Thermochimica-Transport Coupling for MSR Offgas Composition Estimation

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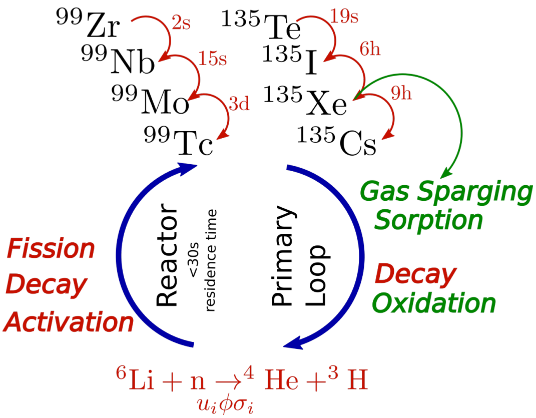
Target Application

Bateman equations coupled to reaction, advection, diffusion (RAD) produces stiff, linear dominate system.

$$\frac{\partial \mathbf{u}}{\partial t} = \underbrace{\mathbf{L}\mathbf{u}}_{Decay,Trans.} - \underbrace{v\nabla \mathbf{u} + D\nabla^2 \mathbf{u}}_{Adv.,Diffusion} \pm \underbrace{S(\mathbf{u},t)}_{Phase.,Offgas.}$$

where $\mathbf{u}$ is a vector of the primary species. $\mathbf{L}$ contains entries spanning many orders of magnitude, from decay λ, transmutation $\phi\sigma_i$, etc.

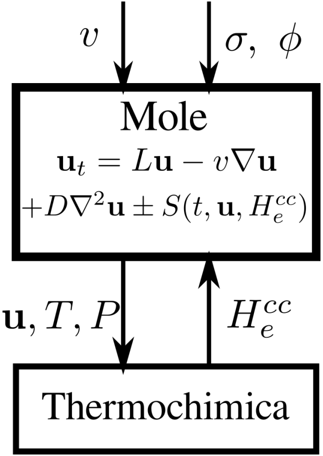
MSR System



ϕ : neutron flux, and σ: cross section.

Target Application

- Mole serves as a testbed for 0D/1D many-species transport methods.
- Includes Aux. kernels for Henry's gas constant and liquid→vapor phase transport.
- Other MOOSE codes (SAM) can utilize the Mole Aux. mass transfer and Henry's gas kernels [1].
- Interface with Thermochimica (TC) to estimate liq. vapor equilibrium constants.
- Prototype new space and time discretization methods methods.



[1] Gurecky, et.al.FY24 Mole Development Updates and SAM-Mole Coupling. `https://doi.org/10.2172/2479034`

High and Low volatile Liq. to Vap. Source Terms

$$S(\mathbf{u},t) = S(u,t)_{HV} + S(u,t)_{TC} - S(u,t)_{offgas} + \dots$$

Henry's gas constant-based models[2] are presently used for high-volatile (HV) individual non-reactive primary isotopes, eg. ^{135}Xe, ^{90}Kr:

$$S(u,t)_{i,hv} = k_i a_b(\mathbf{u})(H_i R T c_{v,i} - c_{a,i})$$

$a_b(\mathbf{u})$ is the interfacial surface area per volume, k_i is the mass transfer coefficient, and H_i is the gas constant for species i. c is *per phase* species concentrations,

$$c_{v,i} = u_{v,i}/\alpha \ , \quad c_{a,i} = u_i/(1-\alpha)$$

where u_i is the total vol. concentration and α is the void fraction.

[2] K. Lee. et.al. Semi-empirical model for Henry's law constant of noble gases in molten salts. Sci. Rep. 2024.

High and Low volatile Liq. to Vap. Source Terms

$$S(\mathbf{u}, t) = S(u, t)_{HV} + S(u, t)_{TC} - S(u, t)_{offgas} + \ldots$$

Using Thermochimica (TC), obtain an additional source term to account for the liquid and vapor phase concentrations of secondary volatile chemical species (CsI, I_2, ...). Take a simplified model to account for low volatile species:

$$S(u, t)_{i,TC} = x_i k_e a_b \left(H_e^{cc} \sum_{i \in e} c_{v,i} - \sum_{i \in e} c_{a,i}\right), \quad H_e^{cc} := \frac{c_{a,e}^{TC}}{c_{v,e}^{TC}} = \frac{u_{a,e}^{TC} \alpha}{u_{v,e}^{TC}(1-\alpha)}$$

with the per-element dimensionless gas constant H_e^{cc} determined by TC. This value determines the liq. vap. equilibrium value for element, e. k_e is an effective per-element mass transfer coefficient, x_i is the isotopic fraction.

High and Low volatile Liq. to Vap. Source Terms

Let j be a secondary species index: $\{c_{j=0}, c_{j=1}, ...\} = \{c_{\text{CsI}}, c_{\text{I}_2}, ...\}$

$$k_e = \sum_j w_j k_j$$

The weights, w_j are computed from stoichiometric fractions. Potential simplification: Set $K_{eff} = k_e a_b$ to a large, fixed constant. What is the end result and advantage?

- Drive the per-element liquid/vapor ratios to the TC equilibrium value.
- Stiff linear term(s) can be handled via CRAM-like methods.

$$S(u,t)_{i,TC} = x_i K_{eff} (H_e^{cc} \sum_{i \in e} c_{v,i} - \sum_{i \in e} c_{a,i})$$

0D Transient, Simplified 4 Species TC Coupling Test Case

Salt at t=0: LiF-BeF2-ZrF4-UF4 (65-29.1-5-0.9) + $^{135}I_l$ + $^{135}Cs_l$

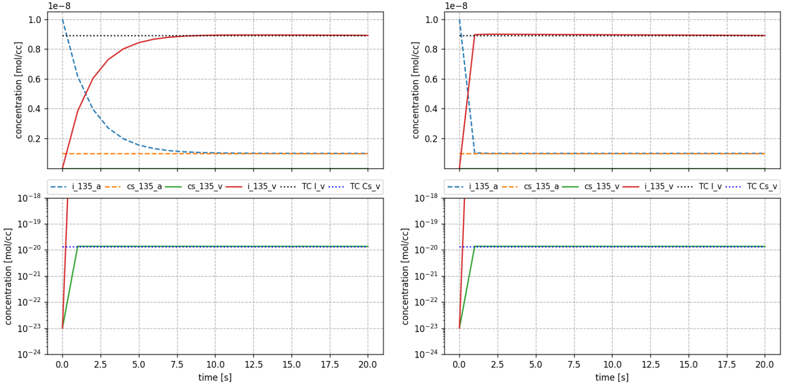


Figure 1: Left: $k_m a_b = 0.5$. Right: $k_m a_b = 5.0$. T=950K, P=1atm.

4.53×10^{-9} mol/cc IG.

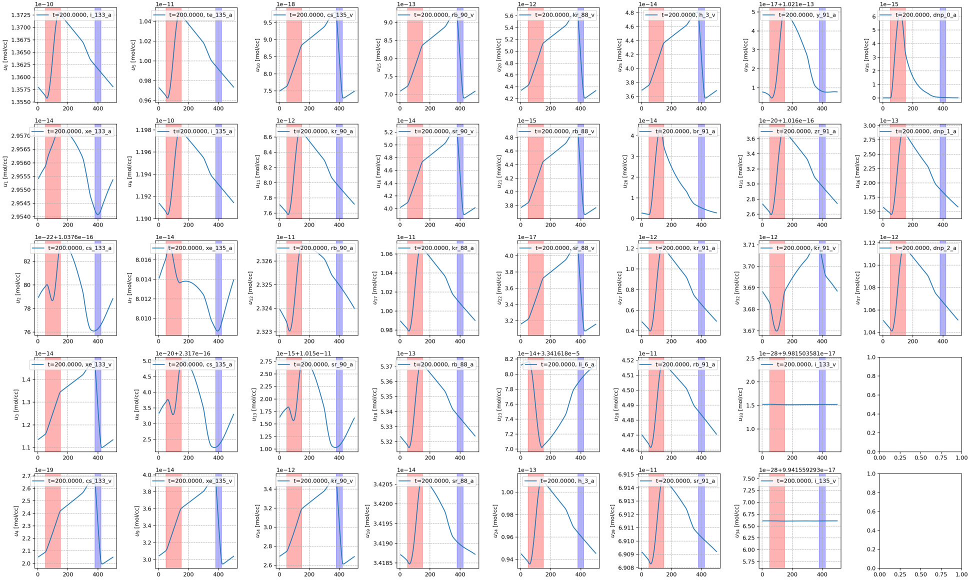
Species	mol frc.
I_2	0.966
I	3.32×10^{-2}
UF_5	1.91×10^{-4}
ZrF_4	1.06×10^{-5}
BeF_2	9.82×10^{-6}
...	...
$CsZrF_5$	4.11×10^{-13}
CsI	4.14×10^{-14}

TC ideal gas phase composition at 20s.

1D Nonlinear RAD, MSR, 38-species

$$\frac{\partial \mathbf{u}}{\partial t} = \underbrace{L\mathbf{u}}_{Decay,Trans.} - \underbrace{(1/A)(\nabla(Av\mathbf{u}) + D\nabla \cdot (A\nabla \mathbf{u}))}_{Adv.,Diffusion} \pm \underbrace{S(\mathbf{u}, t)}_{liq \to vap.}$$

- Track 38 primary species.
- Preliminary, One Way TC coupling (no feedback).
- Physics Included: Adv., Diff., Bateman, high-vol. liquid-gas transfer, gas removal, fission, n-capture. Placeholder reactor parameters and geometry.
- 3^{nd} order FEM - Gauss-Lobatto. Periodic BCs. Nonconstant pipe diameter.
- v_{RX} =0.2 m/s, $\Delta t = 2.0$(s). $t_f = 86400$ (s). Loop length 5(m).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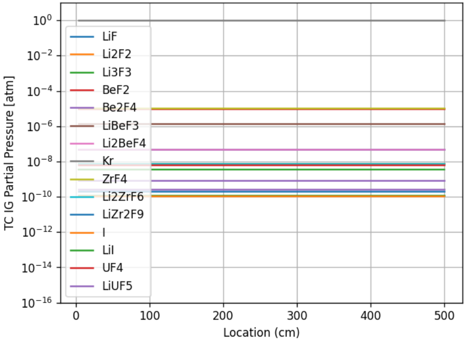
Nonlinear RAD, MSR, 38-species. Void Frac.



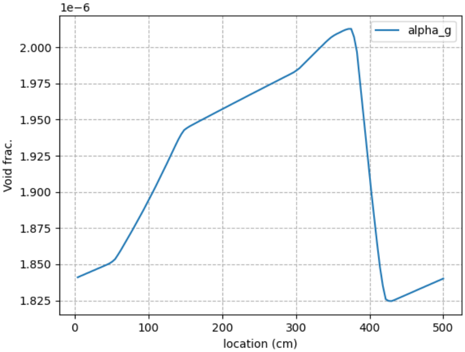


Figure 2: Left: Gas TC species partial pressures, $t = 200(s)$. Right: Void Frac. profile.

Questions

Appendix

Nonlinear RAD, MSR, 38-species. Jacobian Eigs.

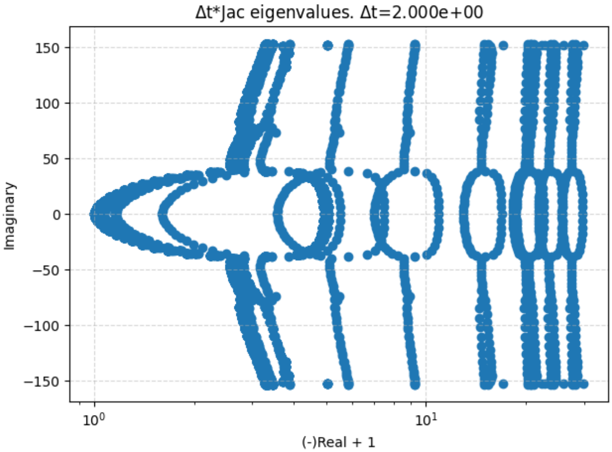


Figure 3: Scaled Eigs of Jacobian at t =86400(s). CFL: 156.

High and Low volatile Liq. to Vap. Source Terms

$$S(\mathbf{u}, t) = S(u, t)_{HV} + S(u, t)_{TC} - S(u, t)_{offgas} + \dots$$

The offgas sink term is estimated by:

$$S(u, t)_{offgas,i} = \frac{k_i v_b A_b u_{v,i}}{V_{cell}}$$

where v_b is the bubble rise velocity, carrying vapor primary species with concentrations $c_{v,i}$, and A_b is the free-surface area, k_i is a dimensionless constant.