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Title: Progress in understanding reactive ejecta experiments

Author(s): Regele, Jonathan David
Ouellet, Frederick

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Los Alamos
NATIONAL LABORATORY

Progress in understanding reactive ejecta experiments

Jonathan D. Regele and Fred Ouellet

XCP-4 Continuum Models and Numerical Methods

Contributors: J. D. Schwarzkopf, W. T. Buttler, R. K. Schulze, A. K. Harrison, D. G. Sheppard,

August 30, 2022

LA-UR-?????

Outline

- Background and motivation
- Solid ejecta
- Liquid ejecta
- Conclusions

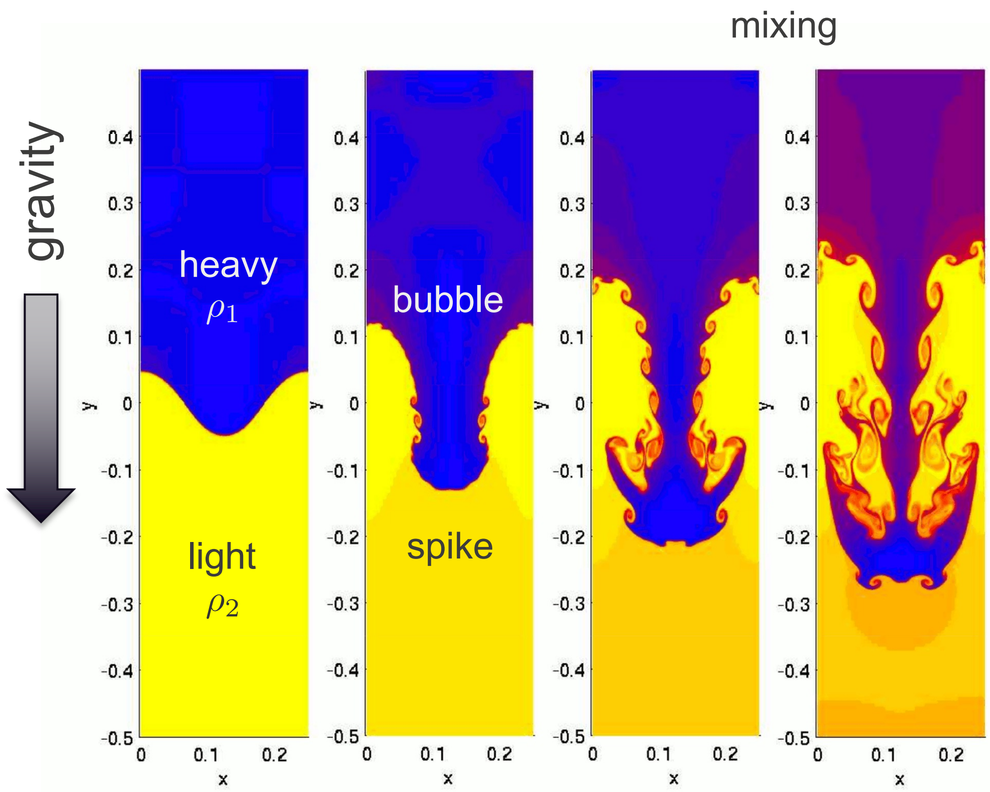
Background and Motivation

Rayleigh-Taylor Instability

- Two unmixed hydrodynamically unstable fluids
- Gravitational field pulls heavy into light fluid
- Spike and bubble form
- Atwood number characterizes mixing behavior

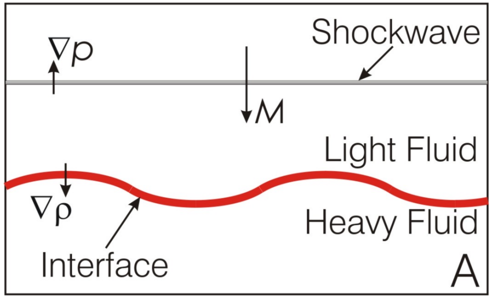
$$A = \frac{\rho_1 - \rho_2}{\rho_1 + \rho_2}$$

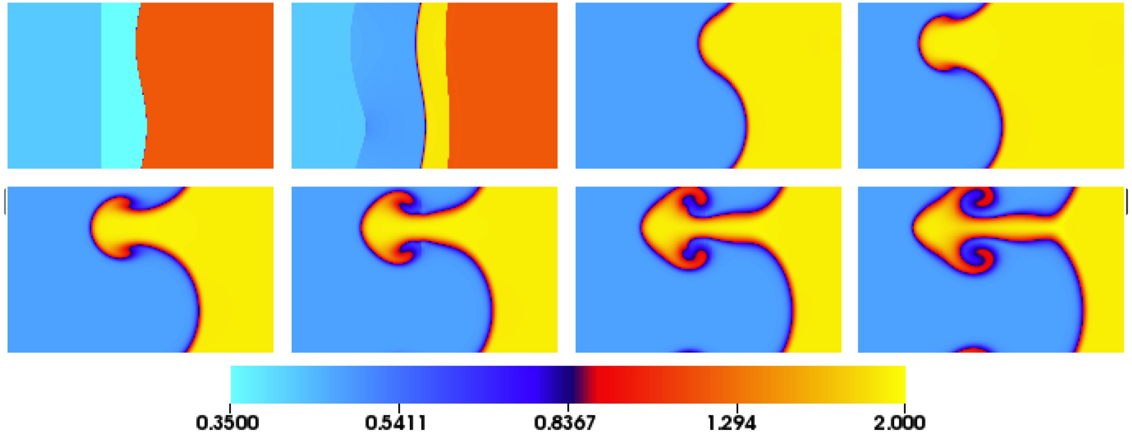
- Common in many natural and engineering settings
  - Radiant heating in floor



Richtmyer-Meshkov Instability

- Impulsive acceleration limit case of R-T instability
- Shock wave impacts perturbed interface
- Shock can pass from light to heavy or heavy to light
- Baroclinic torque stretches the interface → mixing





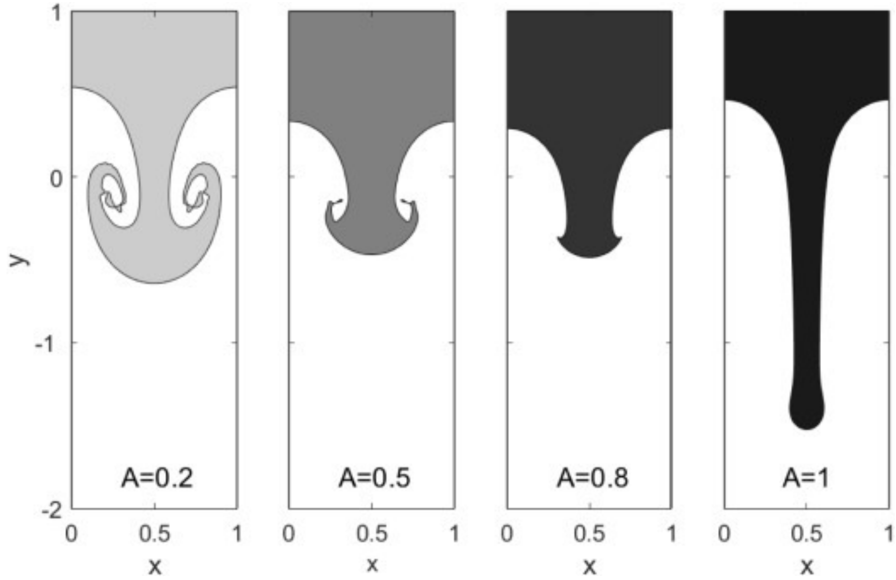
Frapolli, N.; Chikatamarla, S.; Karlin, *Entropy* **2020**, *22*, 370

Liquid-gas RM instabilities

- Mixing rate decreases as density difference increases $\rho_1 \gg \rho_2$

$$A \to 1$$

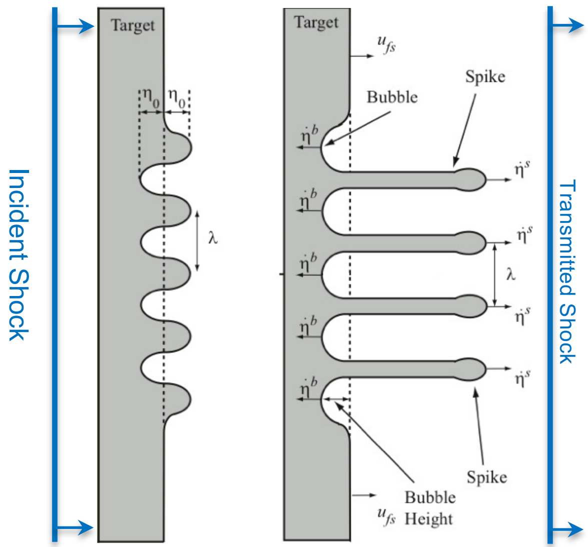
- In liquid-gas interfaces, long spikes form
- Mixing occurs as surface-tension linked breakup



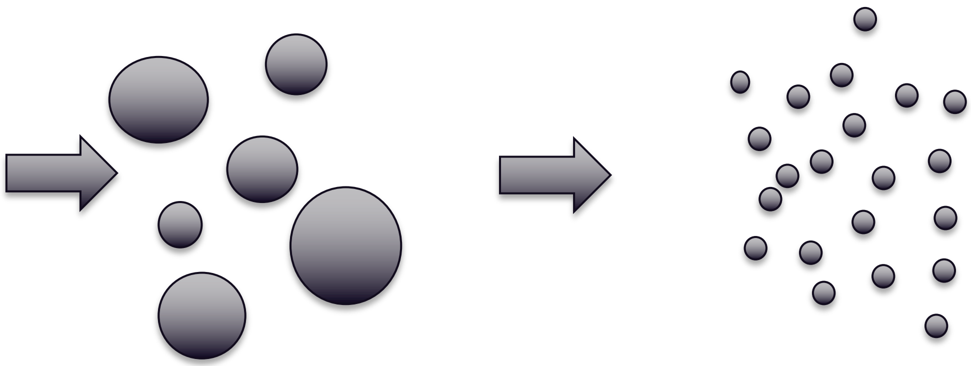
S. Shin, S-I. Sohn, W. Hwang, European Journal of Mechanics - B/Fluids, 91, 2022, pp141-151.

Two-phase Richtmyer-Meshkov instability

Source



Droplet breakup



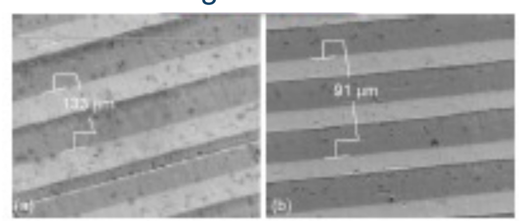
- Shock from high explosive (HE) impacts solid target material with etched surface perturbation
- Shock melts solid material and forms liquid spikes
- Spikes break into droplets, which may or may not break up further
- Chemical reactions may occur

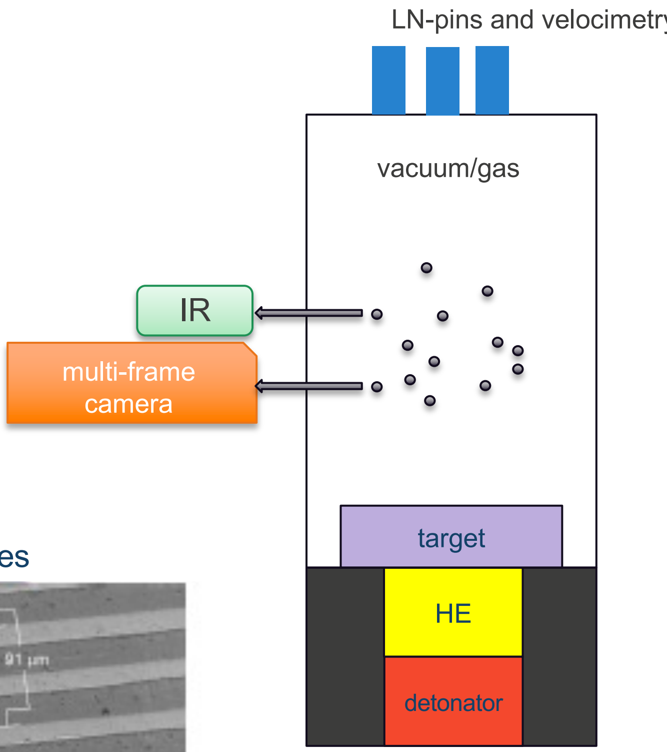
Solid ejecta model

Cerium ejecta experiments

- 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
- LDV for ejecta velocities
- Lithium niobate-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

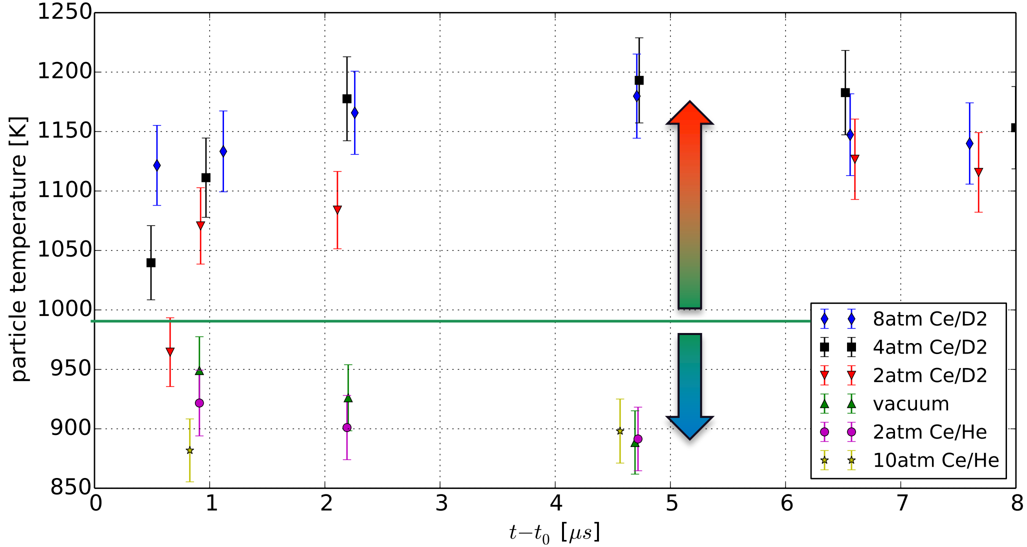




- Buttler LDRD report LA-UR-19-21158
- Buttler et al., Ejecta Transport, Breakup and Conversion, J. Dynamic Behavior Materials, 2017.

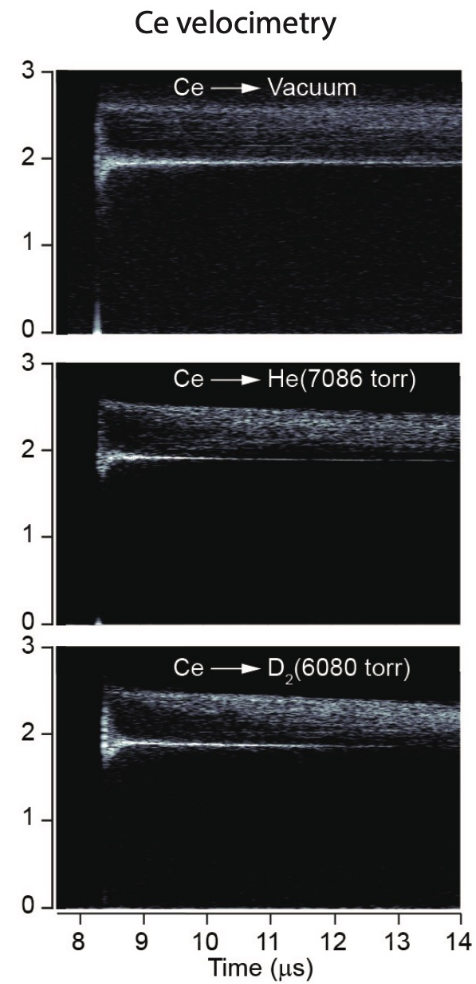
Radiance temperature observations from experiments

- 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



Solid ejecta velocities

- Velocity is constant when ejected into vacuum
- Velocity decreases in gas
- Particles slightly denser in reacting gas

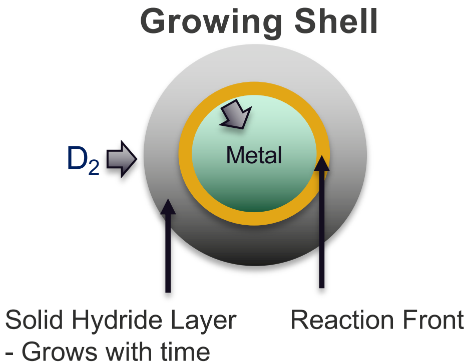


Physical assumptions for model development

- Similar to high ash content coal and aluminum particle combustion

- Assume liquid ejecta droplets are hydrodynamically stable or solid

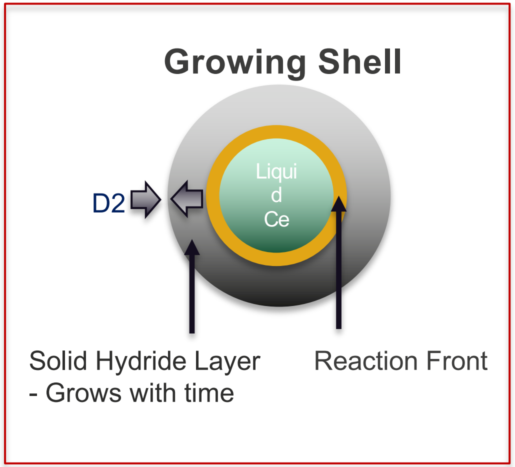
- Exothermic chemical reactions occur at metal surface
  - Reaction is diffusion-controlled → diffusion through hydride layer
  - Shell remains intact
  - Outer diameter is constant → neglect density change in hydride layer

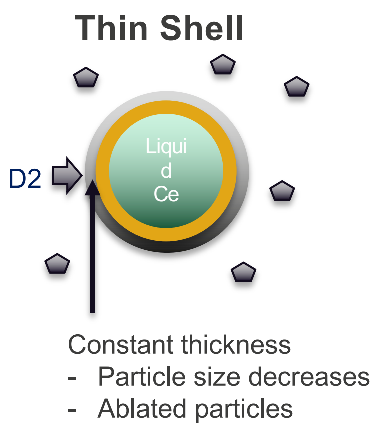


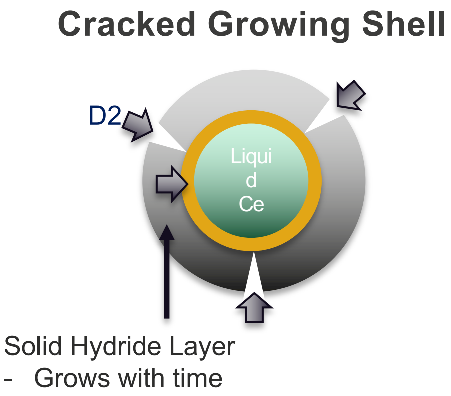
- Heat transfer is dominated by convection
  - Radiation is small in high density gas with moderate temperature differences
  - Temperature inside particle is uniform → lumped capacitance model
  - Biot number = 0.15 << 1

Additional Considerations

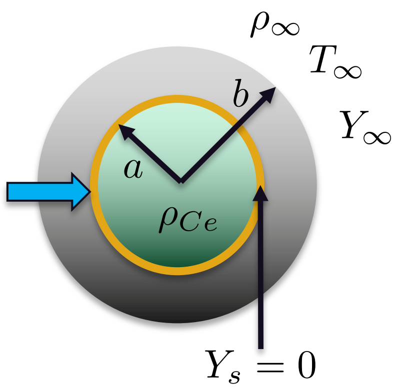
- Cerium surface kinetics are rapid and quickly form a hydride film/shell
- Schulze calculations suggest that kinetic reactions are only rate-limiting at early stages
  - Reactions are likely to be diffusion limited
- Unclear how structurally sound the hydride layer is
  - Does it grow in size until reaction is finished?
  - Does the hydride layer ablate/flake off?
  - Is the hydride layer cracked creating localized regions of higher reactivity?







Model Overview



- Irreversible diffusion controlled reaction

$$\dot{m}_{Ce} = i 4\pi a b \mathcal{D} \frac{\rho_\infty Y_\infty}{b-a}$$

- Outer radius is constant with inner radius decreasing
  - Mass conservation

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

- Heat released from reaction

$$\dot{Q}_r = \dot{m}_{Ce} h_r$$

- Convective heat transfer

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

- Energy conservation

$$\dot{E}_p = \dot{Q}_r + \dot{Q}_c \qquad \dot{E}_p = m_p c_p \frac{dT_p}{dt}$$

- Integrate ODE along with radius change
  - System of 2 ODE's
  - Also include particle momentum change → 3 ODEs

Stoichiometric mass ratio

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

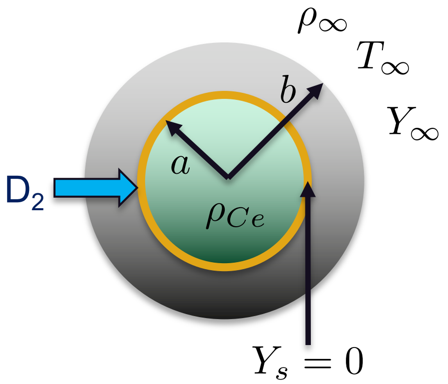
Model Summary

- 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

$$\frac{dT_p}{dt} = \frac{Nu}{2}\frac{1}{\tau_T}(T_\infty - T_p) + \frac{\dot{m}_{Ce}h_f}{m_p c_p}$$

Convective heat transfer — Chemical heat release

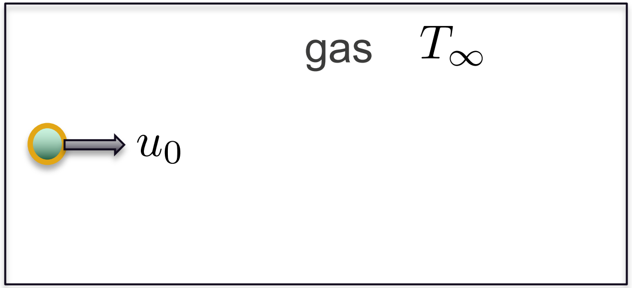
$$\tau_T = \frac{\rho_p c_p (2b)^2}{12k_c}$$

Thermal relaxation time

- Momentum conservation
  - Focus on early time → constant velocity

Verification and Validation

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



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

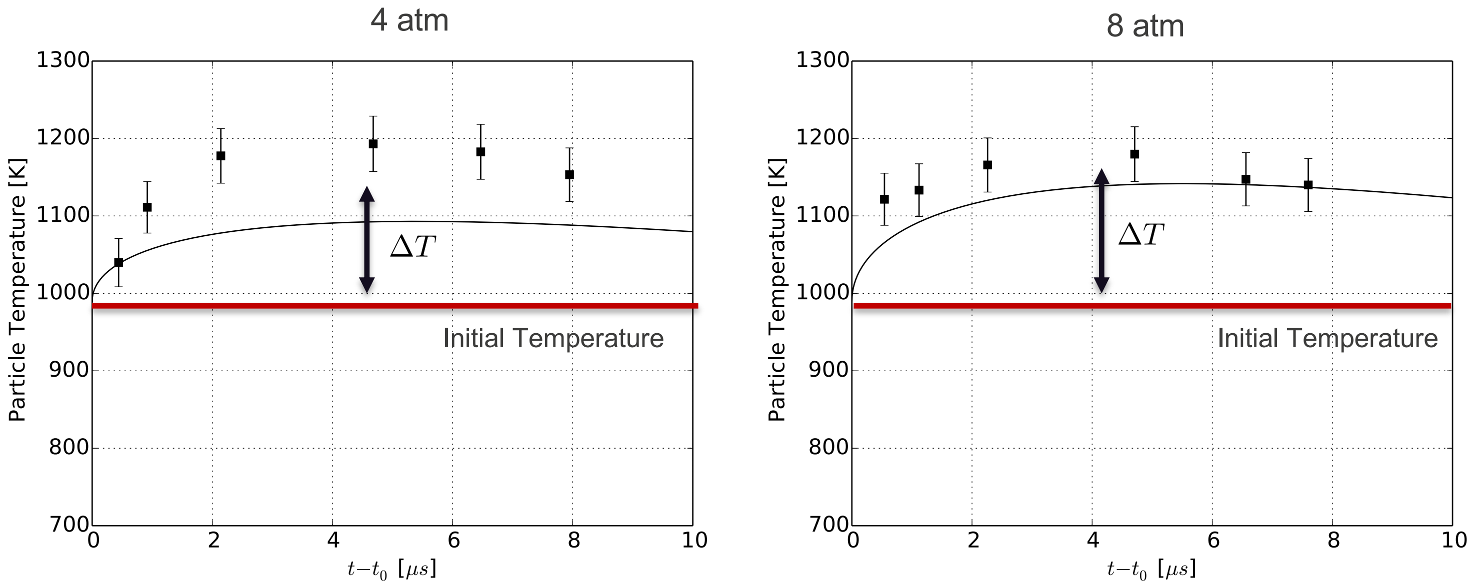
Metal Material Properties

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	$\mathcal{D}(T_p) = 2.083589\text{E-}14 \cdot T_p^{1.941438}$

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

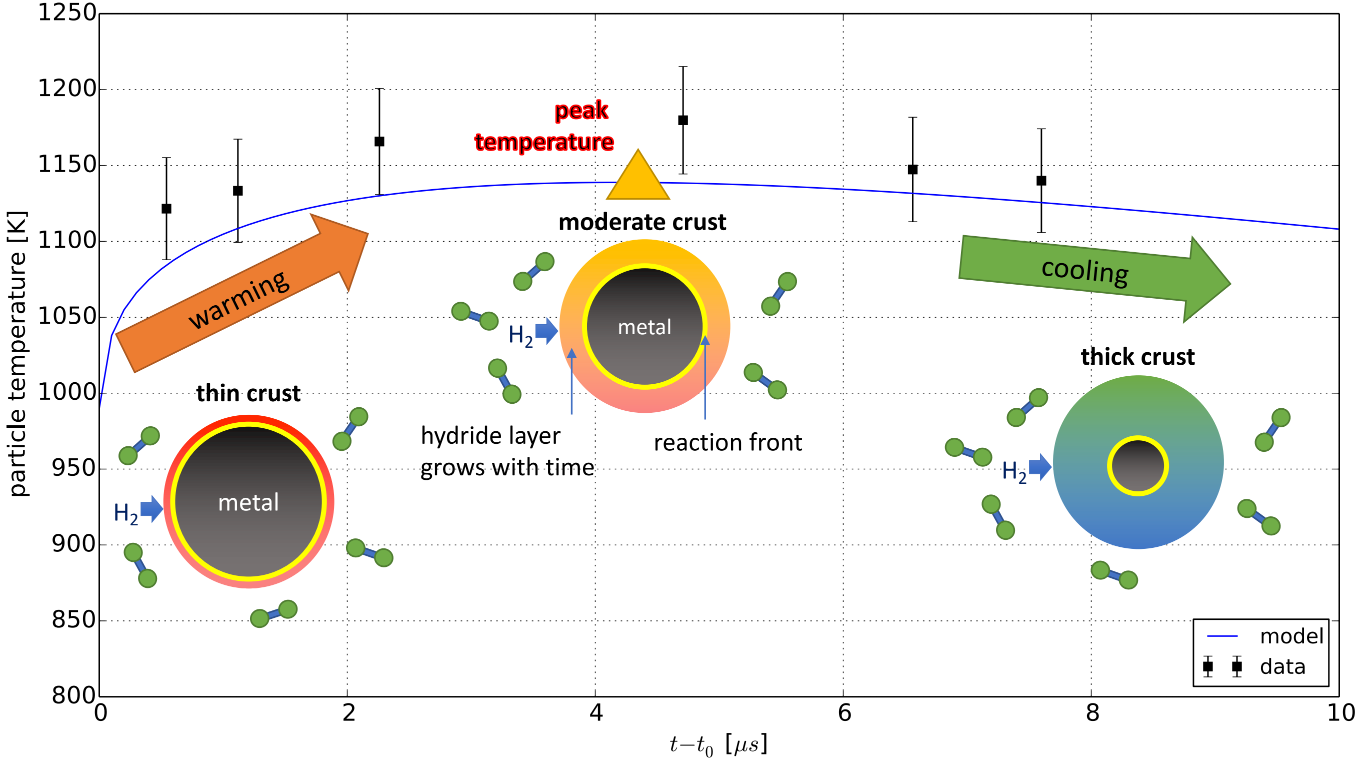
Model temperature comparison with experiments



- Model roughly captures the 100-200 K temperature increase above initial temperature
- Predicted temperatures are low for 4 atm case
- Relatively accurate for 8 atm case considering uncertainty in parameters
- More detailed simulations are anticipated to improve upon these differences

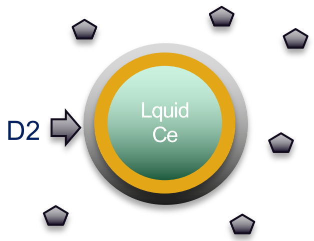
Heat release and transfer processes compete

- Model reproduces particle temperature rise and fall
- Physical assumptions seem reasonable



Constant crust thickness variation

- Hydride crust has constant thickness
- Extra mass flakes off
- Particle radius becomes $r_o = \min(b, a(t) + \delta)$



- Results for solid ejecta vary only slightly with this variant during heating
  - Thicknesses in range 20-50nm
  - Temperature decline is slower

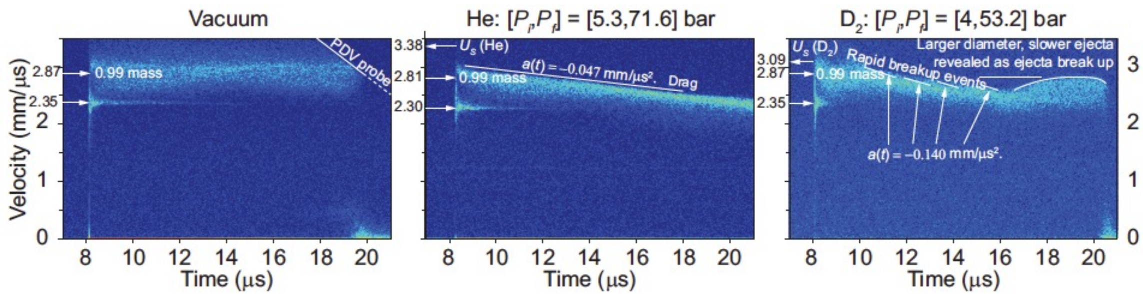
Solid ejecta conclusions

- Simple diffusion-controlled reaction model
- Roughly approximates temperature rise and fall of cerium ejecta particles
- Subsequent work verifies many assumptions are reasonable
  - Nearly constant momentum
  - Constant diameter particle → account for density difference in hydride shell
- Need to create a more accurate validation model

Liquid ejecta

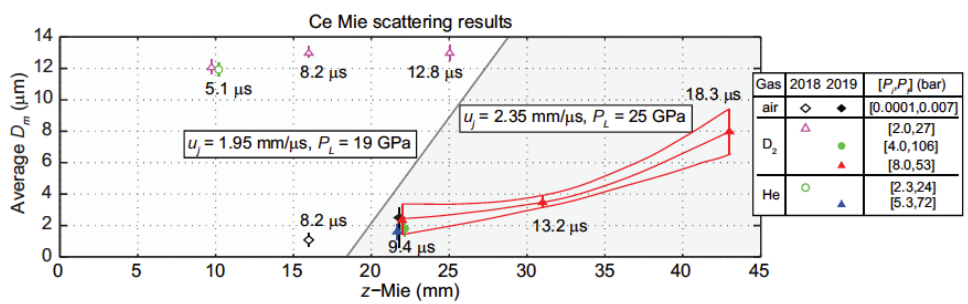
2019 liquid Ce ejecta experiments

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



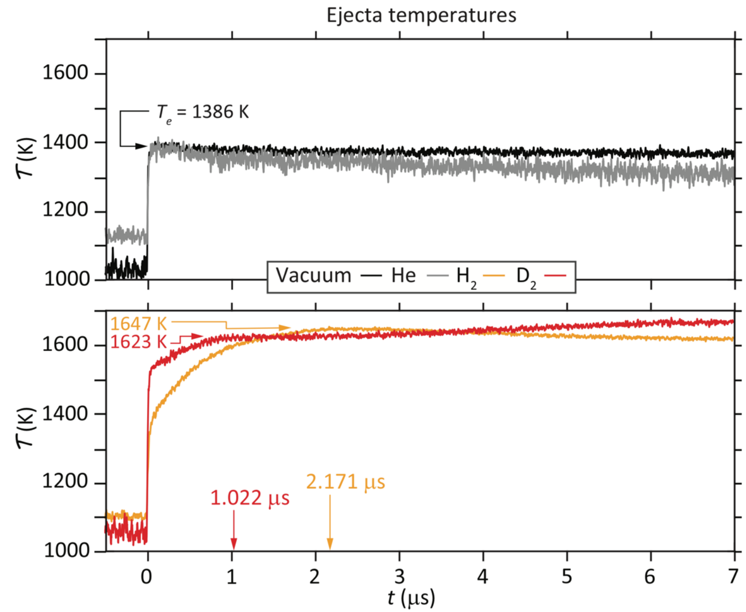
<u>Theories</u>

- Larger diameter, slower ejecta are revealed as leading ejecta breakup



Buttler, Shulze, Charonko, Cooley…, Physica D, 415, 132787, 2021

Particle Temperatures

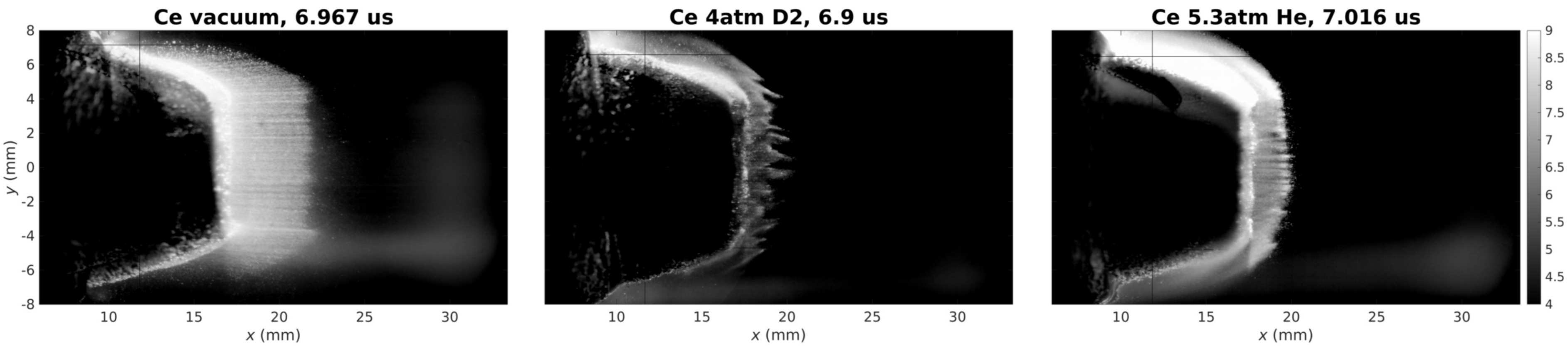


Buttler, Hartsfield, Schulze…, LA-UR-22-21121

- Temperature for nonreacting cases decays with time from initial temperature of 1386 K
- Temperature reaches about 1600 K in reacting shots
  - endothermic process?
  - hydride melting?
  - Something else?

PIV image

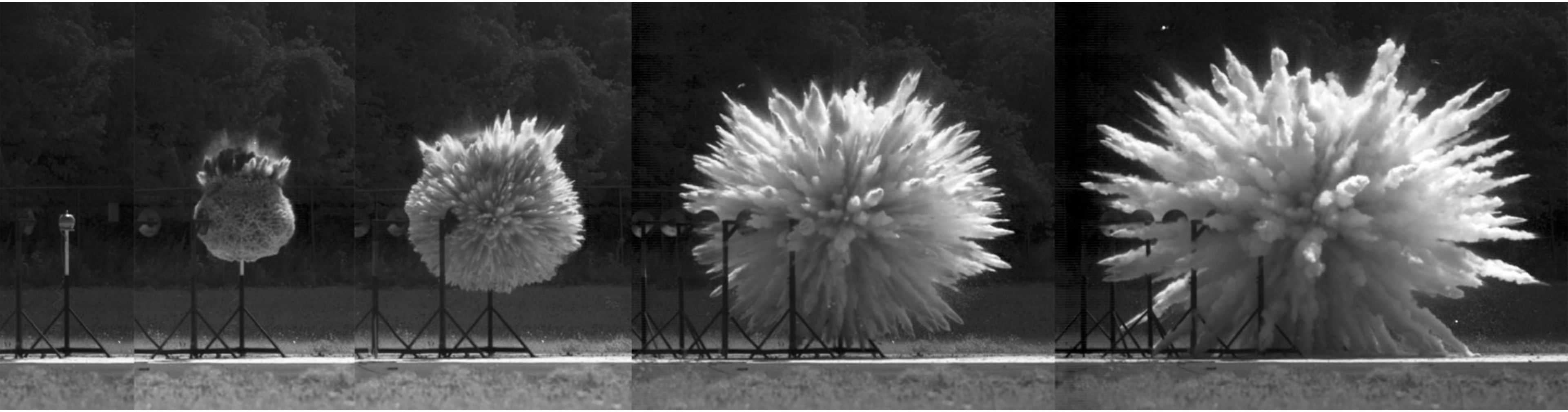
- Nonreacting shots create sheet with near constant velocity at edge
  - Jets coincide with perturbation wavelength
  - Consistent particle size
- Reacting shot reveals much longer perturbation period
  - Wider distribution of particle sizes



LA-UR-19-32716

Explosive dispersal of particles

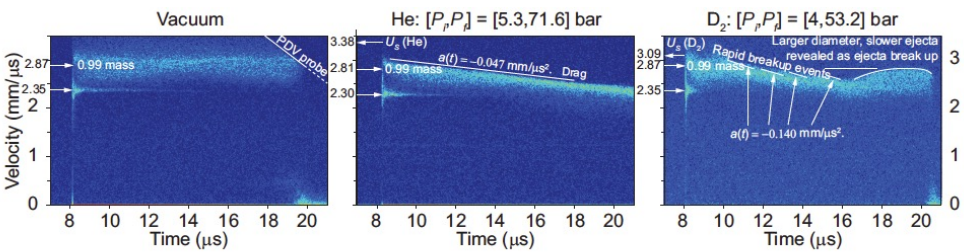
- Packed bed of sand particles inside glass container
- Explosive at center
- Formation of jets
- May occur from polydisperse ejecta particles
- Still not clear how this process works

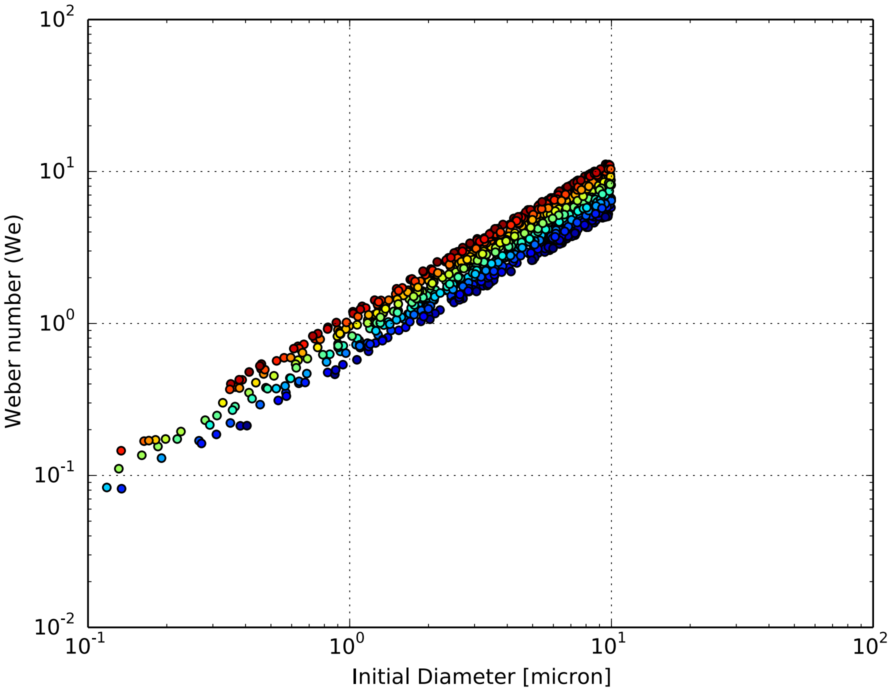


D.L. Frost, Y. Gregoire, O. Petel, S. Goroshin, F. Zhang, Phys. Fluids, 24 091109, 2012.

Hydrodynamic stability

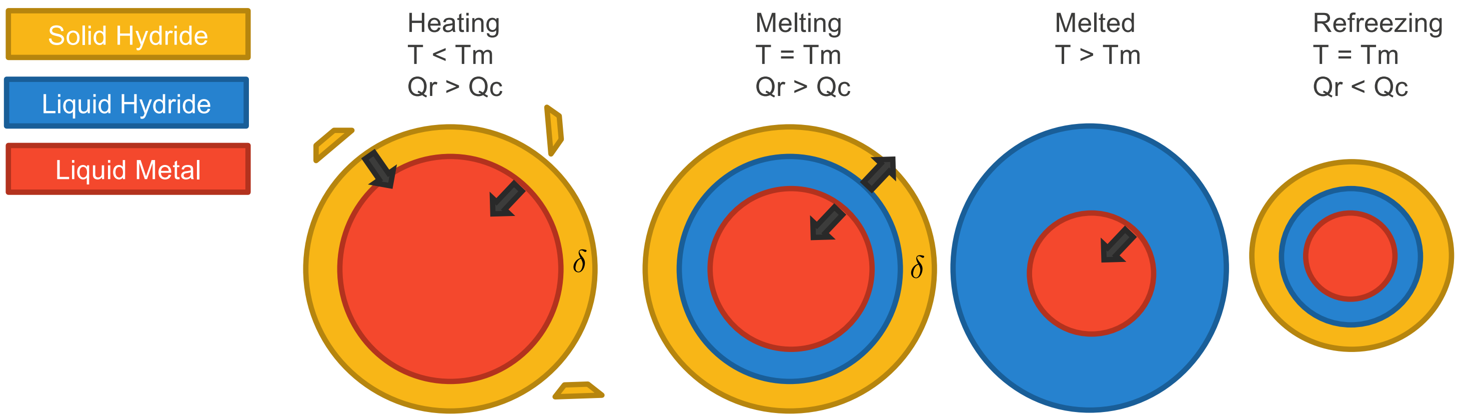
- Assume surface tension of liquid hydride is roughly equal to liquid cerium
- Weber number is mostly below critical value (11) for wide range of sizes
  - Particles are likely hydrodynamically stable
- Polydispersity originate at formation?
  - Why not observed in nonreacting cases?





Liquid ejecta model – current working hypothesis

- Liquid ejecta model – current working hypothesis



Heating

- Consuming metal
- Forming solid hydride
- Reaction limited by diffusion
  through hydride layer
- Hydride flakes off and leaves
Thin layer of hydride

Melting

- Liquid heals solid and makes solid ductile
- Shell remains intact
- Finite amount of time to melt O(micro-sec)

Thickness δ

Melted

- Energy from reaction is used for conversion and also phase change
- Diffusion rate through liquid is 10x faster than solid
- Temperature can rise and fall

Refreezing

- Reaction surface Becomes small
- Heat release is Less than loss
- Hydride freezes
- Still ductile
- Doesn't shed particles until totally frozen

<u>Estimated Inputs:</u>

Heat of fusion
Melt condition
Thermal conductivity
Specific heat
Shell thickness*

* Calculate using hoop stress

Hydride Shell Melting Model

1) Compute the total energy required to melt the shell when $T_p = T_{melt}$

$$H_f = h_f^o m_{shell} = \frac{4}{3}\pi h_f^o \rho_{shell}(b^3 - a^3)$$

2) All Δe_p goes toward melt and not toward ΔT_p

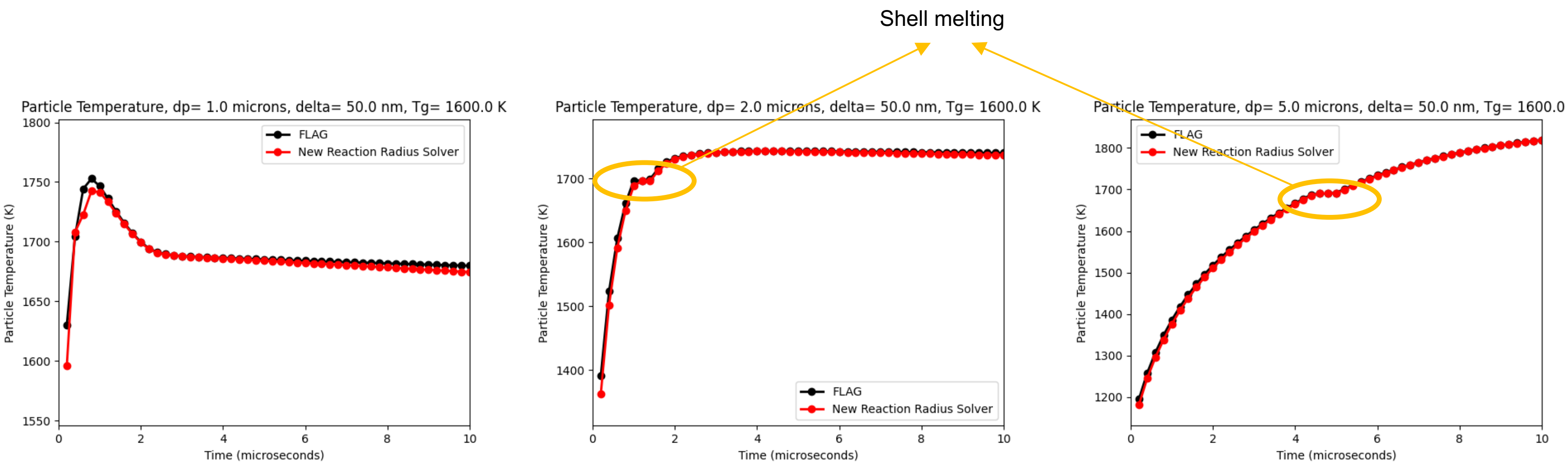
3) Dynamically update the heat required to melt the shell due to the particle's energy change as

$$H_f^{n+1} = H_f^n + \left(\frac{dH_f}{dt}\right)^{n\ (or\ n+1)} \Delta t - m_p \Delta e_p$$

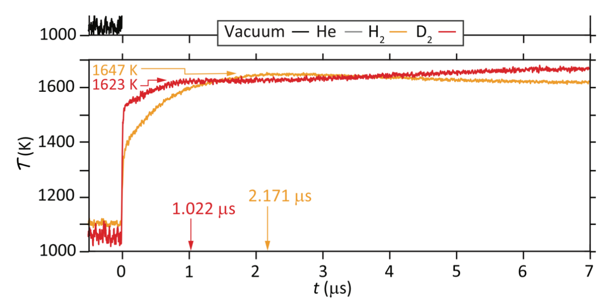
where $\frac{dH_f}{dt} \approx 4\pi h_f^o \rho_{shell}(b^2 - a^2) \cdot \frac{da}{dt}$ accounts for the changing shell size on the required heat

4) Continue process until $H_f^{n+1} = 0$ and the shell is melted

Melt Model Results: Tg = 1600 K, shell thickness = 50nm



- Tested melt model with single particle and gas temperature fixed
- Estimated latent heat of fusion from Lithium hydride
- Thermal conductivity and specific heat estimated from cerium
- Causes particle temperature to pause for less than a microsecond
- Need reliable properties
- Cloud behavior may be more complex



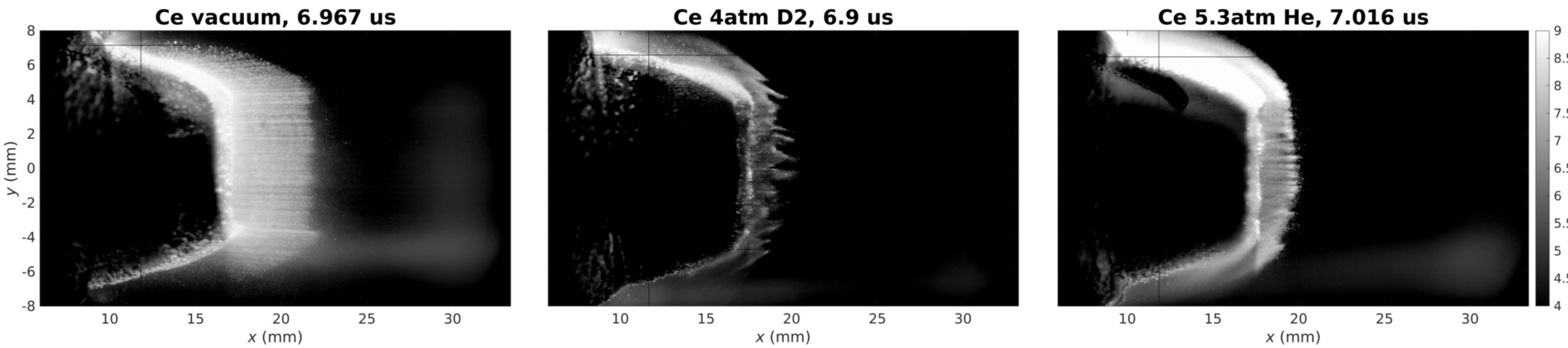
Liquid ejecta conclusions

- Unstable velocity is believed to be from polydisperse ejecta
  - Large particles have more momentum than small
  - Jets form as seen in other explosive dispersal experiments
  - Only occurs in reacting cases

- Model provides a way to create smaller particles from reaction
- Also has mechanism to capture temperature plateau
- Still need material properties for validation

Summary

- Understanding reacting ejecta processes is ongoing
- Model development will take several years
- Contingent upon reliable material properties
- More canonical experiments would be advantageous as well



Impact of two-way momentum coupling

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