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A simple hydride model for cerium ejecta particles

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Funding: ASC/PEM/Mix and Burn
ASC/IC/LAP
LDRD

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Ejecta Working Group Seminar

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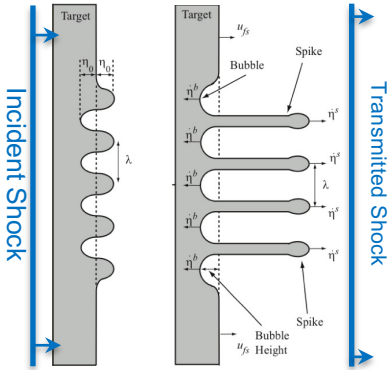
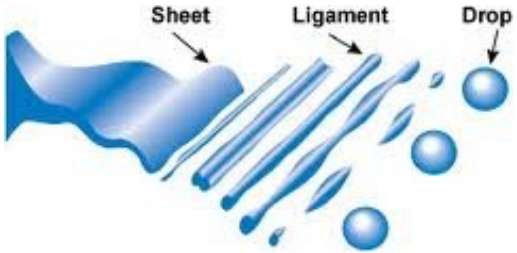
Motivation – Ejecta processes

source

breakup





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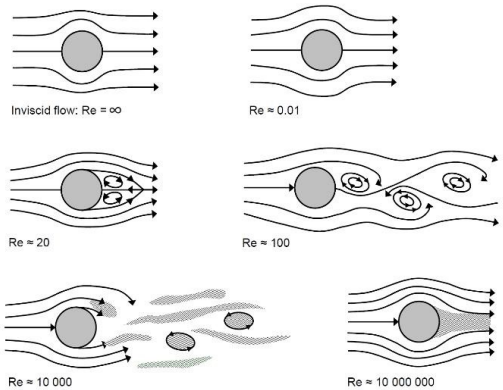
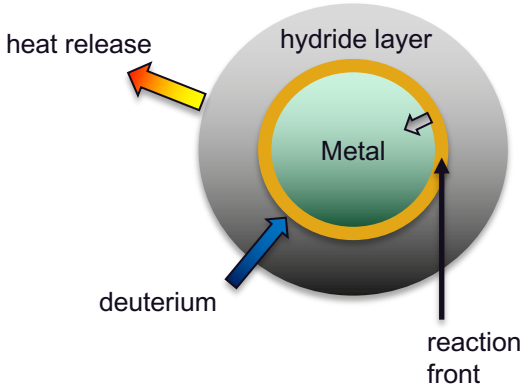


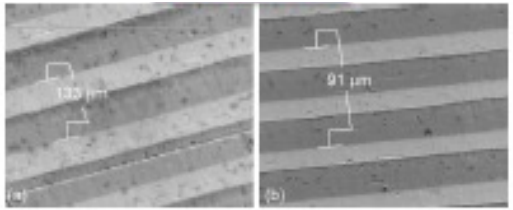
conversion

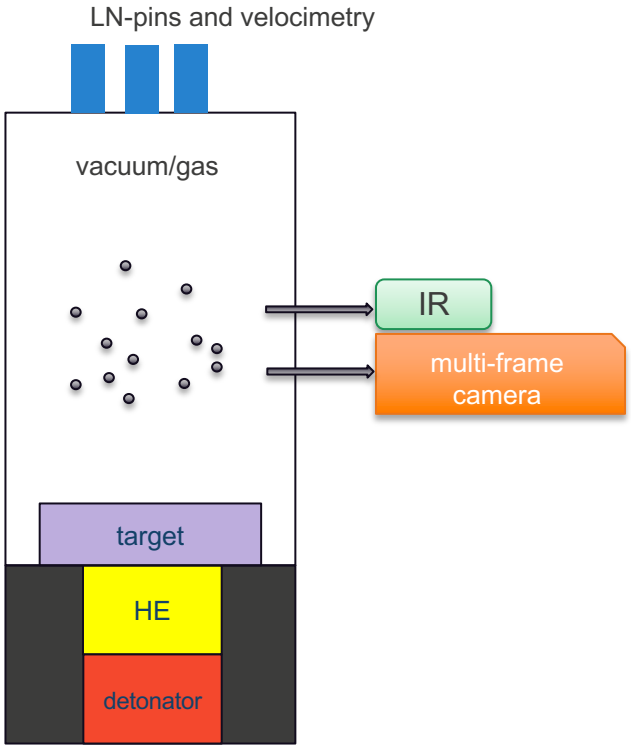


LDRD Experiment (PI: W. Buttler, LA-UR-19-21158)

- HE drives shock wave into target → cerium and tin ejecta into
  - Vacuum, He, D_2 at 2, 4, 8 atm
- IR imaging to measure radiance temperature
- PDV for ejecta velocities
- LN-pins for mass
- Initial ejecta temperature is 990K
- Mie scattering measures particle size
  - 12 micron mean diameter for cerium
  - 2 micron mean diameter for tin
- Inferred solid cerium ejecta particles

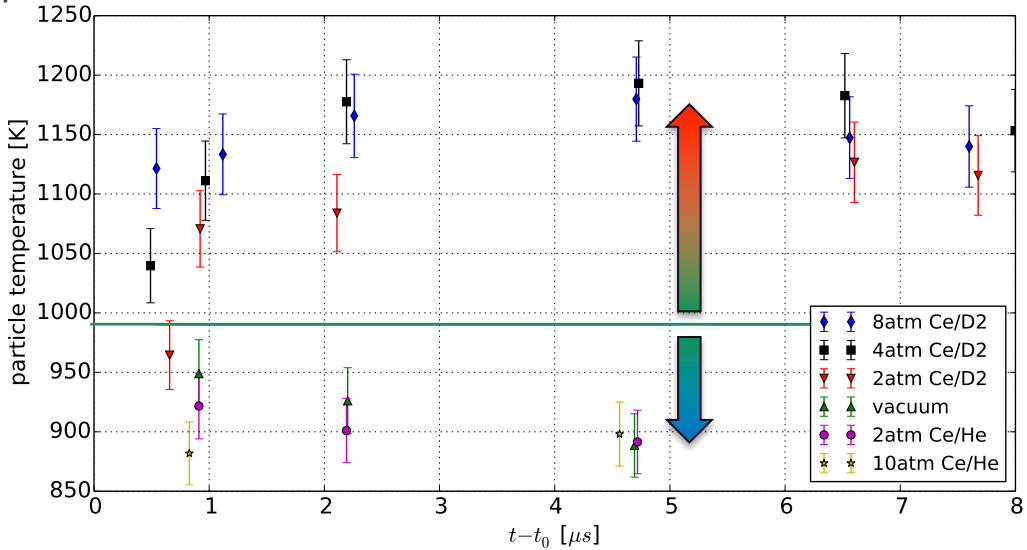
target finishes





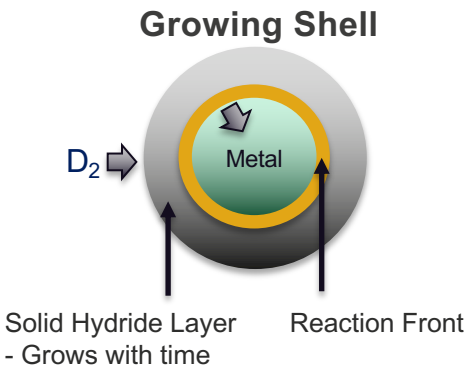
Radiance temperature observations from experiments

- Vacuum and helium media
  - Particle temperature drops (non-reactive)
- Deuterium medium
  - Particle temperature rises 200 K above initial temperature for t < 5 microseconds
  - Exothermic reaction
  - Maximum temperature reached after ~ 5 microseconds
  - Temperature drops for t > 5 microseconds



Physical assumptions for model development

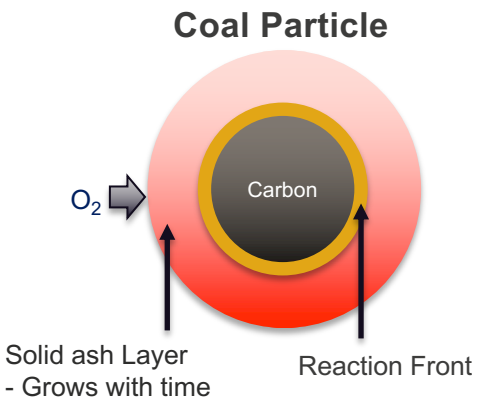
- Assume liquid ejecta droplets are hydrodynamically stable or solid
- Exothermic chemical reactions occur at particle surface
  - Reaction is diffusion-controlled → diffusion through hydride layer
  - Shell remains intact
- Heat transfer is dominated by convection
  - Radiation is small in high density gas with moderate temperature differences
  - Temperature inside particle is uniform → lumped capacitance model
  - Biot number = 0.15 << 1

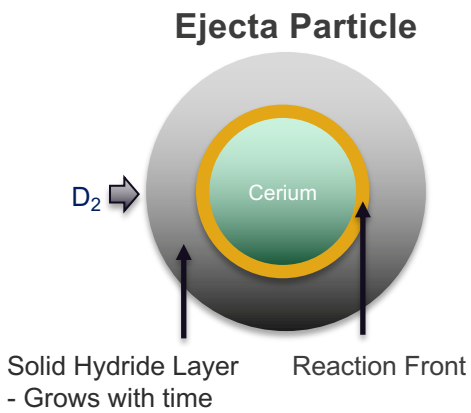


- Buttler LDRD report LA-UR-19-21158
- Buttler et al., Ejecta Transport, Breakup and Conversion, J. Dynamic Behavior Materials, 2017.

Coal combustion analog for solid core

- High ash content coal combustion models are very similar*



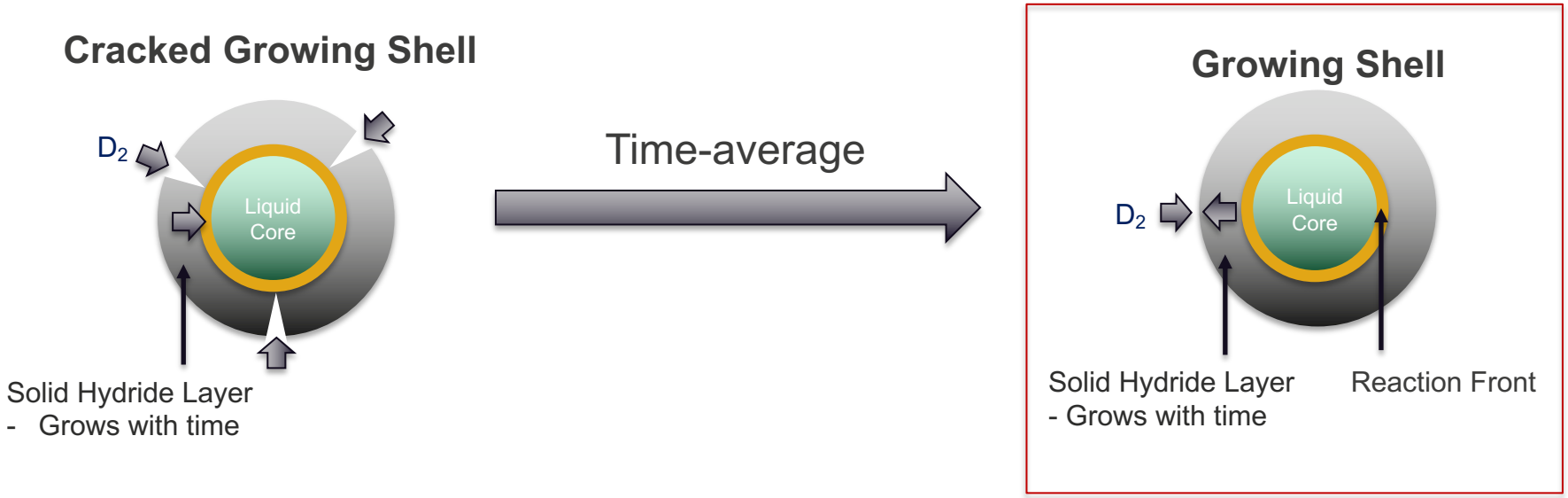


- Ash layer quickly forms on surface of particle
- Oxygen diffusion through ash layer controls carbon oxidation rate
- Thickness of ash layer grows with time

- I. Glassman, Combustion, Ed. 3, 1996

Aluminum oxidation analog for liquid core

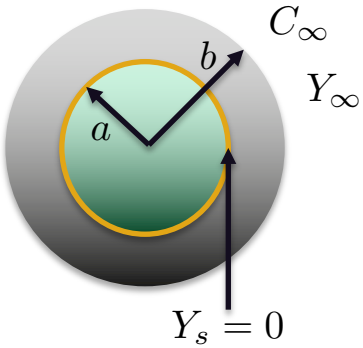
- Aluminum particles oxide in a similar fashion
- Oxide crust cracks → exposes fresh aluminum
  - Rapid kinetics based reaction “heals” cracks
- Simplify by using effective diffusion coefficient → largest uncertainty



Mass Conservation

- D_2 diffuses through CeD_2 layer
- Quasi-steady diffusion through particle (lumped capacitance)

$$\frac{d}{dr}\left(r^2\frac{dY}{dr}\right) = 0 \quad \Rightarrow \quad Y(r) = Y_\infty \frac{ab}{a-b}\left(\frac{1}{r} - \frac{1}{a}\right)$$



- Deuterium diffusion rate through particle $\dot{m}_{D_2} = \mathcal{D} 4\pi ab \frac{\rho_\infty Y_\infty}{b-a}$

- Cerium consumption rate $Ce + D_2 \rightarrow CeD_2$

  (i kg) Ce + (1 kg) D_2 → (i+1 kg) CeD_2 $\quad i = \frac{W_{D_2}}{W_{Ce}} \quad \dot{m}_{Ce} = i\dot{m}_{D_2}$

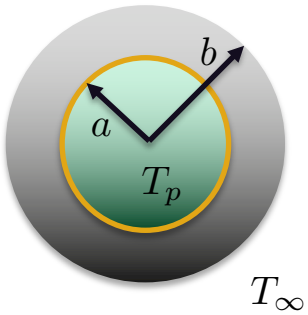
- Heat release rate $\dot{Q}_r = \dot{m}_{Ce} h_f$

- Assume outer radius remains relatively constant

- Rate of change of inner cerium mass $\dot{m}_{Ce} = \rho_{Ce} 4\pi a^2 \frac{da}{dt}$

- Obtain ODE for inner radius $\frac{da}{dt} = i\frac{\rho_\infty}{\rho_{Ce}}\frac{Y_\infty \mathcal{D}}{b-a}\frac{b}{a}$

Energy Conservation



- Use lumped capacitance model
  - Thermal conduction inside particle faster than across gas/solid interface
  - Temperature is constant inside particle

- Convective heat transfer at surface

$$\dot{Q}_c = Nu\pi k_c 2b(T_\infty - T_p)$$

- Ranz-Marshall correlation (1952) for forced convection

$$Nu = 2 + 0.6Re_r^{1/2}Pr^{1/3}$$

- Energy rate of change for particle

Convection

$$\dot{E}_p = \dot{Q}_r + \dot{Q}_c$$

Reaction

- Energy contained inside particle

$$\dot{E}_p = m_p c_p \frac{dT_p}{dt}$$

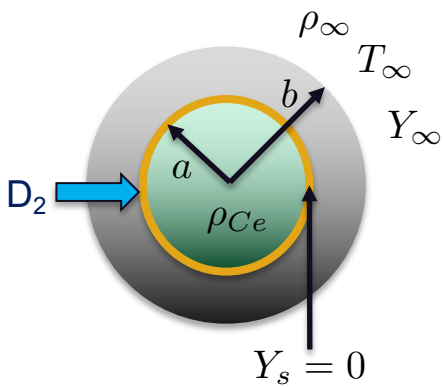
Model Summary

- Mass conservation

$$\frac{da}{dt} = i\frac{\rho_\infty}{\rho_{Ce}}\frac{Y_\infty \mathcal{D}}{b-a}\frac{b}{a}$$

$$i = \frac{W_{D_2}}{W_{Ce}}$$

$$\dot{m}_{Ce} = i4\pi ab\mathcal{D}\frac{\rho_\infty Y_\infty}{b-a}$$



- Energy conservation

$$\frac{de_p}{dt} = \frac{Nu}{2}\frac{c_p}{\tau_T}(T_\infty - T_p) + \frac{\dot{m}_{Ce}h_f}{m_p}$$

Convective heat transfer

Chemical heat release

$$\tau_T = \frac{\rho_p c_p (2b)^2}{12k_c}$$

Thermal relaxation time

- Momentum conservation
  - Focus on early time → constant velocity

Numerical Implementation

Numerical Implementation

- Initial reaction rate is fast
  - Potential division by zero

$$\frac{da}{dt} = i\frac{\rho_\infty}{\rho_{Ce}}\frac{Y_\infty \mathcal{D}}{b-a}\frac{b}{a}$$

- Use n+1/2 to prevent division by zero

- Naive discretization of reaction radius equation is inefficient
  - Accuracy highly dependent on timestep size
  - Numerical integration requires $O(10^5)$ timesteps per particle reaction time
  - Timestep can be much smaller than hydro timestep

- Seek analytical solution for mass conservation equation

Analytical Solution - Transformation

- Assume that all parameters are constant except the reaction radius a
- Integrate to obtain

$$1 - 3\left(\frac{a}{b}\right)^2 + 2\left(\frac{a}{b}\right)^3 = \frac{t}{\tau_r}$$

- Reaction time:

$$\tau_r = \frac{1}{6i}\frac{\rho_{Ce}}{\rho_\infty}\frac{b^2}{\mathcal{D}Y_\infty}$$

- Cubic equation:

$$0 = 2r^3 - 3r^2 + (1 - t_*) \qquad t_* = t/\tau_r \qquad r = a/b$$

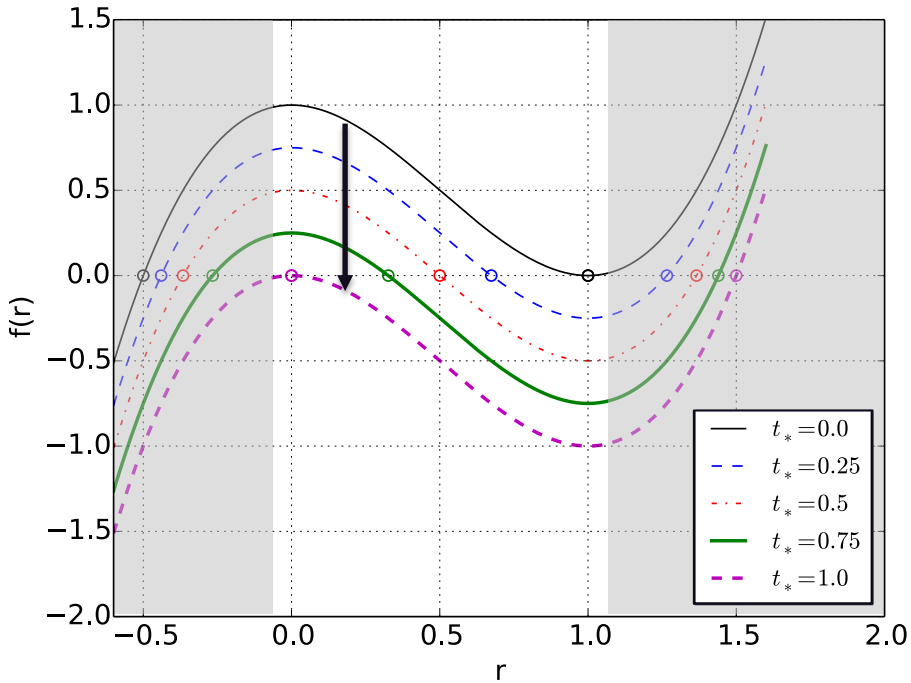
- Solution validity:

$$t_* \in [0, 1] \qquad r \in [0, 1]$$

Graphical representation of solution

- Valid within $r \in [0, 1]$
- Solution starts at $r = 1$
- Moves left with increasing t_*
- Final value at $r = 0$

- Middle roots are valid



Analytical Solution

- Transform into depressed cubic: $$0 = 2y^3 - \frac{3}{2}y + \frac{1 - t_*}{2} \qquad y = r + 1/2$$

- Roots:

$$r_1 = \cos\theta + \frac{1}{2}$$

$$\boxed{r_2 = \cos\left(\frac{2\pi}{3} - \theta\right) + \frac{1}{2} \qquad \theta = \frac{1}{3}\arccos(2t_* - 1)}$$

$$r_3 = \cos\left(\frac{2\pi}{3} + \theta\right) + \frac{1}{2}$$

- Solution: $$r(t_*) = \cos\left(\frac{2\pi}{3} - \theta(t_*)\right) + \frac{1}{2} \qquad \theta(t_*) = \frac{1}{3}\arccos(2t_* - 1)$$

Numerical Implementation

- At end of previous timestep $t_* = t_*^n$ → $t_*^{n+1} = t_*^n + \Delta t_*$
- Radius at t_*^n $r^n = r(t_*^n) = \cos\left(\frac{2\pi}{3} - \theta^n\right) + \frac{1}{2}$
- Radius at t_*^{n+1} $r^{n+1} = r(t_*^{n+1}) = \cos\left(\frac{2\pi}{3} - \theta^{n+1}\right) + \frac{1}{2}$
- Express new radius as radius change

$$\Delta r_n^{n+1} = r^{n+1} - r^n = \cos\left(\frac{2\pi}{3} - \theta^{n+1}\right) - \cos\left(\frac{2\pi}{3} - \theta^n\right)$$

Transform to original dimensional variables

- Dimensional reaction radius: $a^n = br^n$ $\quad a^{n+1} = br^{n+1}$
- New reaction radius: $a^{n+1} = a^n + b\Delta r_n^{n+1}$
- Time nondimensionalization assumes τ_r const: $t_*^n = \dfrac{t^n}{\tau_r}$
- Accommodate variable properties by
  – Approximate as constant during timestep $\tau_r \to \tau_r^n$

$$\theta^n = \frac{1}{3}\arccos\left(2\frac{t^n}{\tau_r^n} - 1\right) \qquad \theta^{n+1} = \frac{1}{3}\arccos\left(2\left[\frac{t^n + \Delta t}{\tau_r^n}\right] - 1\right)$$

Whole step discretization

$$a^{n+1} = a^n + b\Delta r_n^{n+1}$$

$$\Delta r_n^{n+1} = \cos\left(\frac{2\pi}{3} - \theta^{n+1}\right) - \cos\left(\frac{2\pi}{3} - \theta^n\right)$$

$$\frac{1}{3}\arccos\left(2\left[\frac{t^n + \Delta t}{\tau_r^n}\right] - 1\right) = \theta^{n+1}$$

$$\theta^n = \frac{1}{3}\arccos\left(2\frac{t^n}{\tau_r^n} - 1\right)$$

- Solution is exact for constant τ_r

Integration with energy equation

$$\frac{de_p}{dt} = \frac{Nu}{2}\frac{c_p}{\tau_T}(T_\infty - T_p) + \frac{\dot{m}_{Ce} h_f}{m_p}$$

- Energy equation is integrated with first order explicit scheme
  - Use average particle temperature and mass conversion rate at n+1/2

$$\frac{\Delta e_p}{\Delta t} = \frac{Nu}{2}\frac{c_p}{\tau_T}(T_\infty - T_p^{n+1/2}) + \frac{\dot{m}_{Ce}^{n+1/2} h_f}{m_p}$$

$$T^{n+1/2} = \frac{T_p^n + T_p^{n+1}}{2} \qquad e^{n+1/2} = c_p T_p^{n+1/2}$$

$$\frac{\Delta e_p}{\Delta t}\left(1 + \frac{\Delta t}{\tau_T}\frac{Nu}{4}\right) = \frac{Nu}{2}\frac{c_p}{\tau_T}(T_\infty - T_p^n) + \frac{\dot{m}_{Ce}^{n+1/2} h_f}{m_p} \qquad e^{n+1} = e^n + \Delta e$$

- Need mass conversion rate at n+1/2

Mass conversion rate at n+1/2

- Mass conversion rate: $$\dot{m}_{Ce}^{n+1/2} = i4\pi a^{n+1/2} b\mathcal{D} \frac{\rho_\infty Y_\infty}{b - a^{n+1/2}}$$

- Radius at n+1/2: $$a^{n+1/2} = a^n + b\Delta r_n^{n+1/2}$$

$$\Delta r_n^{n+1/2} = \cos\left(\frac{2\pi}{3} - \theta^{n+1/2}\right) - \cos\left(\frac{2\pi}{3} - \theta^n\right)$$

$$\theta^{n+1/2} = \frac{1}{3}\arccos\left(2\left[\frac{t^n + \Delta t/2}{\tau_r^n}\right] - 1\right)$$

Obtain final state at n+1

- Find updated energy

$$\frac{\Delta e_p}{\Delta t}\left(1+\frac{\Delta t}{\tau_T}\frac{Nu}{4}\right)=\frac{Nu}{2}\frac{c_p}{\tau_T}(T_\infty-T_p^n)+\frac{\dot{m}_{Ce}^{n+1/2}h_f}{m_p}$$

$$e^{n+1}=e^n+\Delta e$$

- Integrate radius another half step

$$a^{n+1}=a^{n+1/2}+b\Delta r_{n+1/2}^{n+1}$$

$$\Delta r_{n+1/2}^{n+1}=\cos\left(\frac{2\pi}{3}-\theta^{n+1}\right)-\cos\left(\frac{2\pi}{3}-\theta^{n+1/2}\right)$$

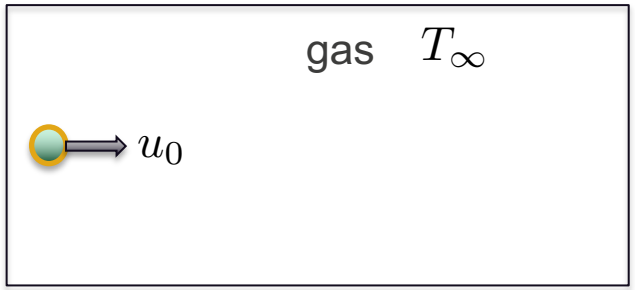
$$\frac{1}{3}\arccos\left(2\left[\frac{t^{n+1/2}+\Delta t/2}{\tau_r^n}\right]-1\right)=\theta^{n+1}$$

$$\theta^{n+1/2}=\frac{1}{3}\arccos\left(2\left[\frac{t^n+\Delta t/2}{\tau_r^n}\right]-1\right)$$

Verification, Validation and Performance

Model verification

- Setup simple verification problem
  - Single particle inside domain
  - Constant velocity
  - Constant gas temperature



- Reaction radius ODE → constant diffusion coefficient

$$\frac{da}{dt} = i\frac{\rho_\infty}{\rho_{Ce}}\frac{Y_\infty \mathcal{D}}{b-a}\frac{b}{a}$$

- Energy equation
  - Verified each term independently
  - Adiabatic heat release
  - Thermal relaxation

$$\frac{de_p}{dt} = \frac{Nu}{2}\frac{c_p}{\tau_T}(T_\infty - T_p) + \frac{\dot{m}_{Ce}h_f}{m_p}$$

Validation – Change to post-shock conditions

- Tested model with Ce experiment at 4 and 8 atm pre-shock conditions

Initial Conditions

Initial Gas Pressure	**4**	**8**	**atm**
Shocked Gas Temperature	720	740	K
Shocked Metal Temperature	990	990	K
Shocked Gas Density	2.3	4.5	kg/m^3
Relative Velocity	480	470	m/s

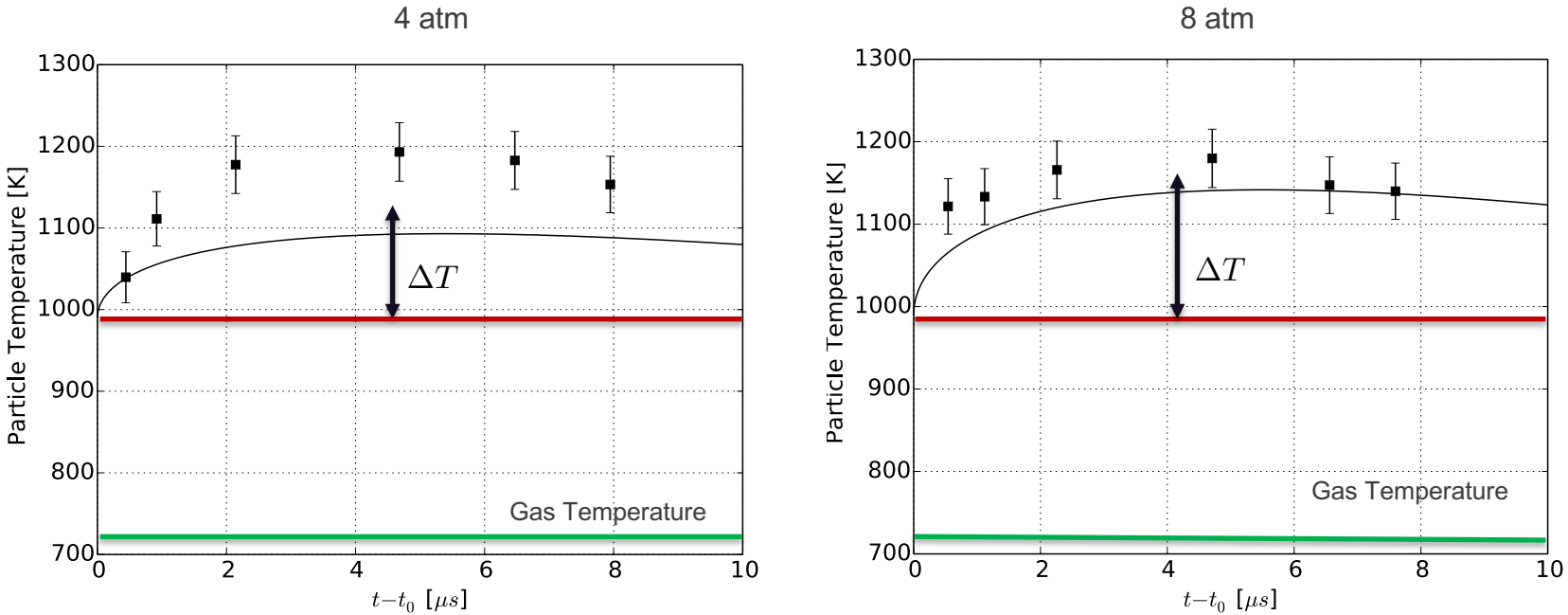
Gas Properties

Specific heat	7.25 kJ/kg-K
Thermal conductivity	0.138 W/m-K
Viscosity	1.72E-5 Pa-s
Specific heat ratio	1.4

Metal Material Properties

Specific heat	269 J/kg-K
Particle diameter	13 micron
Particle density	6.7 g/cc
CeD_2 heat of formation	210 kJ/mol
Diffusion coefficient	$\mathcal{D}(T_p) = 2.083589\text{E-}14 \cdot T_p^{1.941438}$

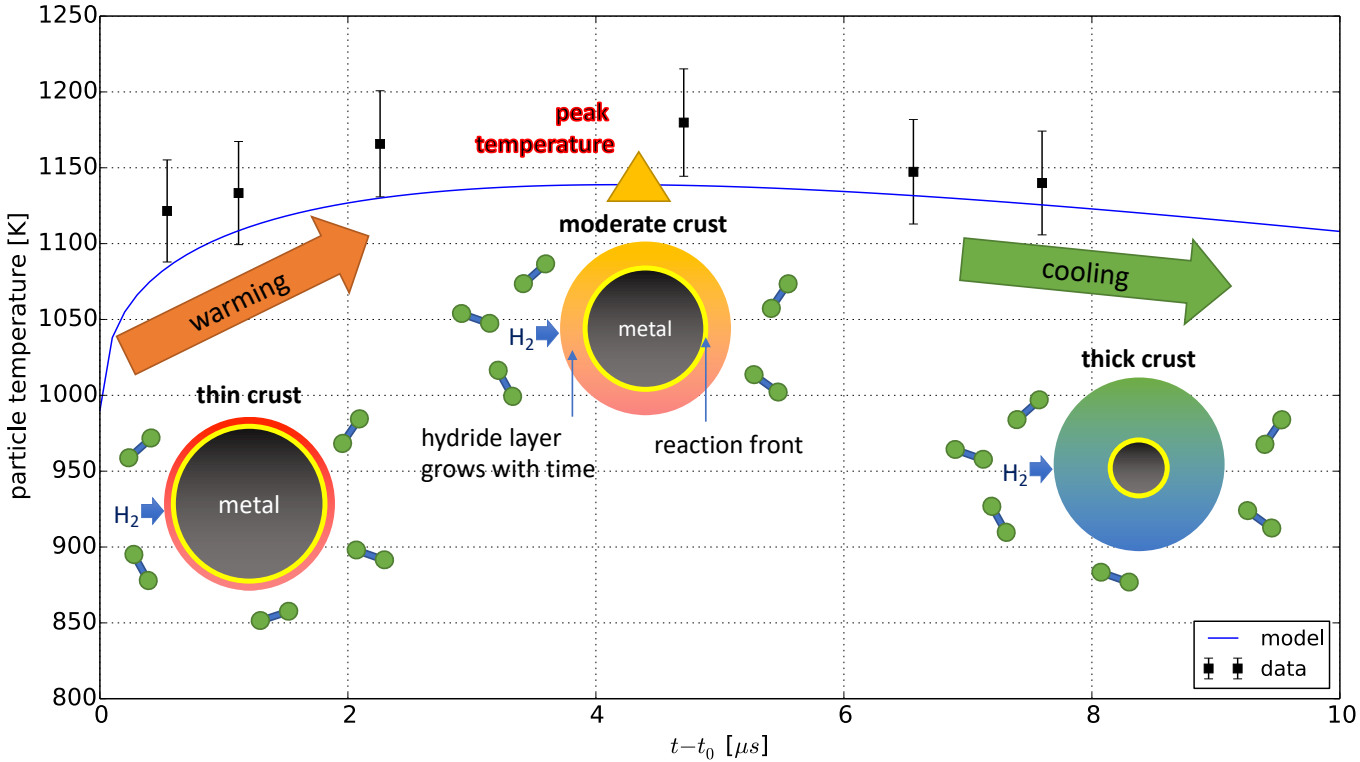
Particle temperature comparison



- Model roughly captures the 200 K increase in particle temperature
- Predicted temperatures are low for 4 atm case
- Relatively accurate for 8 atm case

Competing processes

- 8 atm data
- Physical assumptions seem reasonable

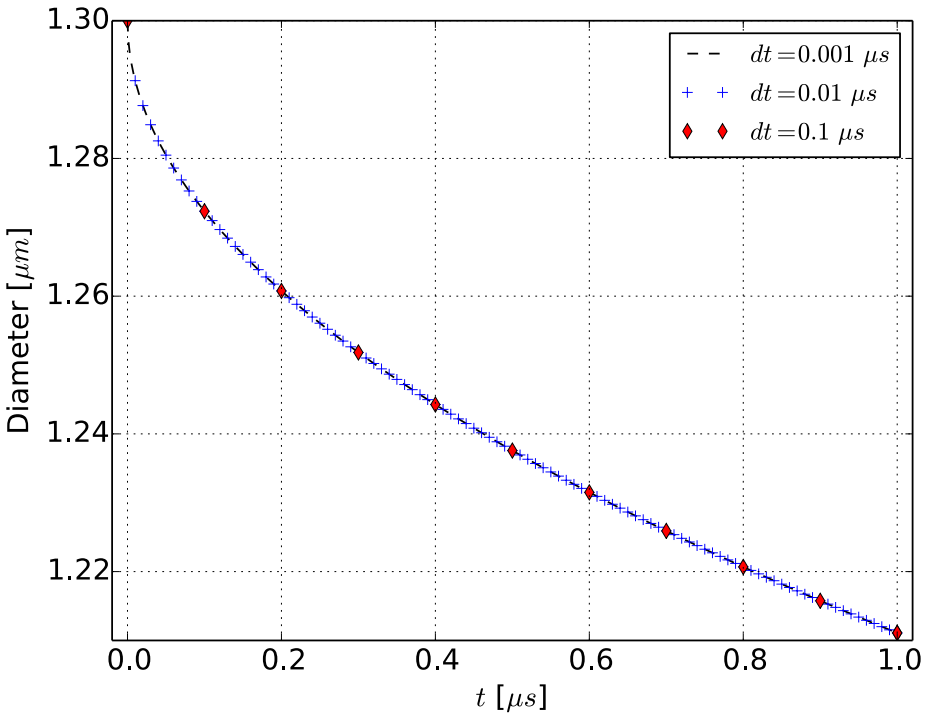


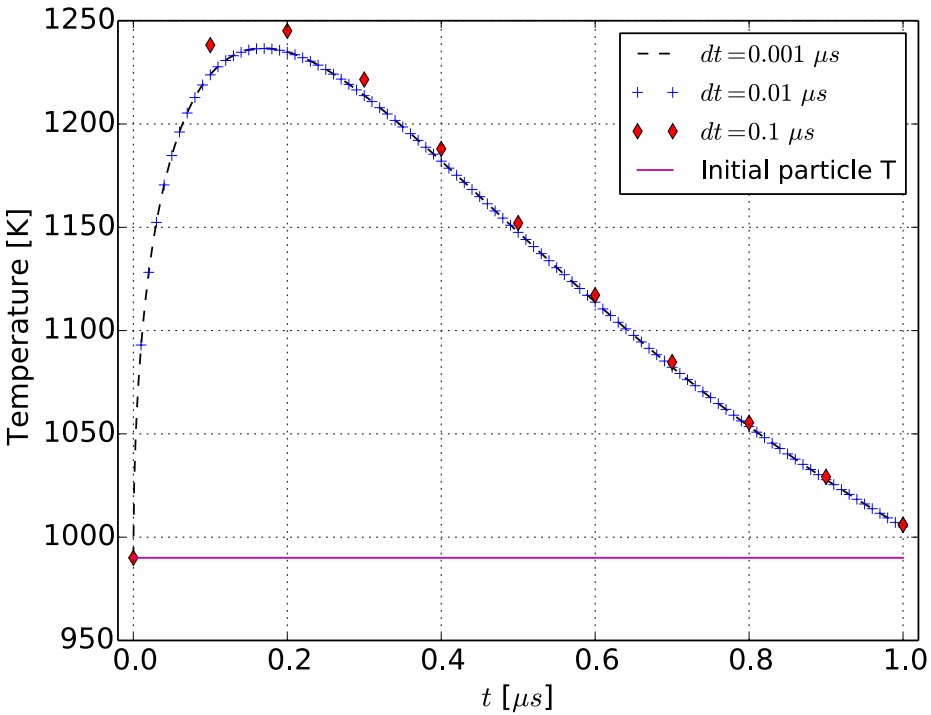
Model performance with small particles

- Particles in experiments are 13 microns in diameter → large
- Test model with 1.3 and 0.13 micron particles

1.3 micron particles

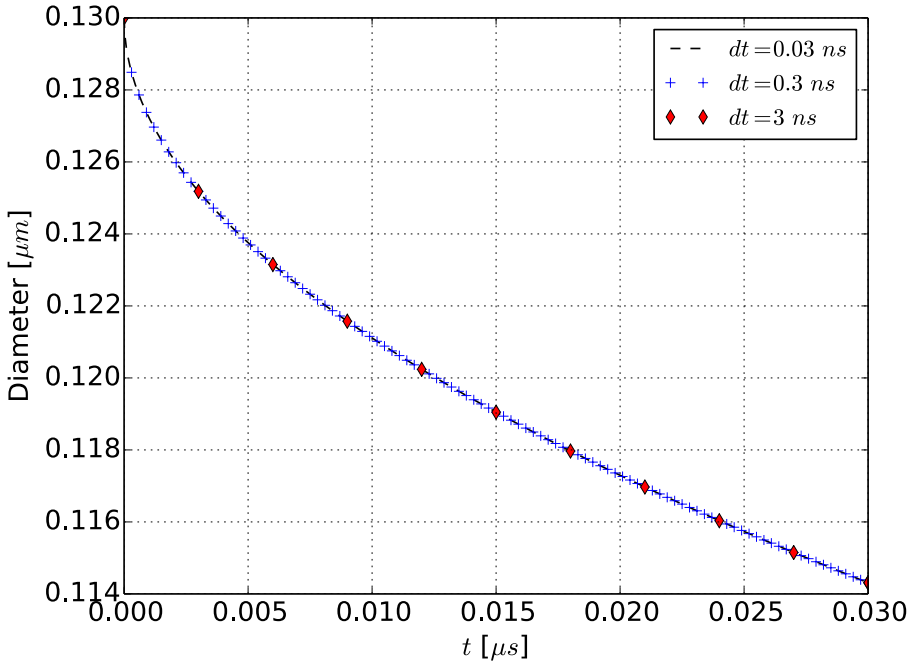
- Reaction radius independent of timestep size
- Temperature slightly overestimated for largest timestep

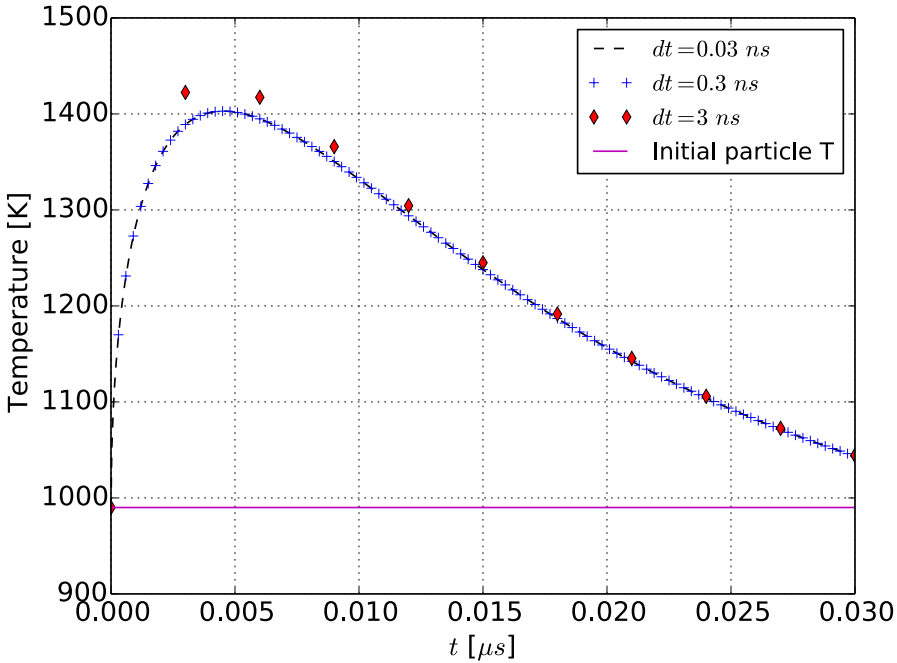




0.13 micron particles

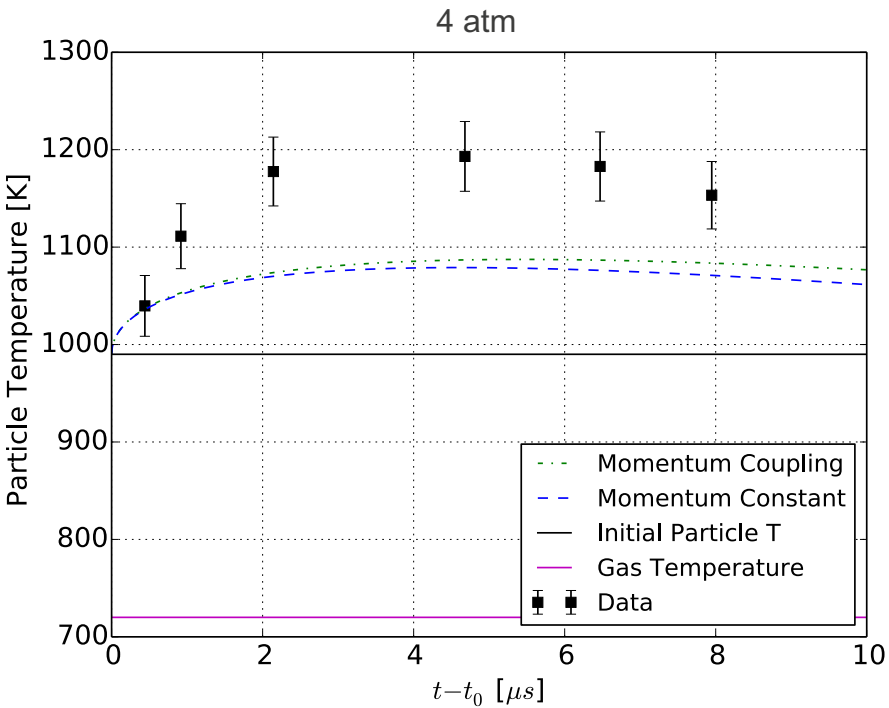
- Reaction radius independent of timestep size
- Smaller timesteps at beginning increase accuracy
- May need adaptive timestep

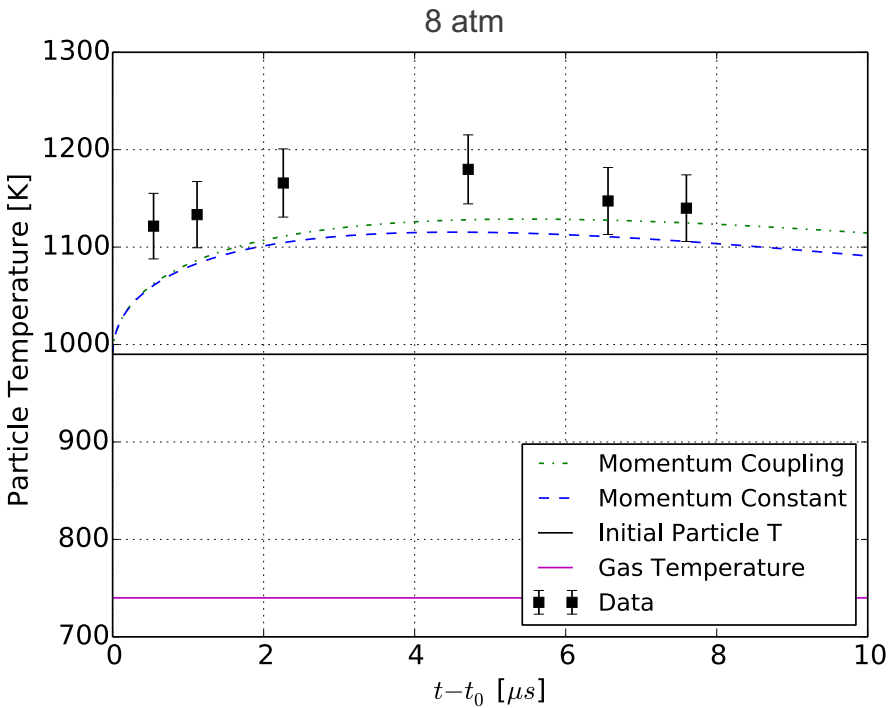




Momentum Coupling effects

- Momentum coupling slows particles
  - Reduces heat conduction → increases particle temperature



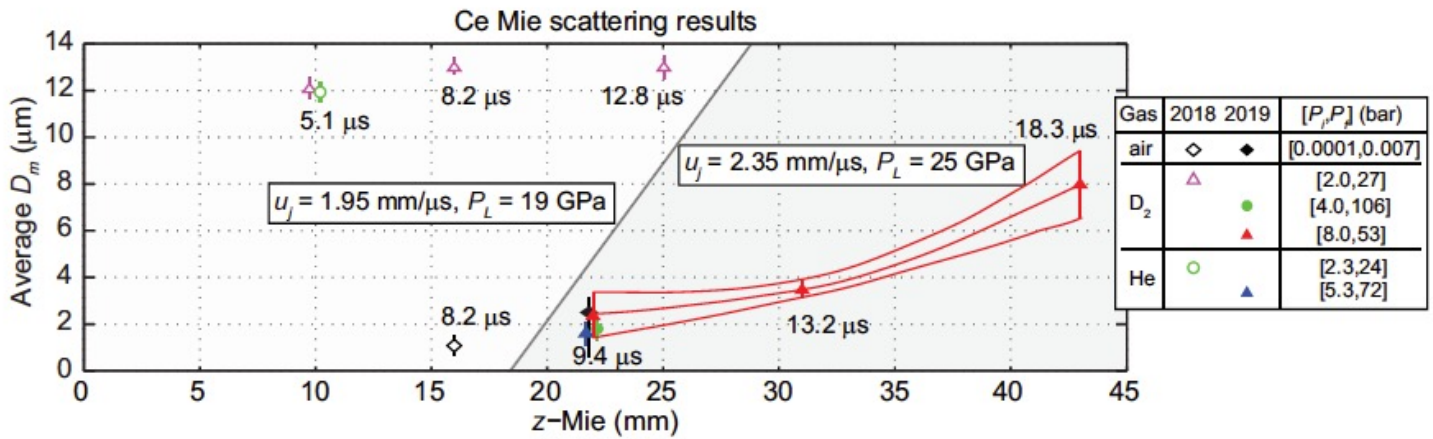


Summary

- Simple diffusion-controlled reaction model
- Roughly approximates temperature increase of cerium ejecta particles
  - No tuning of parameters
  - Greatest uncertainty exist in diffusion coefficient
- Need more detail to accurately capture temperature rise
- Exact solution increases accuracy and minimizes computational cost
  - Both reaction radius and particle temperature
- Particle temperatures are reasonably accurate for range of sizes
  - Adaptive time-stepping may be required for greater accuracy

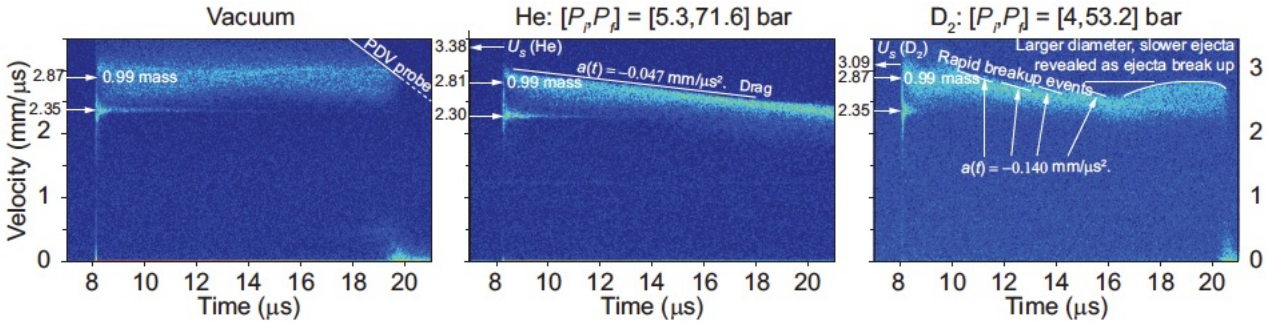
Future Work

- Unstable liquid ejecta particle model
  - Droplets are not hydrodynamically stable
  - Free to deform and breakup into smaller droplets
  - Hydride reactions occur simultaneously
- 2019 liquid cerium ejecta experimental results very interesting
  - Average particle size increases with time/distance for reacting cases



2019 liquid Ce ejecta experiments (PI: Buttler, [20170082DR])

- Cerium ejecting into
  - Vacuum → constant velocity
  - Helium → monotonically decreasing particle velocities
  - Deuterium → non-monotonic velocity reduction



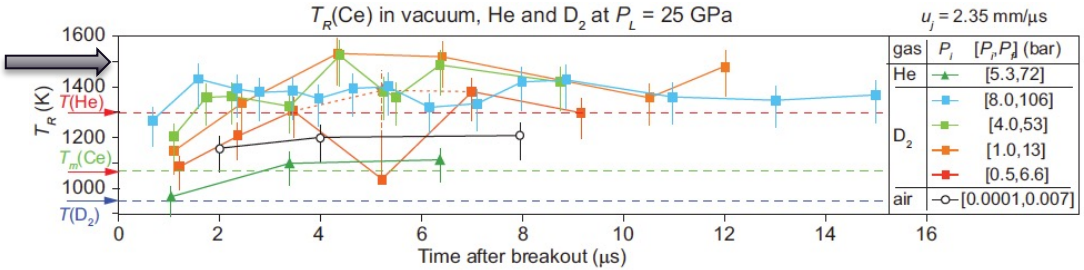
Theories

- Larger diameter, slower ejecta are revealed as leading ejecta breakup
- Hydride shell holds the liquid droplet together and delays breakup

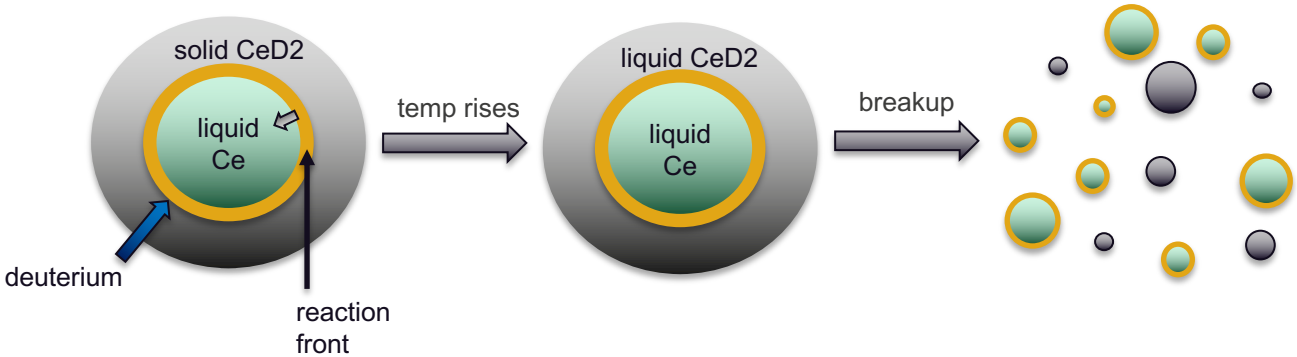
- What is the mechanism that causes the loss of shell structural integrity?

What causes shell structure to disappear?

Max temperature

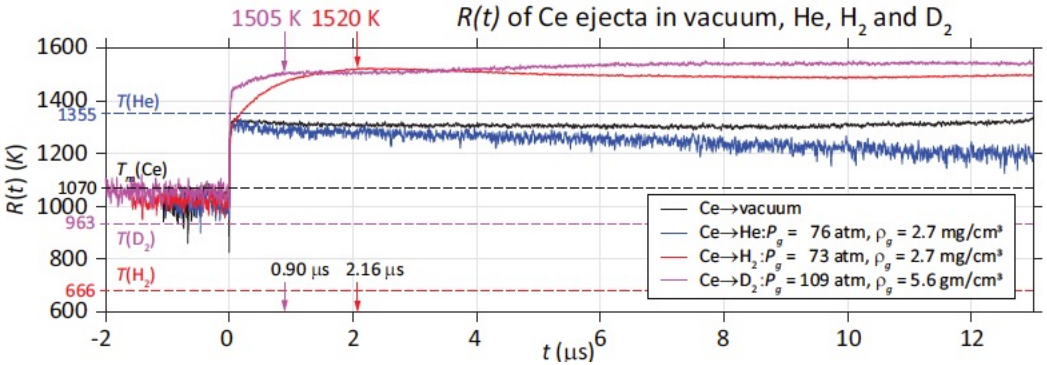


- Liquid Ce particles reach a maximum temperature around 1400-1500K
  - Temperature dependent strength or melting?
  - Implicitly assumes that shell forms before the droplets breakup fully: We → 10



Future Work

- Higher fidelity Radiance data confirms max temperature



- Need to know particle temperature instead of radiance
  - Buttler & Hartsfield working on emissivity free measurement technique