed for direct coupling to multi-physics codes
  - Fortran 90
  - API available for C++
- Open-source (https://github.com/ORNL-CEES/thermochimica)
- Works directly with FactSage-derived DAT files
- Capabilities:
    - Stoichiometric phases
    - Ideal gas (IDMX)
    - Regular substitutional models (QKTO, RKMP)
    - Compound Energy Formalism (SUBL)
    - Modified quasi-chemical model (SUBG, SUBQ)

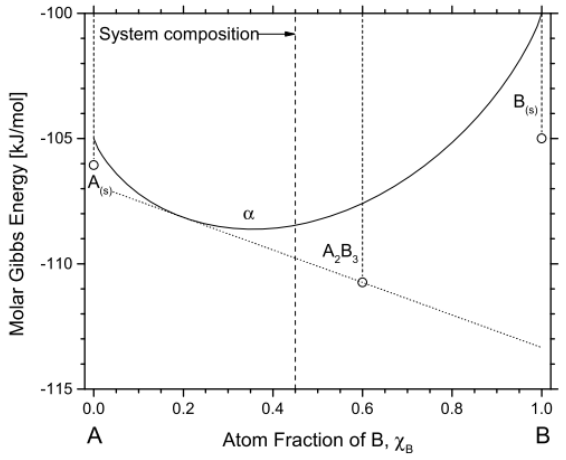


https://github.com/ORNL-CEES/thermochimica

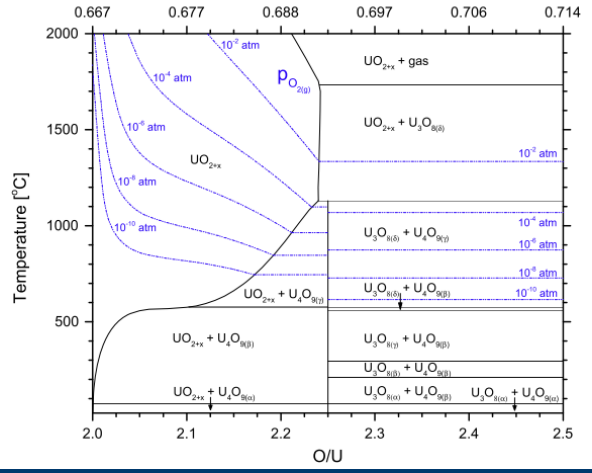
Computational Details

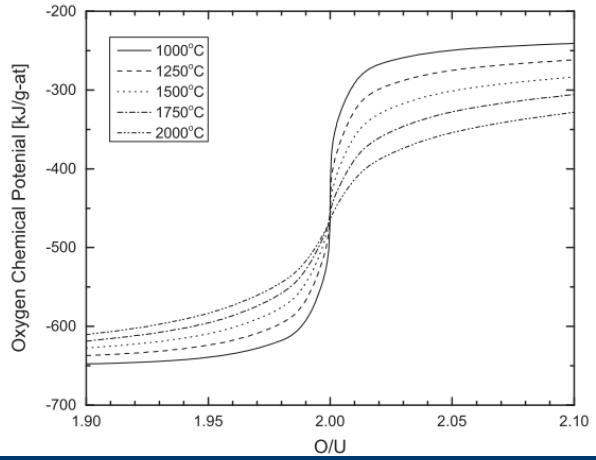


(a) $A_{(s)}$ is in equilibrium with $\alpha_{(s)}$.



(b) $A_2B_{3(s)}$ is in equilibrium with α.





- Equilibrium thermodynamics
- Isothermal / isobaric conditions
- Constrained optimization of Gibbs energy
- Point calculations (no geometry)
- Determines stable phases and their compositions
- Applications:
  - Equilibrium point calculations
  - Phase diagrams
  - Multiphysics coupling

M. H. A. Piro, M. J. Welland, M. Stan , J. Nucl. Mater., 464 (2011) 48–52.

Recent Work

Oxygen Transport in UO_2 (MOOSE/Bison)

- Chemical diffusion model for isothermal conditions can be written as:

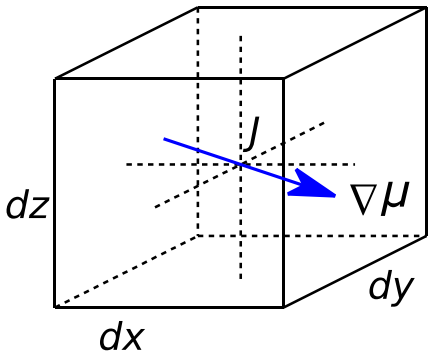
$$v_k = M_k F_k \qquad J_k = n_k v_k = n_k M_k F_k = -M_k n_k \nabla \mu_{gk}$$

- Mobility of O for $UO_{2\pm x}$ (Moore et al. 2013)

$$M_O = M_{Va} + M_{I_O}$$

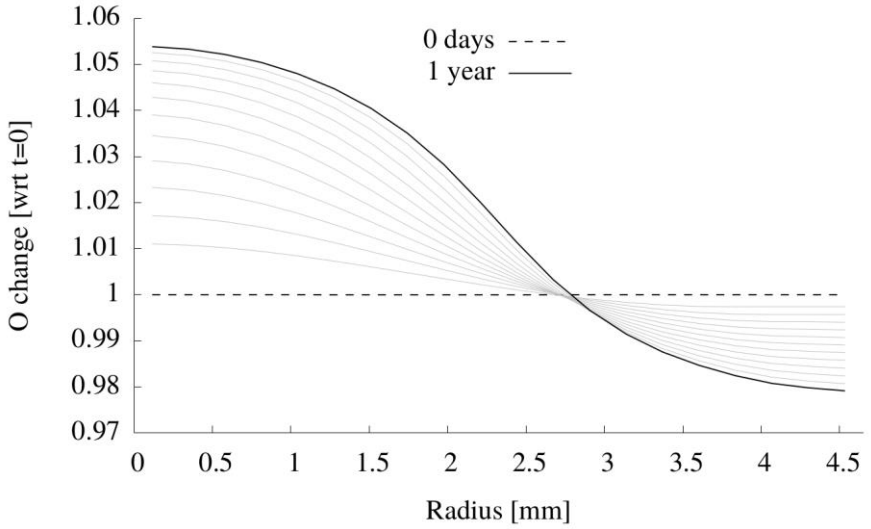
$$M_{Va} = y_{Va}(1 - y_{Va}) M_{Va}^0 \exp\left(\frac{-Q_{Va} + y_{Va}(1 - y_{Va})(A - BT)}{RT}\right)$$

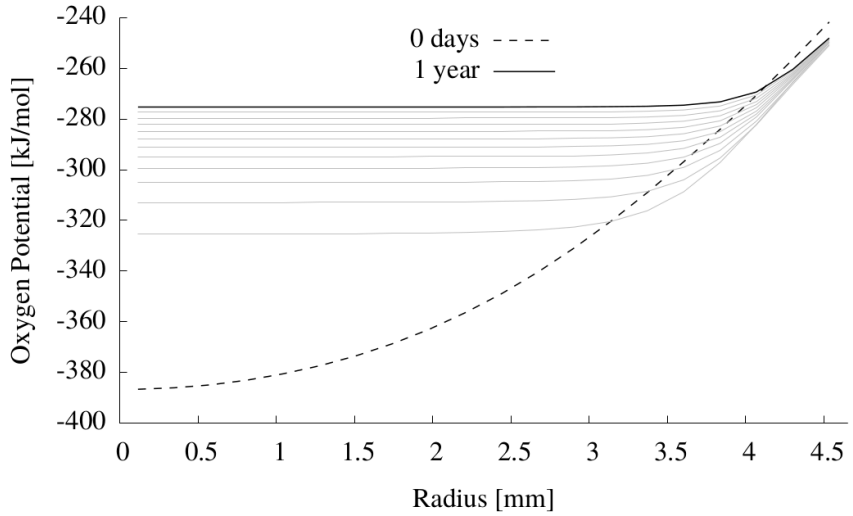
$$M_{I_O} = y_{I_O}\left(1 - y_{y_{I_O}}\right) M_{y_{I_O}}^0 \exp\left(\frac{-Q_{y_{I_O}} + y_{y_{I_O}}\left(1 - y_{y_{I_O}}\right) A}{RT}\right)$$



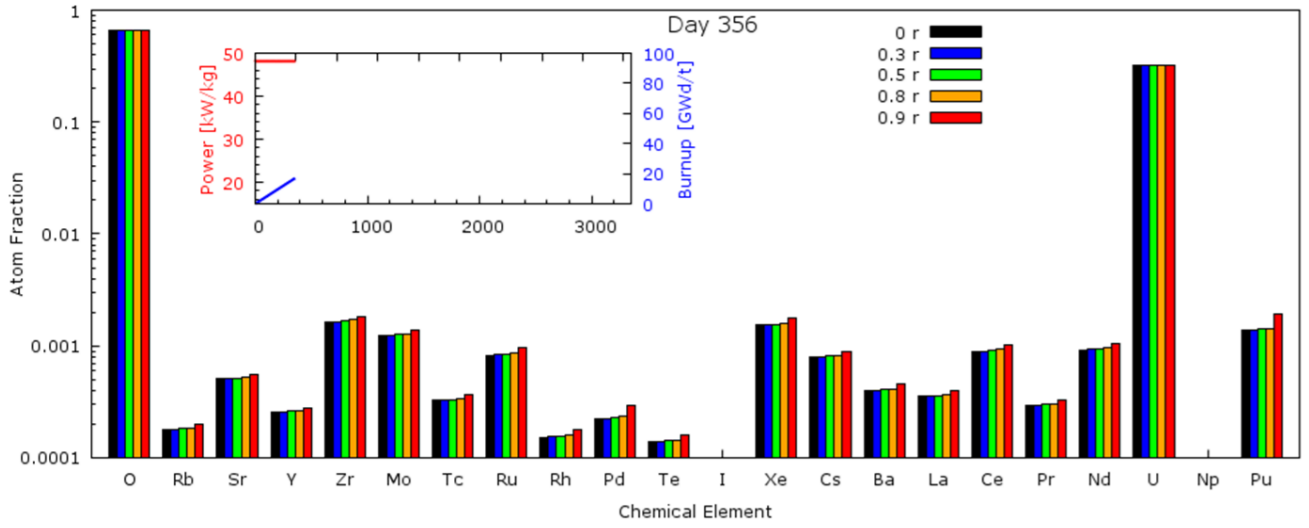
- Material states (y_{Va}, y_{I_O}) are from thermodynamic calculations
- Implemented in Bison

Oxygen Transport in UO_2 (MOOSE/Bison)





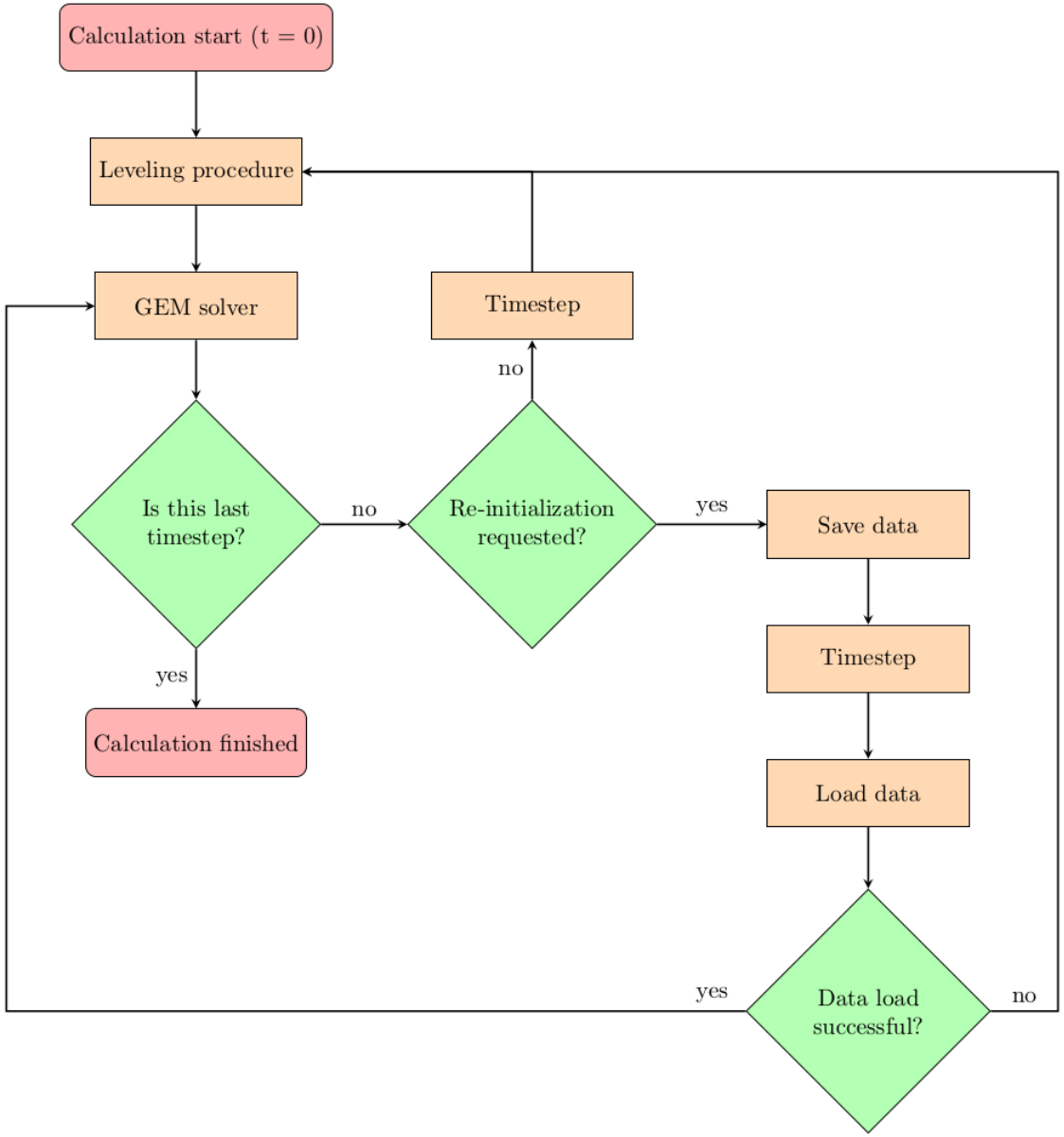
- Material composition is calculated by ORIGEN-S and read into the model as a function of time
- Chemical potential from Thermochimica drives diffusion
- Thermodynamics of Advanced Fuels International Database (TAF-ID) used
- Simulated for 1 year of burnup



S. Simunovic, T.M. Besmann, E. Moore, M. Poschmann, M.H.A. Piro, K.T. Clarno, J.W. McMurray, W.A. Wieselquist, 2019, Under Review.

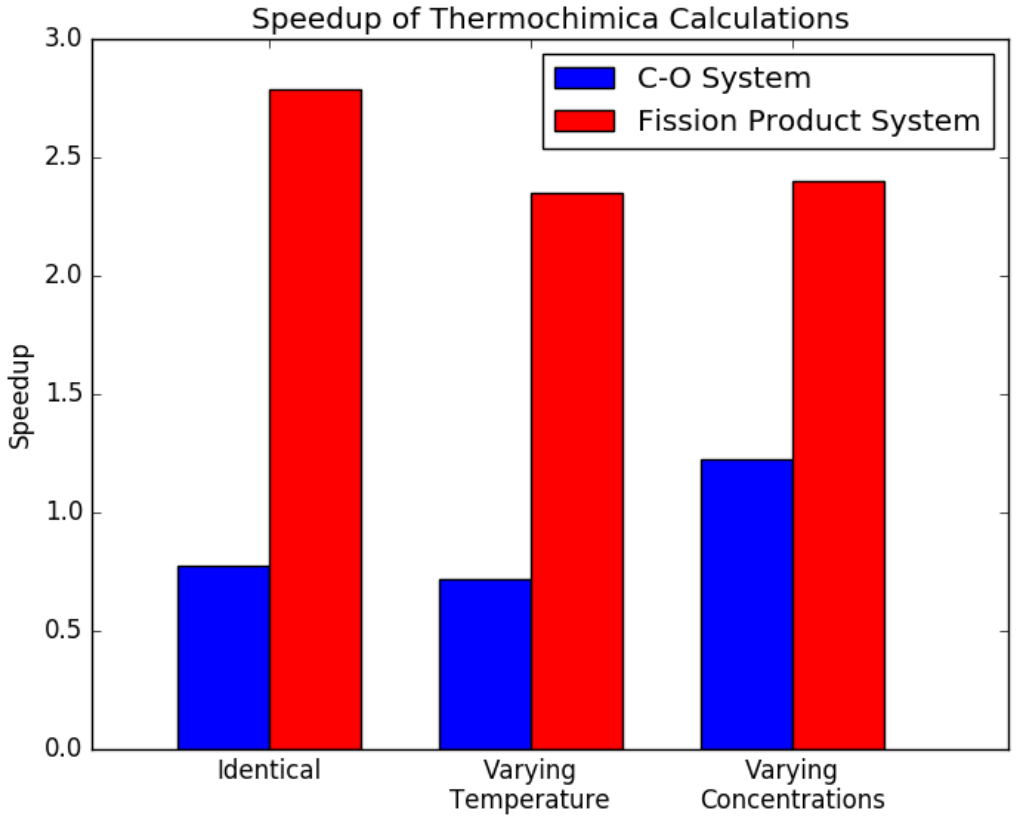
Acceleration of Thermochimica

- Often, consecutive Thermochimica calculations have similar conditions
  (T, P, chemical concentrations)
- Re-initializing using solution from previous time step can provide more accurate initial estimate than leveling solver
- Saves many energy minimization steps



Stand-Alone Speedup

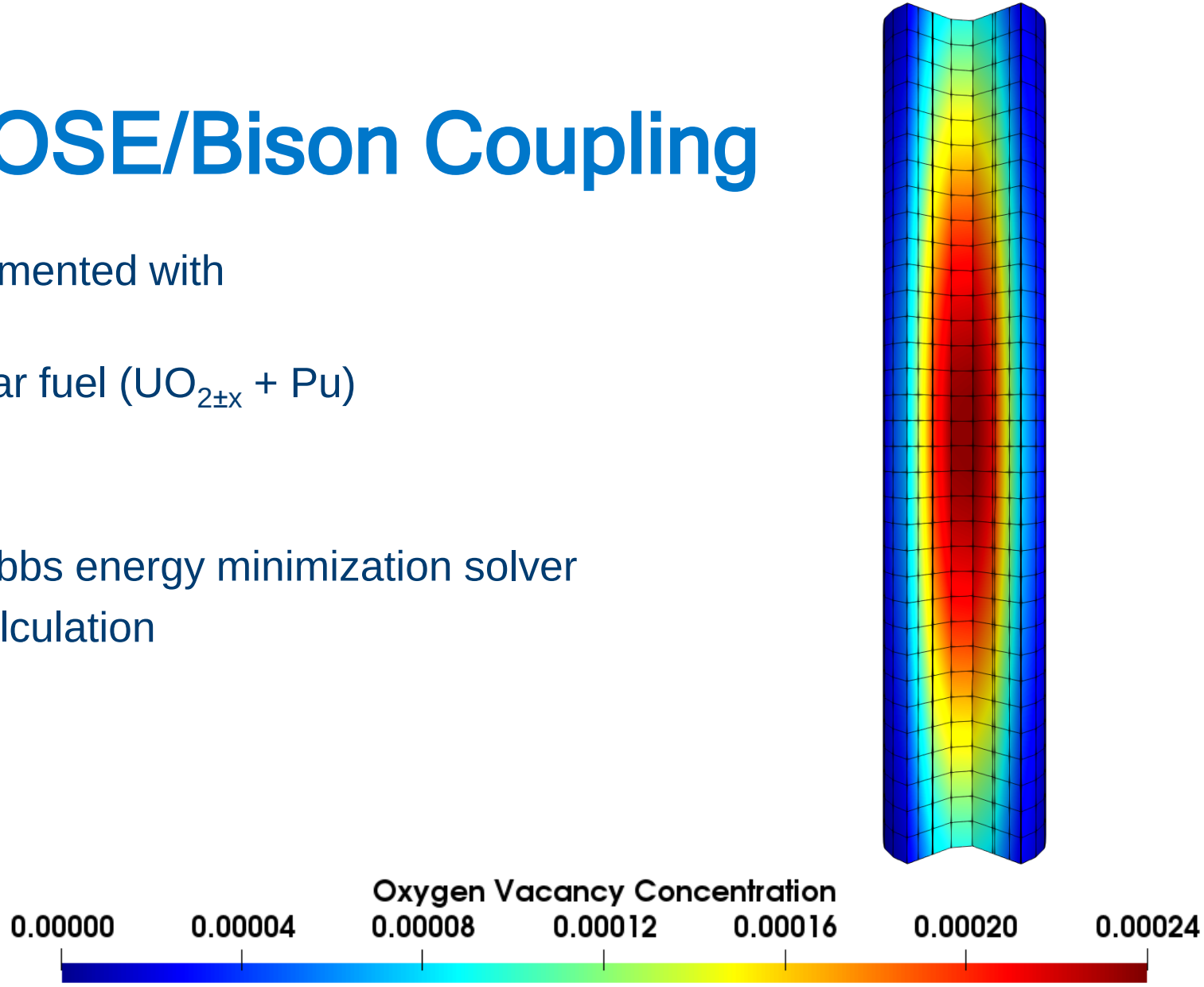
Carbon – Oxygen System	Fission Product System
2 elements	23 elements
1 solution phase	15 solution phases
4 stoichiometric phases	240 stoichiometric phases
0.7x – 1.2x speedup	2.3x – 2.8x speedup



- Speedup better for larger, more complex systems
- Tested for constant and varying conditions
- Speedup of 2x – 3x for most cases
- Negative speedup possible for trivial calculations!

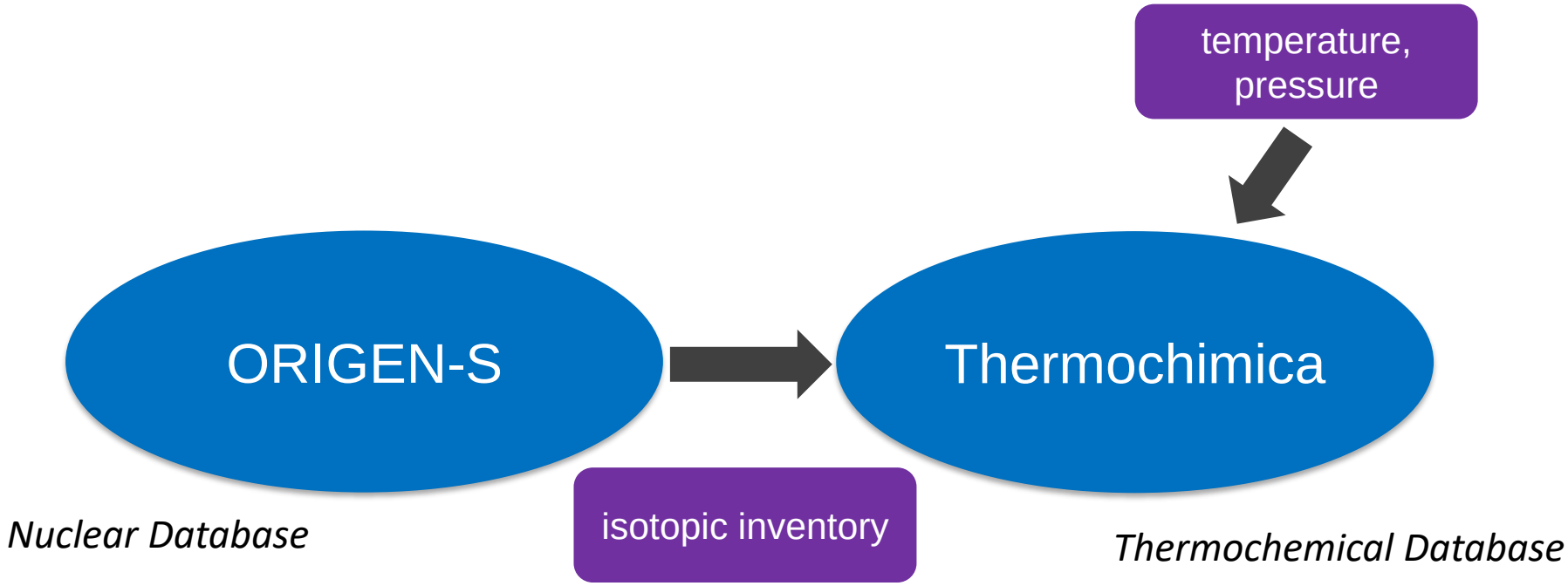
Speedup with MOOSE/Bison Coupling

- 1D and 3D Bison examples implemented with re-initialization
- Diffusion of oxygen in LWR nuclear fuel ($UO_{2\pm x}$ + Pu)
- 3 elements, 8 solution phases
- 18 stoichiometric phases
- Up to 96% reduction in calls to Gibbs energy minimization solver
- Speedup of 2.7x – 7.4x of total calculation



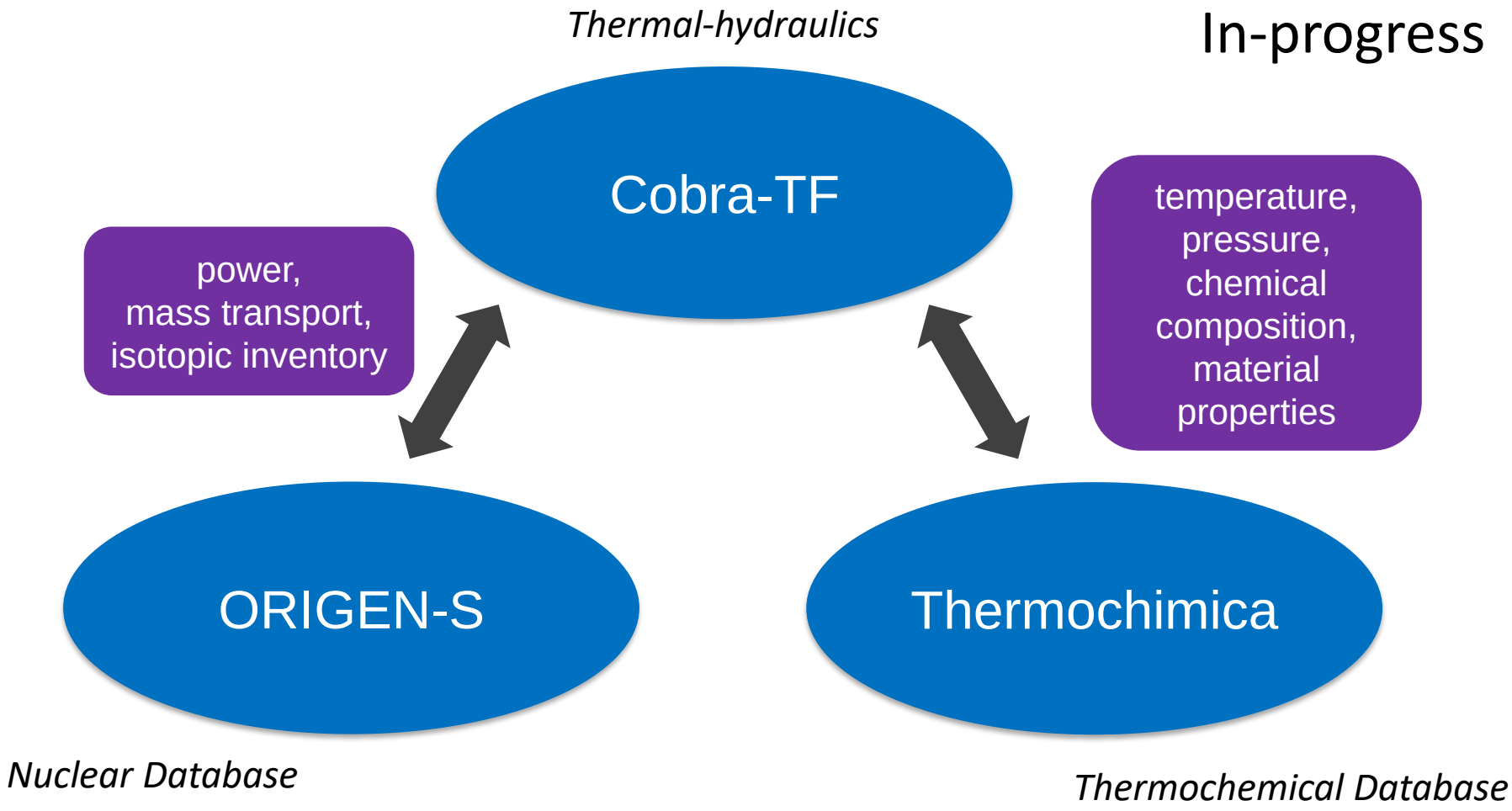
MSR Multi-physics (ORIGEN/CTF)

Current capabilities



In collaboration with B. Collins *et al.* (ORNL).

MSR Multi-physics (ORIGEN/CTF)



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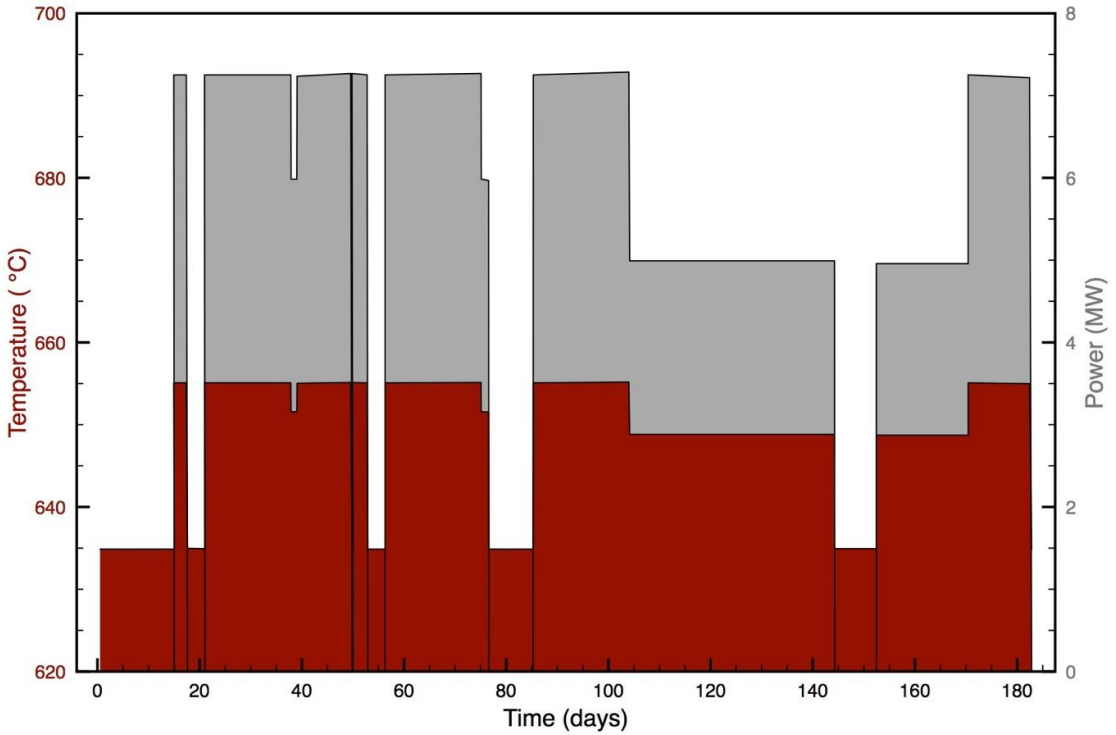
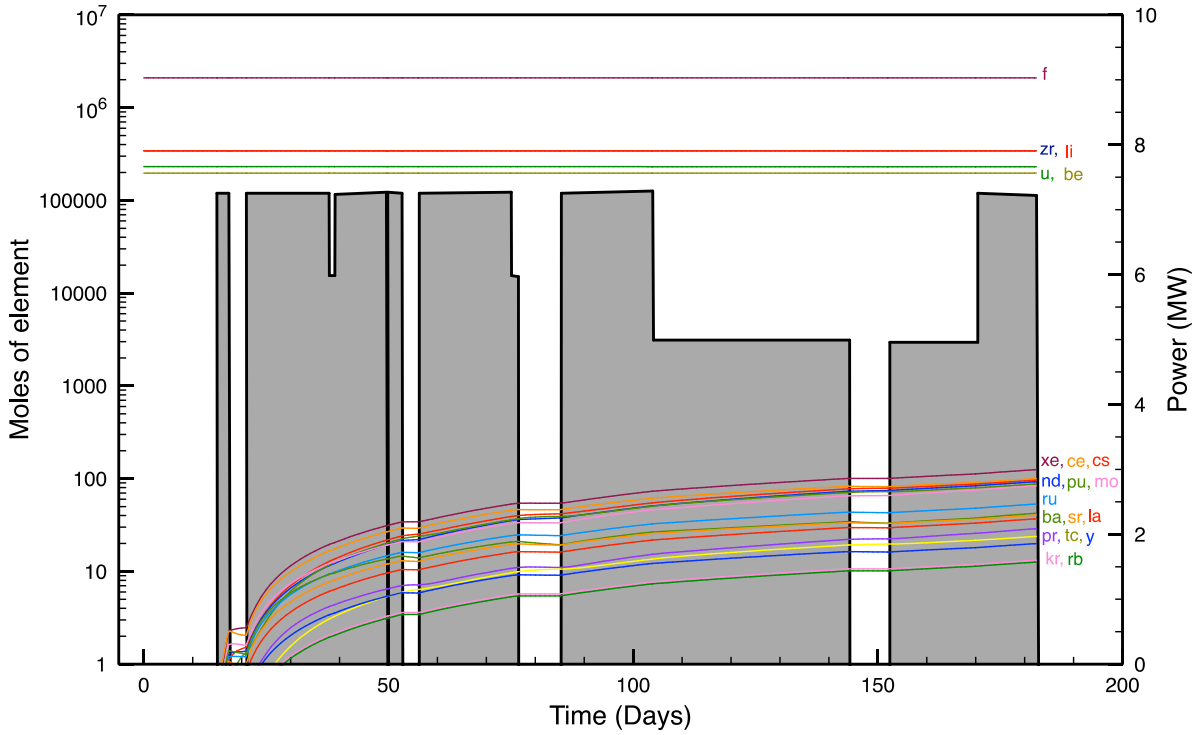
For demonstration purposes, temperature was approximated by taking a linear function of power.

MSR Multi-physics – Power & FP Evolution

Power Profile / Temperature Approximation



Fission Product Evolution in MSRE



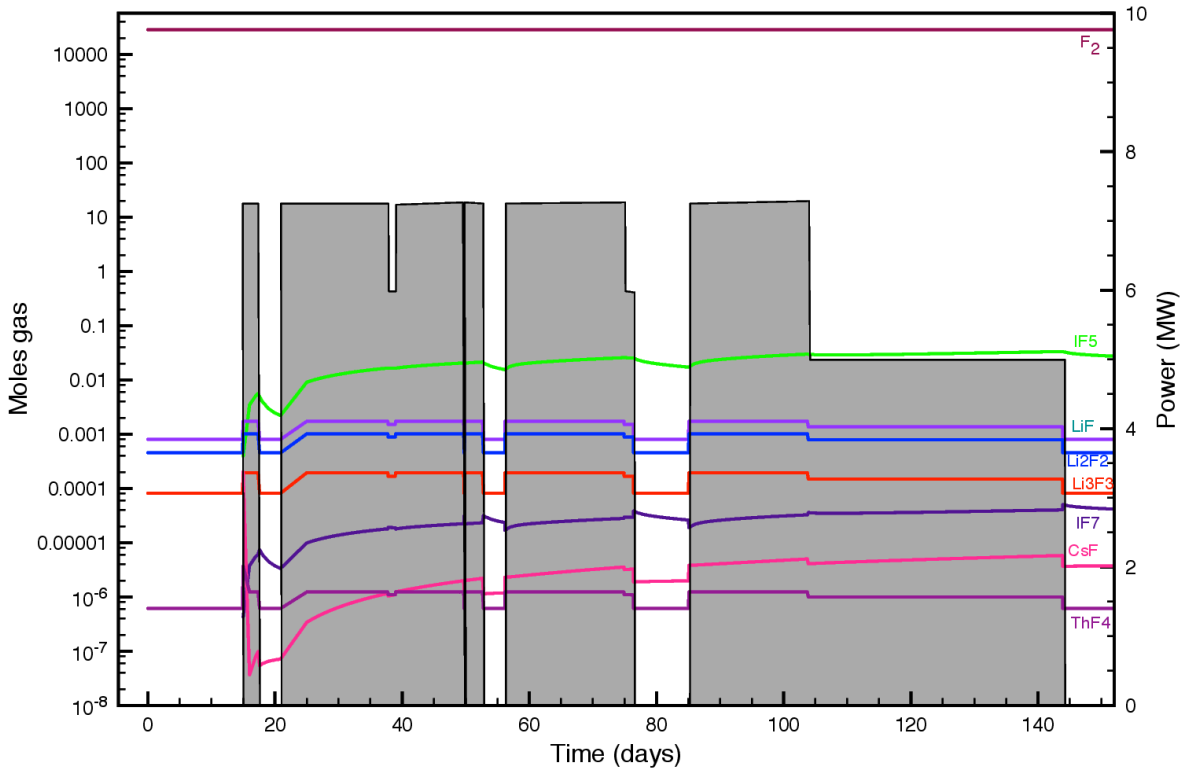
B. Fitzpatrick, *et al.*, to be published.

Disclaimer: these are preliminary results and are intended to demonstrate capabilities.

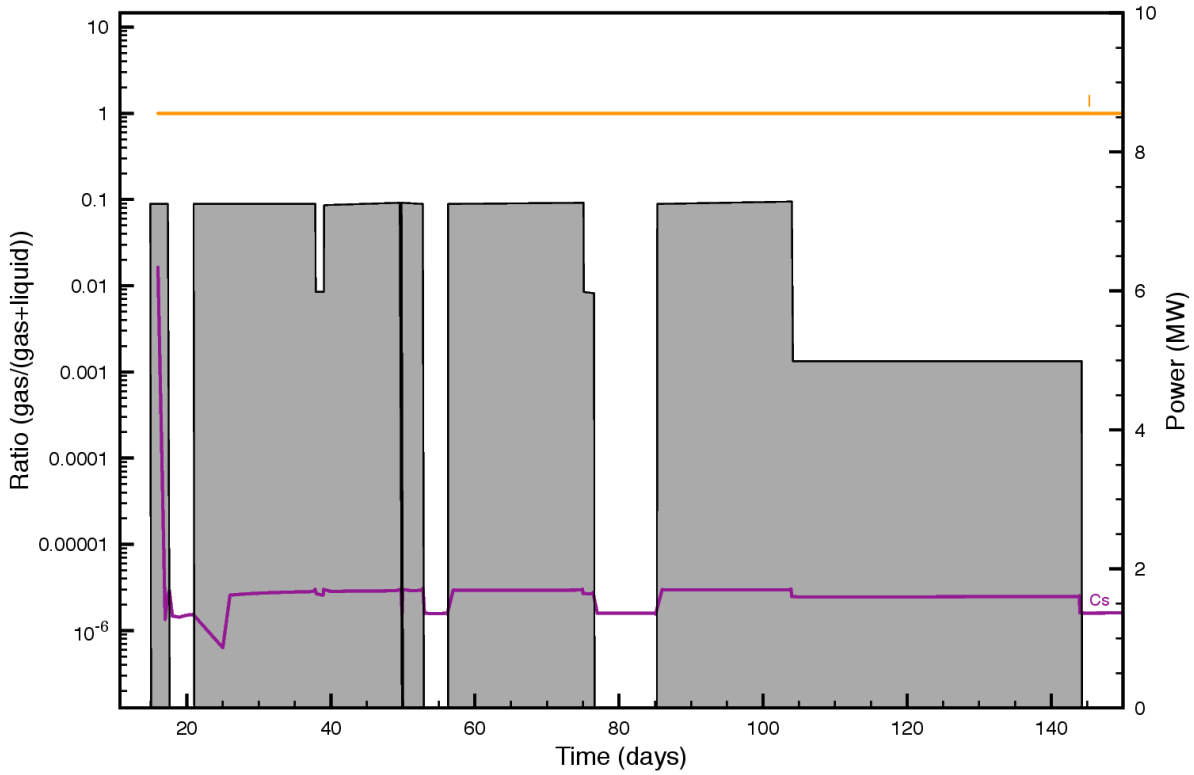
Uranium content from ORIGEN-S was mapped to thorium content in Thermochimica for demonstration purposes.

MSR Multi-physics - Thermochemistry

Gaseous Species



Mole Fraction in Gas Phase



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Disclaimer: these are preliminary results and are intended to demonstrate capabilities.

Data-file of (Li,Cs,Th//F,I) kindly provided by E. Capelli.
E. Capelli, O. Benes, R. Konings, J. Nucl. Mater., 501 (2018) 38-252.

Future Work

- Continued development and testing of capabilities
- Applications to new engineering systems
  - Fuel performance
  - Safety analyses
  - Severe accident scenarios
  - Pellet-clad interaction
- Ongoing support for sponsored projects
  - MOOSE/Bison coupling for UO_2 and metallic fuels
  - ORIGEN/CTF coupling for MSRs
- Your ideas
  - Contact me at max.poschmann@ontariotechu.ca

Conclusions

- Thermochimica is an efficient and flexible code for finding equilibrium conditions and useful thermodynamic quantities
- Thermochimica can be coupled to a variety of useful multiphysics software packages
- Thermochimica has the capabilities needed to simulate many systems, including UO_2 and molten salt nuclear fuels

Thank you!

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