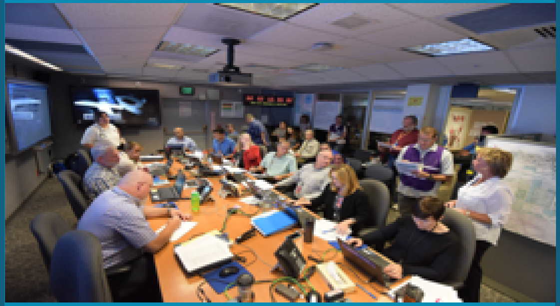
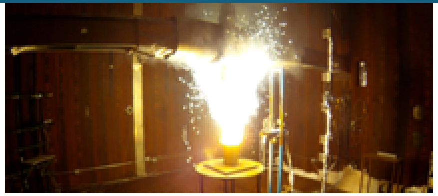
SAND2022-1835C

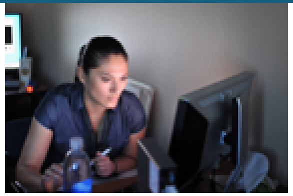
February 25, 2022

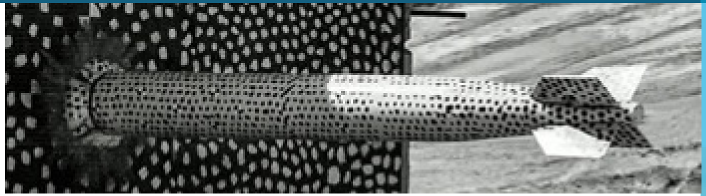
Sandia National Laboratories

Entrainment from Contaminant Accident Scenarios Involving Fire









For the EFCOG Meeting, February 25, 2022

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Dr. Flint Pierce, and Ethan Zepper

Fire Science and Technology Department

Fire Science & Technology

U.S. Department of Energy ENERGY NNSA

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Outline

Last year at the EFCOG meeting Year 1 NSRD was presented focused on emissions from contaminated liquid fuels

- Will review prior work summary
- Will briefly review previously presented literature review results

This final year of the project focus turned to contaminated solids and the exhibition of three campaigns:

- Contaminated PMMA fire simulations compared to historical (BF-1995) release data (Flint Pierce)
- Revisiting the MS-1973 liquid pool fire scenario including conjugate transport (Alex Brown)
- Predicting the Hubbard et al. (2022) data on contaminant release from burning cellulose (Ethan Zepper)

Year 1 Summary

A review of experiments on contaminant entrainment from fires has been completed for solid and liquid combustibles

Evaporation Induced Entrainment is described quantitatively based on MD simulations

- The model is adaptable to permit predictive evolution using a CFD code

Solid emissions are potentially sensitive to:

- The propensity for the char to accumulate around the particles
- Transit of particles into liquid polymers

We are pursuing mechanistic models for predicting airborne release

- Using SIERRA/Fuego as a platform for model development
- Data are lacking to sufficiently characterize the test behaviors

Historical Study Review-solid sources

Limited (fewer than with liquids), highly variable, historical studies exist on entrainment of contaminants from solids during a fire:

Study	Solids	Contaminants	Sizes Pre-test
MS-1973; BNWL-1730	4-5 kg of mixed Cardboard, paper, plastic, rubber, rags, oil, tape	UO_2, Uranium nitrate liquid, air dried uranium nitrate	0.2-30 mm
MS-1973; BNWL-1732	Sandy soil, vegetation, stainless steel,	UO_2, Uranium nitrate liquid, air dried uranium nitrate	0.2-30 mm
Halverson et al., 1987	Polychloroprene, polystyrene, PMMA, cellulose	UO_2, uranyl nitrate hexahydrate (UNH) solution, and UNH salt	MMD of 1 micron, aqueous, and crystal
Bhanti et al., 1988	Styrene divinyl benzene co-polymer	Th in the resin	N/A
Buijs et al. (1988-1992), Pickering et al. (1987-1989)	Primarily PMMA with other materials	Ce, Eu oxides in large-scale tests U, Pu ,and Am oxides in small -scale tests	MMD of 10.5 micron with sieve below 40
Fernandez and Burghoffer (1995)	PMMA	CeO_2	1.75-7.5 micron MMD
Mendoza et al. (2020)	Cheesecloth, plastic bag, PMMA, rubber, cellulose	CeO_2	
Hubbard et al., 2021	Cellulose	Lu, Yb, U, nitrates and mesoparticles of salts	N/A

Comparisons exhibited in this work ← (Fernandez and Burghoffer (1995); Hubbard et al., 2021)

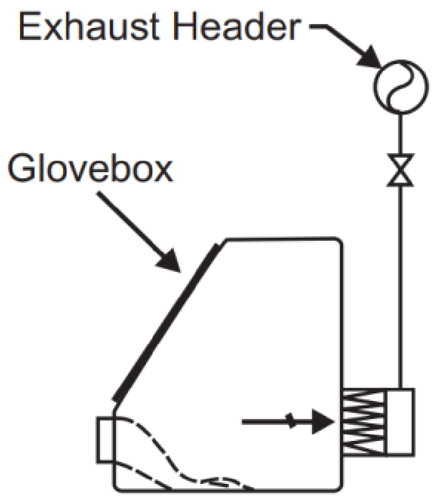
Modeling Contaminant Release from Heated PMMA

Flint Pierce

Modeling Contaminant Release from Heated PMMA

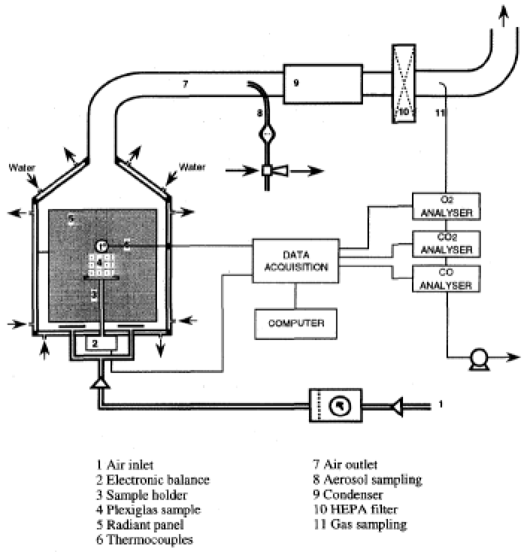
What was done

- Model for contaminant particle release from heated/ melting/pyrolyzing PMMA as present in gloveboxes and other DOE hardware
- Developed using coupled CFD/Thermal/Radiation transport (Sierra modules Fuego_Aria , Nalu)



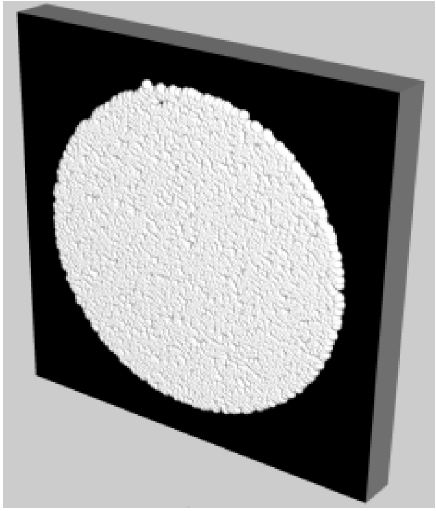
Fernandez and Burghoffer (FB1995) investigated radiatively heated PMMA slabs in chamber with low velocity upward vertical airflow (9-13 m^3/hr)

- 10cm x 10cm PMMA samples
- Heated by front side radiant panel (450K/550K)
- 0.5m high, 0.17m^2 horizontal cross- section chamber
- Airborne Release Fractions (ARFs) determined from particle flow through top of chamber



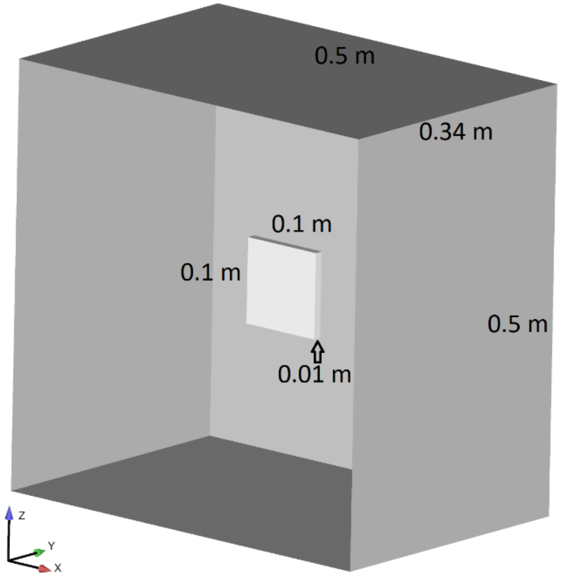
Initial contaminant (CeO_2) particle deposit on PMMA surface

- 1.7µm or 7.5 µm contaminant particle diameter
- 8cm circular deposit



FB1995 scenario modeled in SNL Sierra Thermal/Fluids code suite

- Fuego: Eulerian CFD, Lagrangian contaminant particles
- Aria: heat transport, contaminant diffusion in PMMA
- Nalu: thermal radiation transport

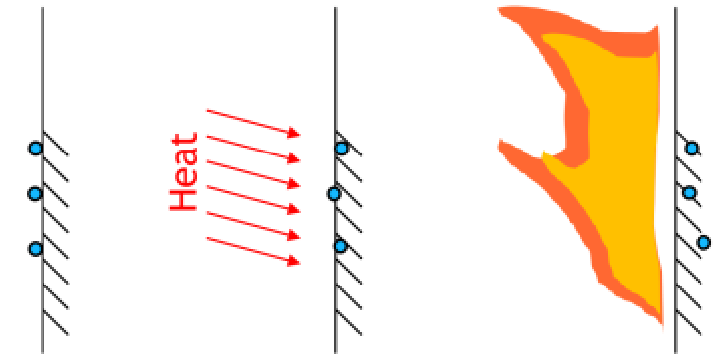


1. Fernandez, Y. and Burghoffer, P., 1995. Radioactive aerosols emission in fires. Aerosol science and technology, 23(2), pp.231-238

Modeling Contaminant Release from Heated PMMA

Why was it done

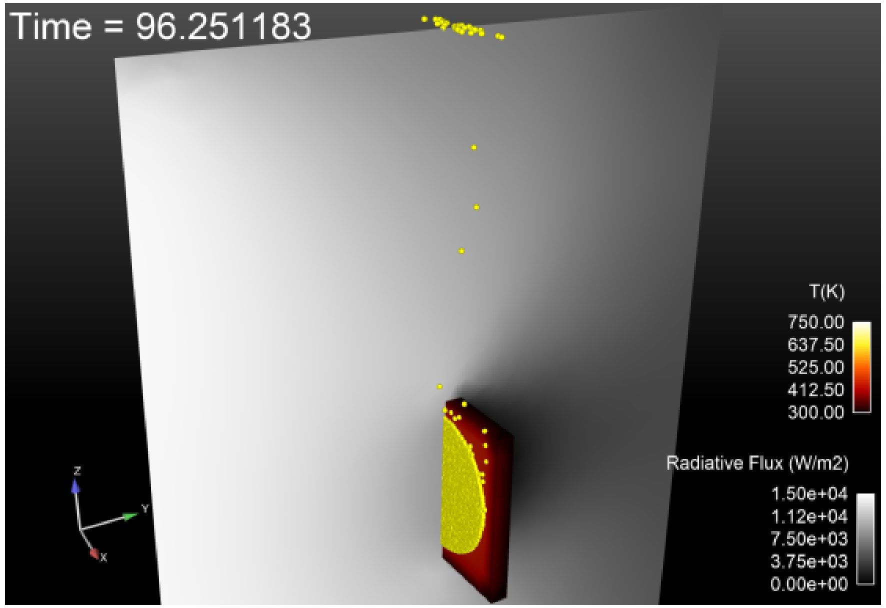
No previous computational model available to simulate entire process



- FB1995 study: ARF relative to heating rate, air flow velocity, contaminant particle size
- Most detailed study of ARF dependence in this type scenario
- *Simulating this case helps reveal underlying principles governing the process and provides means to compare model predictions to available experimental data*

Need to understand ARF dependence on:

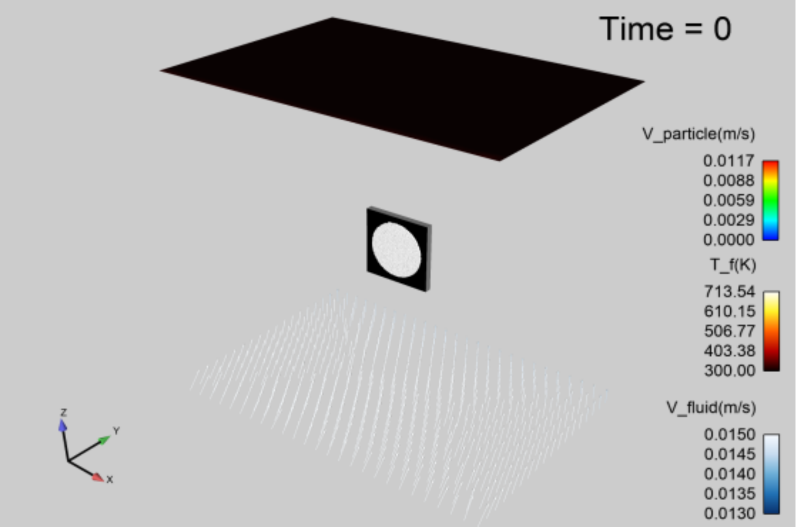
- contaminant release
  - no rigorous model available/little validation data
  - our model – release increases (linearly) with:
    - local temperature
    - contaminant concentration at PMMA surface
- contaminant diffusion in heated/melting PMMA
  - No rigorous model for *large particle diffusion* in PMMA
  - our model
    - As T increases, diffusion increases (linearly)
- Can we reproduce BF1995 results?



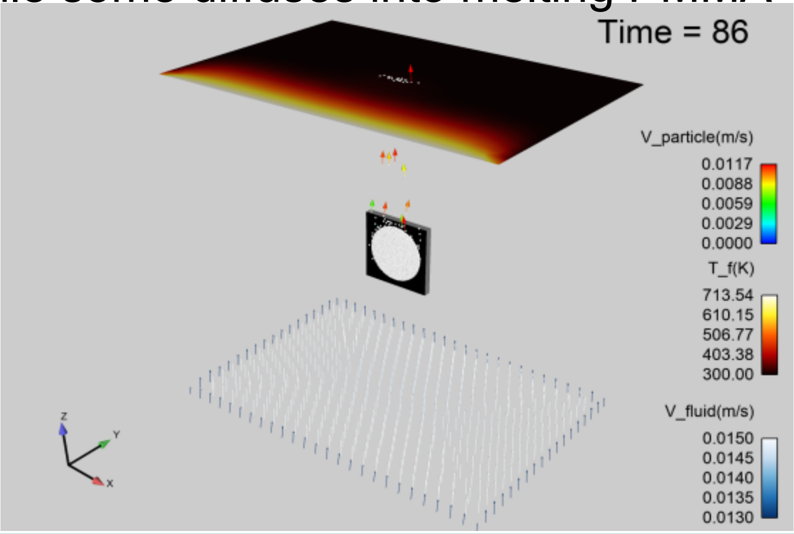
Modeling Contaminant Release from Heated PMMA

Visualizing simulation results over 1 hr of simulation time

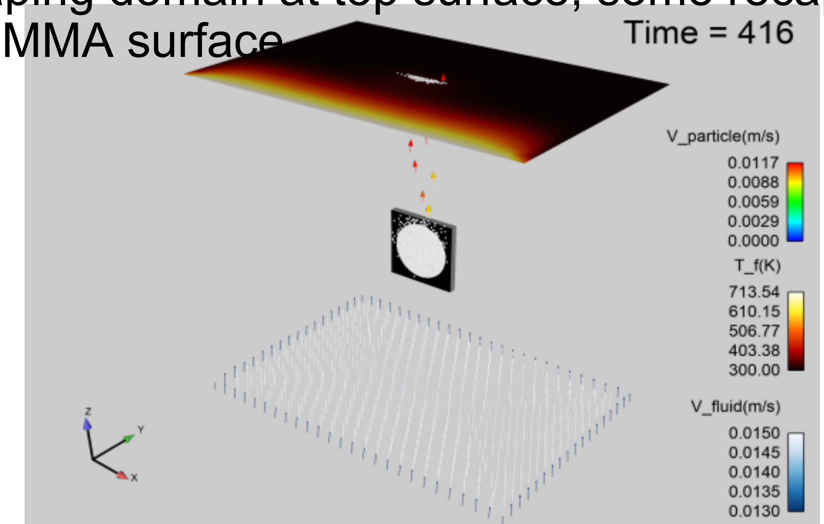
- Start: contaminant embedded on PMMA



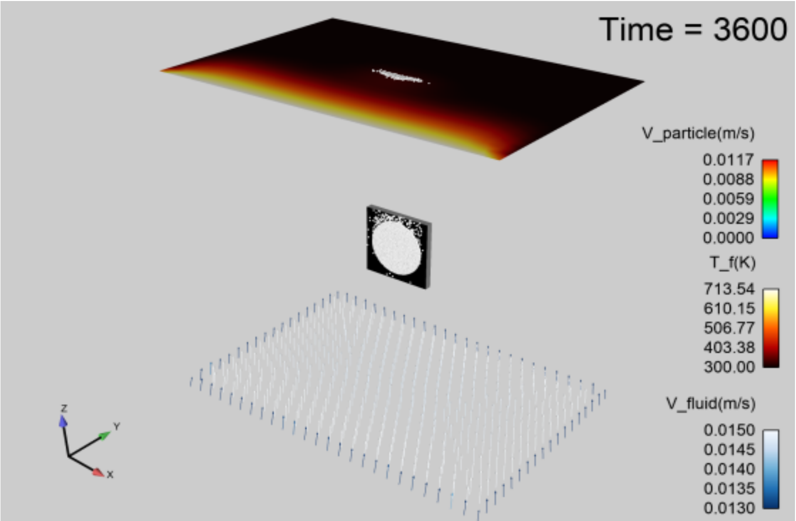
- As PMMA heated, contaminant starts being released, lofted while some diffuses into melting PMMA



- As heating continues, more contaminant lofted, escaping domain at top surface, some recaptured on PMMA surface



- After 1 hour, all releasable contaminant has left domain (ARF)



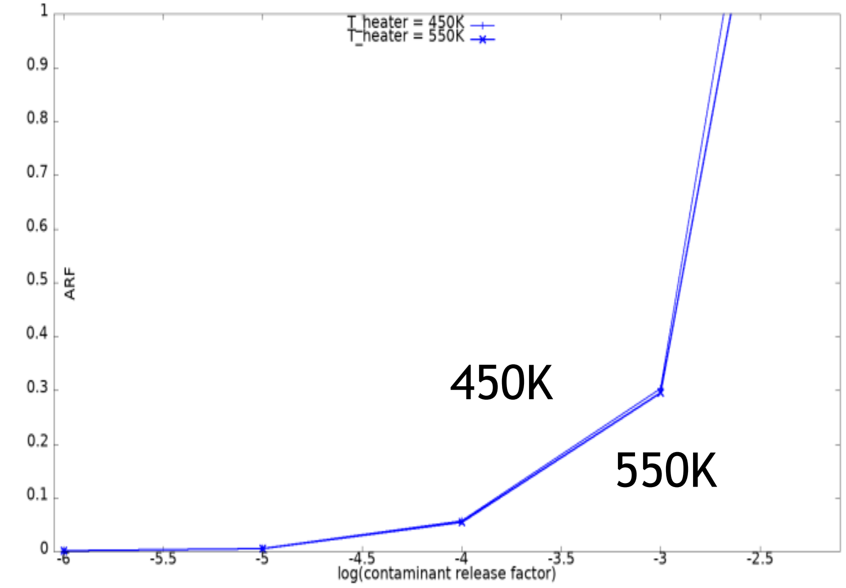
Modeling Contaminant Release from Heated PMMA

What was learned: ARF

- Explored effects
  - contaminant release (linear with T) and
  - contaminant diffusion (linear with T, concentration) in PMMA
- All 8 parameter sets in FB1995
  - $d_{particle}$, T_{heater}, v_{flow}
- Comparison to FB1995
  - FB 1995: ARF = 0.005-0.05
  - This study: ARF extends over that range, depends on model choice of contaminant release and diffusion factors

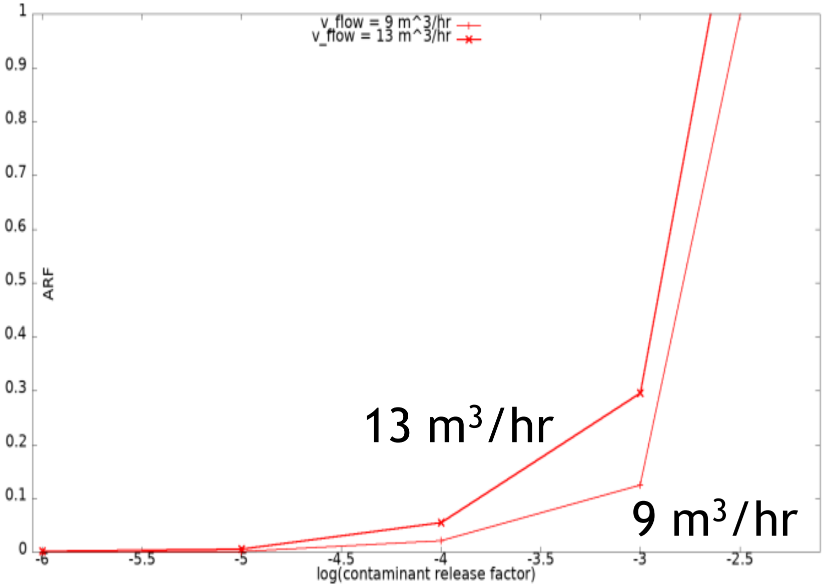
T_{heater} = 450-550K

- *little impact* on ARF
- Contaminant release from PMMA *and* diffusion into PMMA increase linearly with T, canceling each other's effect on ARF
- higher release rate factor → larger ARF (expected)



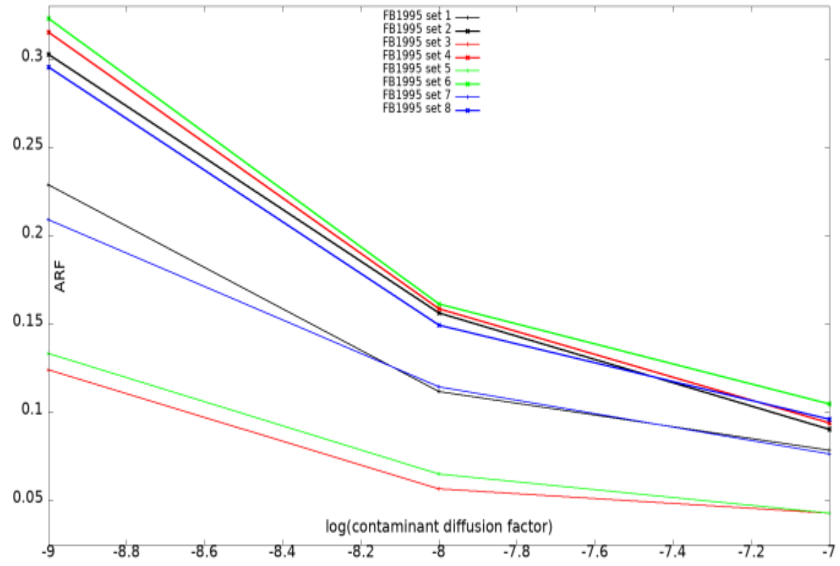
v_{flow}= 9-13 m^3/hr

- higher v_{flow} → larger ARF (expected)
- Easier to pull released contaminant away
- higher release rate factor → larger ARF



contaminant diffusion factor = 10^{-9}-10^{-7}

- ARF decreases with contaminant diffusion factor
- more contaminant diffusion into PMMA = less available for release



Actinide Entrainment Conjugate Analysis

Alex Brown

Actinide Entrainment Conjugate Analysis

What was done

Conjugate analysis means including the heat transport between gas and solid via a coupling method

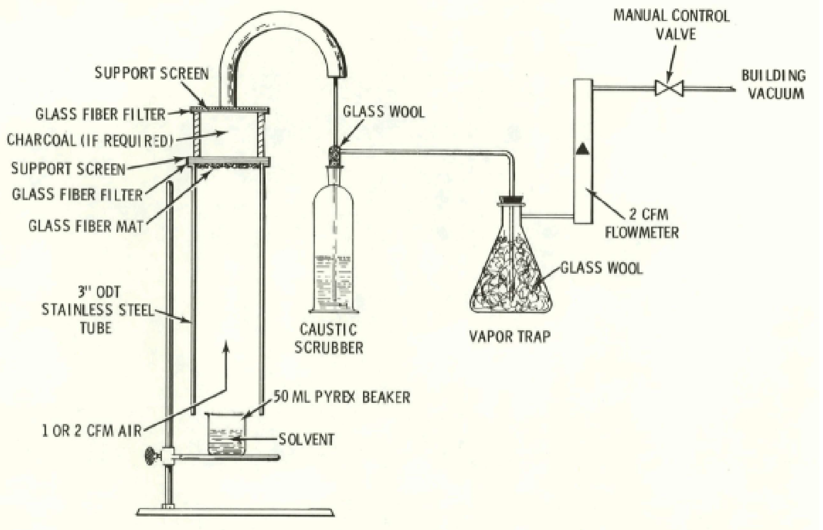
Done (enabled) using SIERRA-Fuego/Aria coupling
Omitted in prior studies

Simulations were made to compare to one of the few datasets key to ARF prescription in DOE-3010

Historical MS-1973 data involved a release from a TBP/Kerosene mixture with contaminant

Results include burn time and Airborne Release Fraction (ARF)

- **Average fuel regression rate (0.22 – 0.39 mm/min)**
- **Average contaminant release rate (4.6e-10 – 7.1e-10 kg/s)**



The simulation included some new parameter assessments compared with prior work:

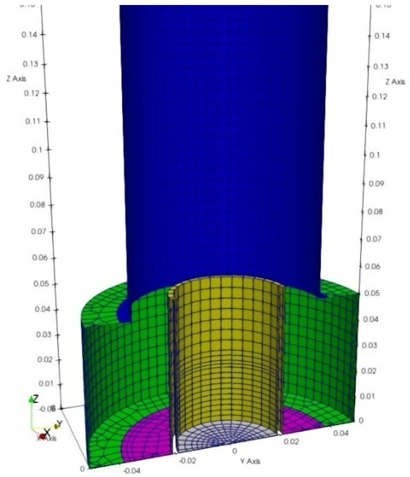
- Mesh refinement
- Beaker particle stick model variations
- Aria heat transport properties
- Particle size distribution and velocity

Simulations also take advantage of some code development that permit higher fidelity modeling

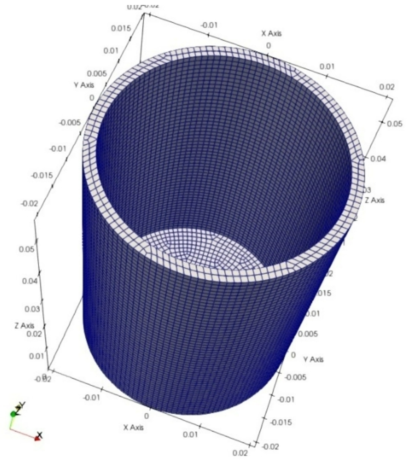
- Comparisons to burn rate and ARF

Jointly solved via Segregated coupling method

The fluid mesh



The (new) solid mesh



Actinide Entrainment Conjugate Analysis

Why was it done

Prior work identified the pool height as a key parameter of sensitivity to the ARF

- Questionable applicability of handbook data to all possible conditions

Prior work lacked model fidelity

- Burn rate was constant at rate estimated from test data
- Container walls were assumed isothermal

New and improved physics allows for a better interrogation of the problem and characterization of the accuracy of the model

The ability to use surrogates for more hazardous compounds like Pu or PuO enables testing and qualification of models

- If the models are sufficiently accurate and reliable, they can replace the need for dangerous tests

The simulation matrix helps understand the effect of model assumptions on ARF and burn rate

ID	Variation from Baseline
1	None
2-fine	Fine mesh (2x in each dimension)
3-stick	Particles all rebound instead of stick to the beaker walls
4-b-ht	Includes increased beaker heat transport parameters
5-ps+	Particle size distribution assumed larger than baseline
6-ps-	Particle size distribution assumed smaller than baseline
7-pv-	Particle velocity assumed smaller than baseline
8-r1	Pool rate model assumption 1
9-r2	Pool rate model assumption 2
10-h1	Pool height assumption 1
11-h2	Pool height assumption 2
12-h3	Pool height assumption 3
13-h4	Pool height assumption 4
14-c1	Contaminant surrogate used
15-op	Opaque beaker
16-b-ht-op	Beaker heat transport parameters and opaque beaker

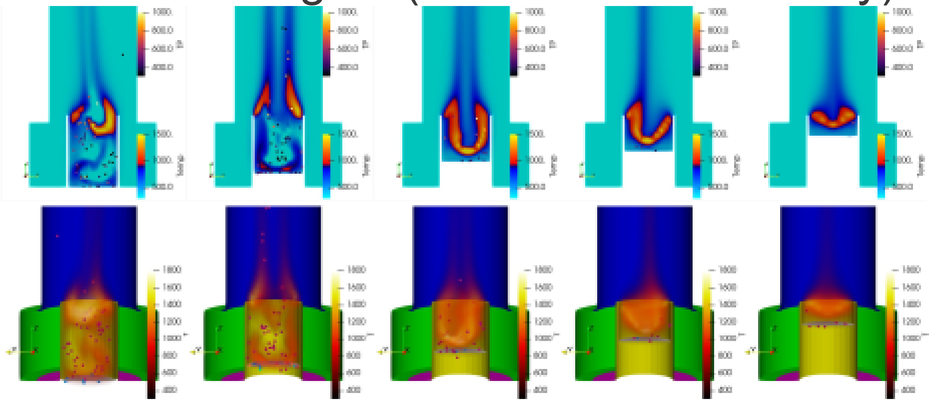
Actinide Entrainment Conjugate Analysis

Novelty of what was done

First to our knowledge simulations done with a predictive pool regression rate and linking that to the particle entrainment

- The model assumes that entrainment is proportional to the burn rate, and the magnitude is defined by correlations of Kataoka and Ishii (1985)
- Size distribution of droplets came from measured distributions (out of HDBK 3010)

Test matrix increased the fidelity of predictions of the liquid at various heights (still need more fidelity)



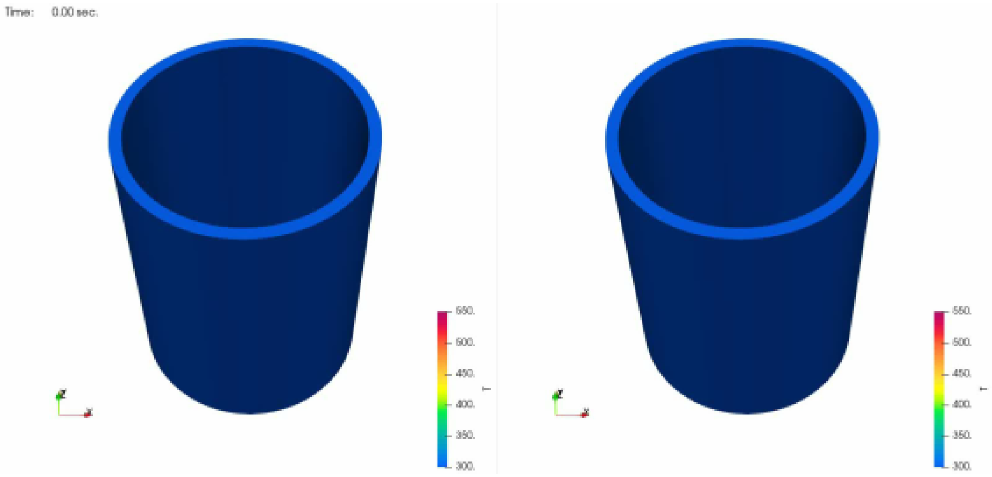
Temperature predictions (K) from the simulation of five liquid heights (low to high left to right)

First to our knowledge conjugate simulation to help understand the effect of the container temperature

- Previous models made an isothermal assumption
- Intended to explore the effect of increased detail in the simulation

Video: Tested effect of predicted beaker temperature

Radiation was either absorbed (right) or past through (left), had major effect on predictions



Actinide Entrainment Conjugate Analysis

What was learned

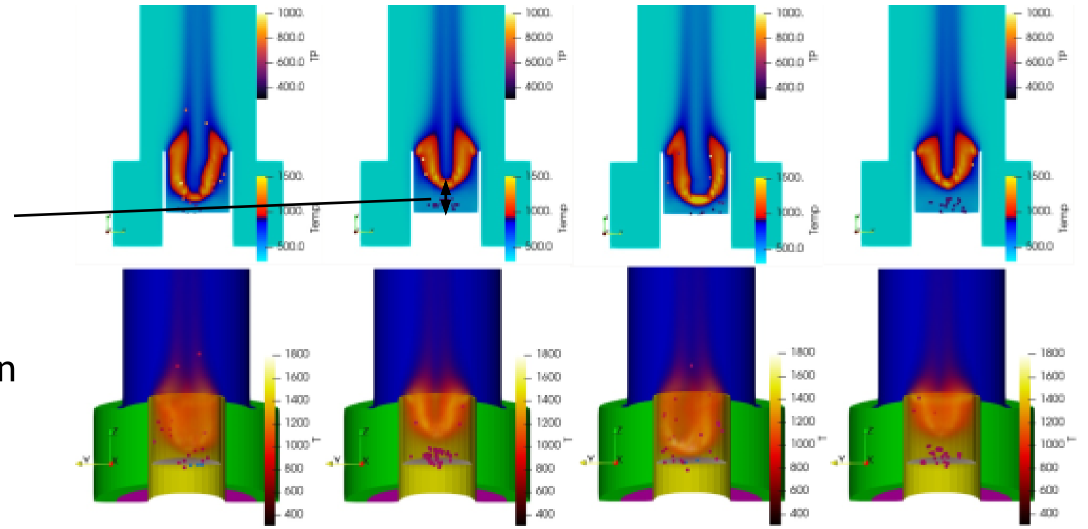
Particles evaporate rapidly, becoming contaminant only

Fuel height effect is <u>significant</u>

Major effect of radiation assumption (transparent or not) container on the magnitude of the release and burn rate

Can articulate code improvements needed to improve simulations

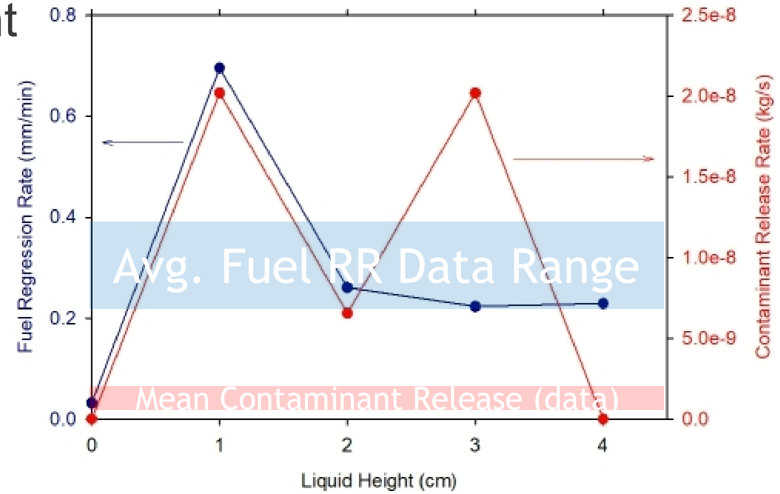
This height appears to make a big difference in pool burn rate and release fraction



Temperature predictions (K) different beaker absorptivity assumptions

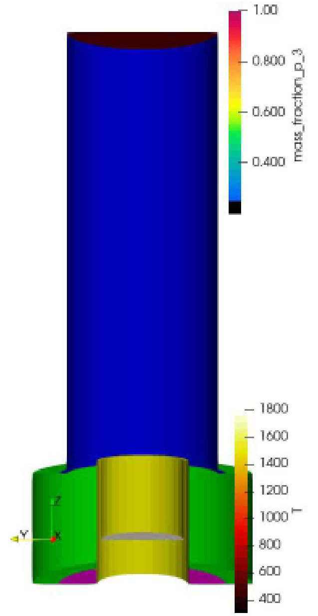
Release and burn rate *very* non-monotonic with fuel height

Transparent beaker results



Suggested topics for future code improvements:

- Receding fuel level via a model for dynamic meshing or multiphase behavior will enable ARF comparisons with data
- Soot/contaminant interactions (affects RF, and possibly ARF)
- Multicomponent fuel sources



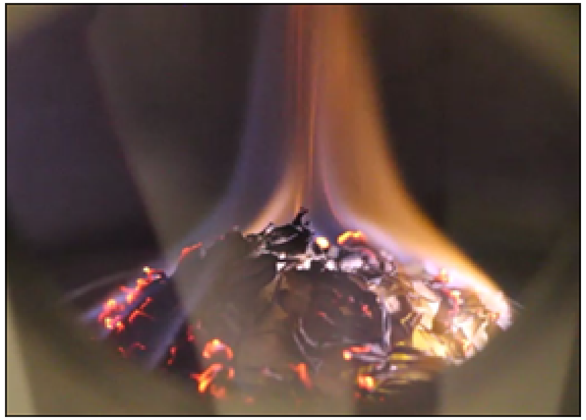
Modeling Contaminant Release from Burning Cellulose

Ethan Zepper

Modeling Contaminant Release from Burning Cellulose

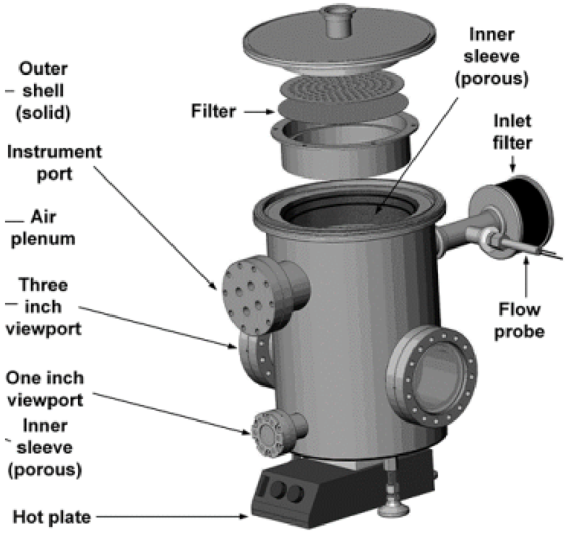
What was done

A novel particle release mechanism for use in CFD simulations was formulated based on predicted progress of the decomposition of organic materials



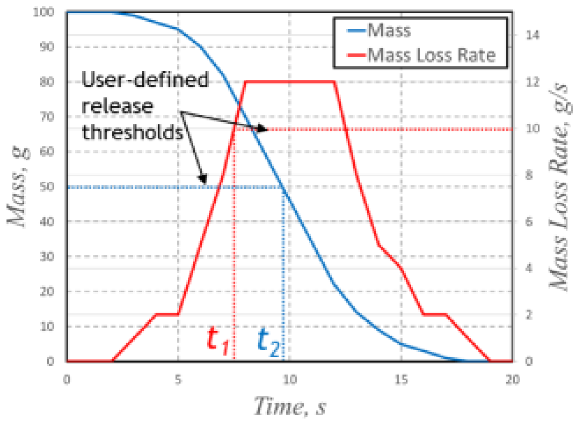
Recent experimental efforts by Hubbard et al.[1] studied the release of contaminant from reacting solid substrates

- 5g of cellulose burned in custom chamber
- Cellulose cut into strips to aid combustion
- 1 wt% contaminant load
  - UO_2, Lu_2O_3, and Yb_2O_3
- Airborne Release Fractions (ARFs) determined from the filter collection system



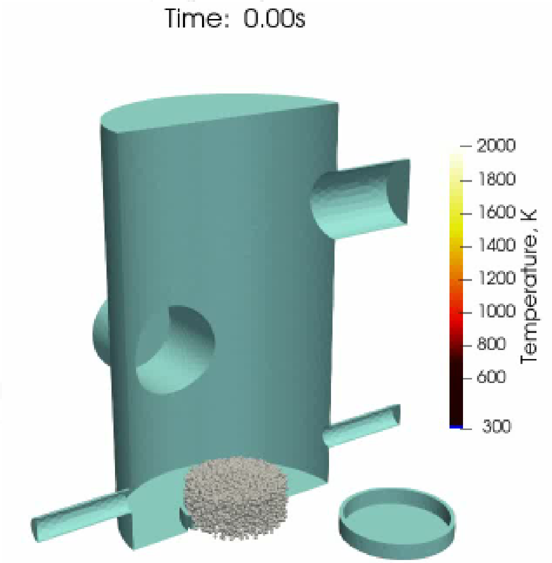
Two user-defined release criteria are based on material (cellulose) decomposition

- Species Mass Loss (blue)
- Mass Loss Rate (red)



Experimental setup modeled using Sierra/Thermal Fluids

- Cellulose strips modeled using Lagrangian particles
  - Surface-area-to-volume ratio matched
- Novel release mechanism applied
- Predicted ARF determined for comparison



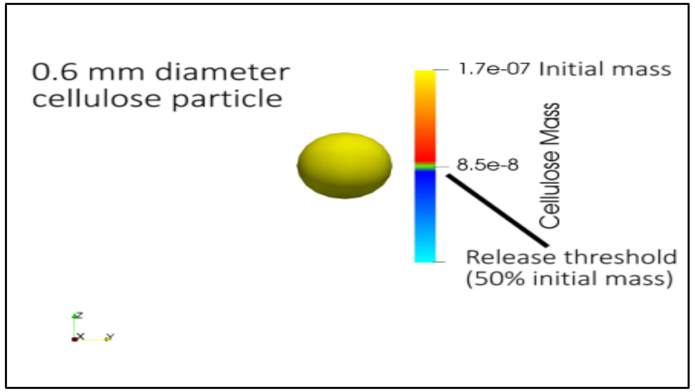
1. J.A. Hubbard, T.J. Boyle, E.T. Zepper, A. Brown, T. Settecerri, J.L. Santarpia, N. Bell, J.A. Zigmond, S. S. Storch, B. J. Maes, N. D. Zayas, D. K. Wimann, M. Ringgold, F. Guerrero, X.J. Robinson, G. A. Lucero, and L. J. Lemieux, "Determination of Airborne Release Fractions from Solid Surrogate Nuclear Waste Fires," Nuclear Technology, vol. 208, no. 1, pp. 137-153, 2 January 2022

Modeling Contaminant Release from Burning Cellulose

Why was it done

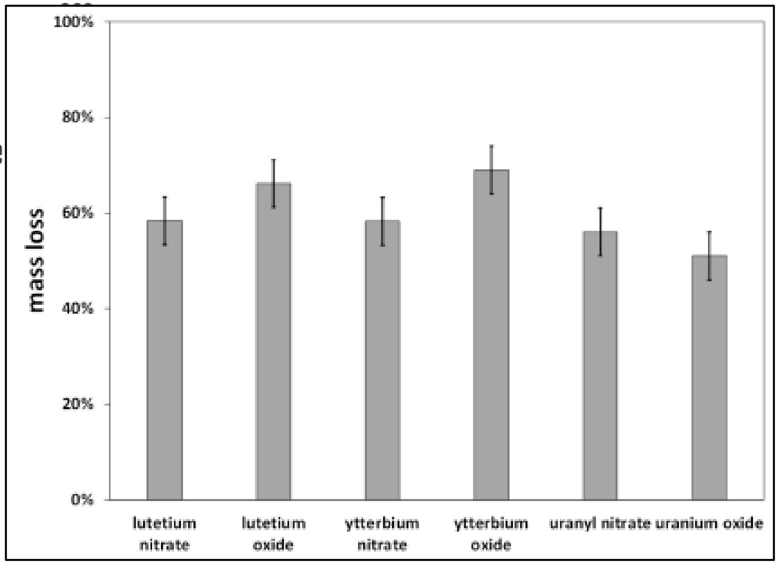
Initial model verification efforts demonstrated the function of the new release mechanisms

Application to a physical scenario was therefore desired



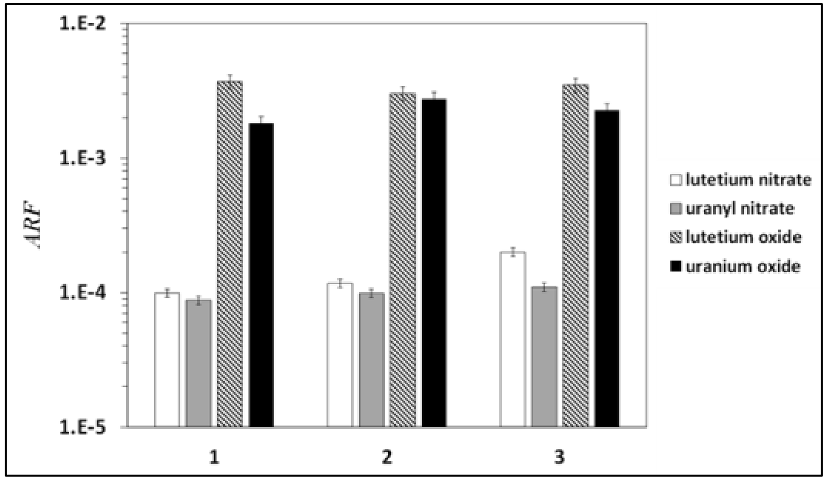
Detailed measurements of the cellulose mass loss and burn duration were available

- Mass loss exceeded 55%
- Flaming burn duration typically exceeded 2 minutes
- Smoldering combustion also noted, but this phenomenon was not modeled



Cellulose combustion experiments from the Hubbard et al. provided high-quality validation data and thus seemed a natural fit for model validation

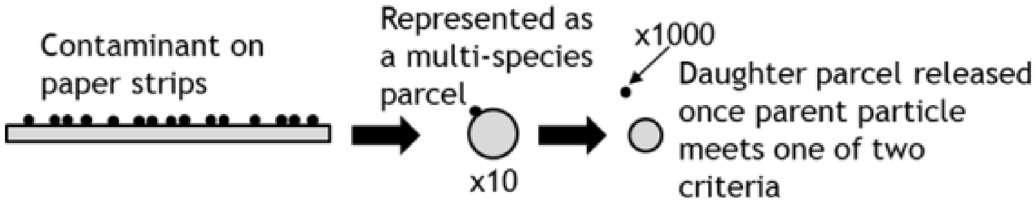
ARF measurements from the Hubbard et al. experimental effort provided a clear comparison opportunity for evaluation of the new release model



Modeling Contaminant Release from Burning Cellulose

Novelty of what was done

A generalized engineering model for contaminant release or resuspension did not currently exist, to the authors' knowledge, prior to the development of this two-factor release mechanism in Sierra/Fuego



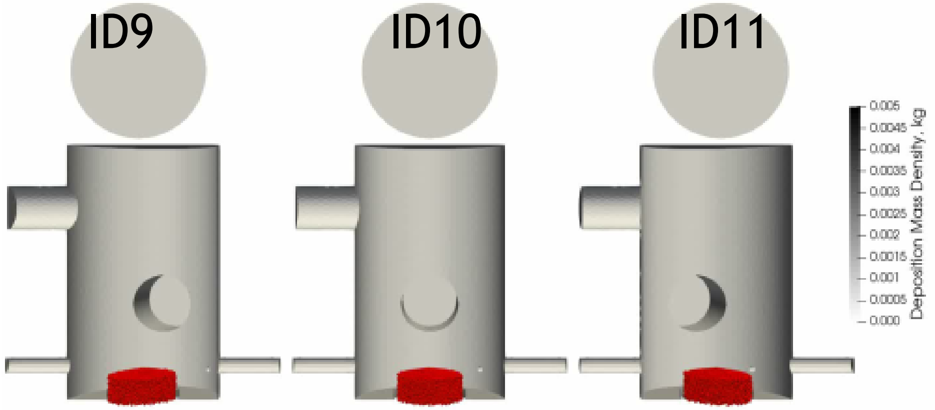
Parametric Analysis Scenario Matrix and Results

ID	Scenario	Extinction Time, s	% Mass Loss	ARF
1	Baseline (UO2)	56.1	59.5%	0.891
2	Surrogate (PuO_2)	55.9	59.4%	0.889
3	Surrogate (Lu_2O_3)	56.0	59.5%	0.888
4	Surrogate (Yb_2O_3)	56.3	59.5%	0.889
5	Species Mass Loss (90%)	55.9	59.4%	0.891
6	Species Mass Loss (95%)	56.8	59.5%	0.888
7	Mass Loss Rate (1%/ms)	56.3	59.4%	0.890
8	Mass Loss Rate (10%/ms)	56.2	59.4%	0.888
9	Species Mass Loss (99.99%)	56.1	59.4	0.877
10	Species Mass Loss – disabled	56.3	58.5	9.31E-3
11	Mass Loss Rate (1%/ms; Species Mass Loss disabled)	56.1	58.7	0.211
12	Mass Loss Rate (10%/ms; Species Mass Loss disabled)	56.2	58.5	8.55E-4

To understand model sensitivities, a parametric study was performed

- Sensitivities of the two entrainment threshold parameters (species mass loss and mass loss rate) were explored
- 4 contaminants with varying material properties
  - Uranium dioxide (UO_2)
  - Plutonium dioxide (PuO_2)
  - Lutetium oxide (Lu_2O_3)
  - Ytterbium oxide (Yb_2O_3)

For the parameters studied, disabling the species mass loss criteria (i.e. only employing the mass loss rate) criteria proved to be the most sensitive parameter.

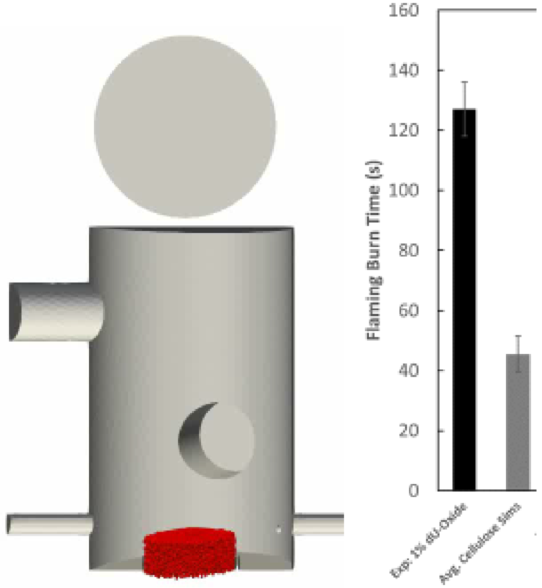


Modeling Contaminant Release from Burning Cellulose

What was learned

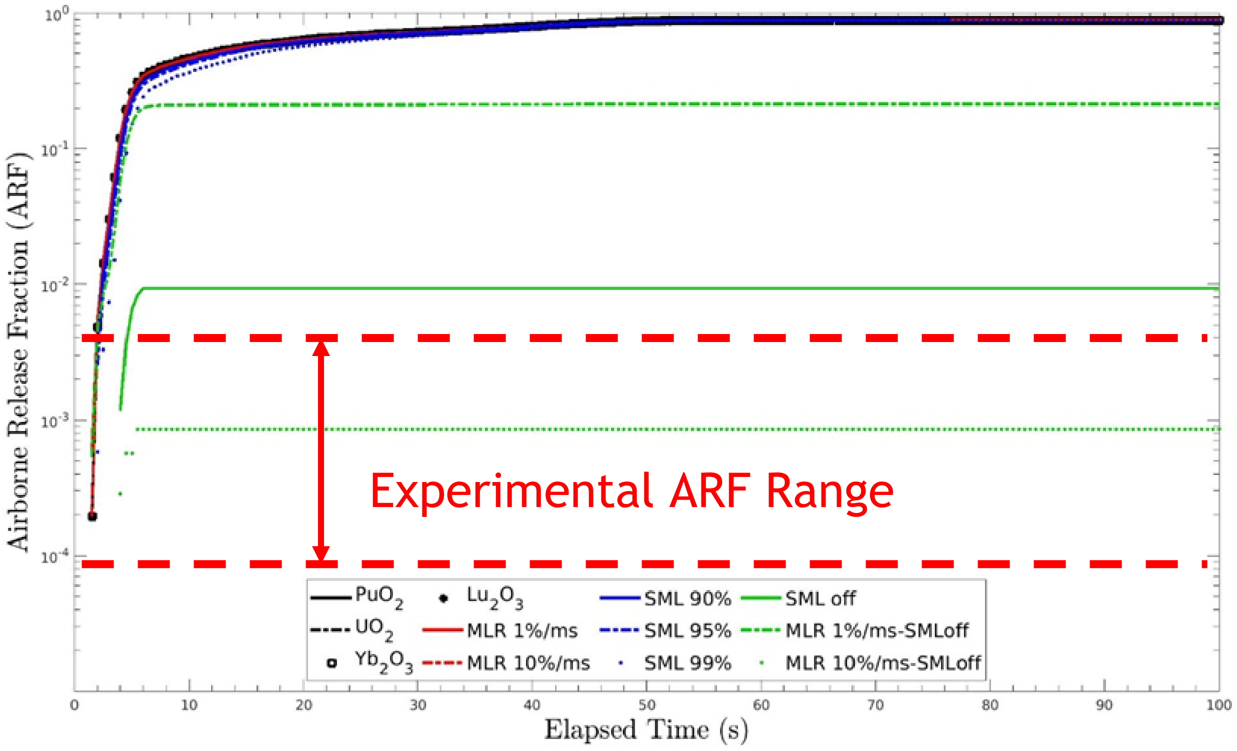
Lagrangian particle representation of cellulose strip material bed did not accurately represented the experimentally-observed fire

- Inter-particle thermal conductivity (via node mapping) would enable models with more accurate physical representations of the cellulose strips



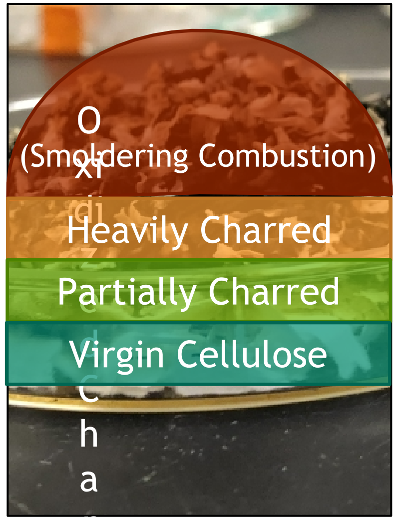
One permutation fell within the experimental ARF ranges by disabling the species mass loss condition

- Likely due to statistical representation of particles (parceling) and the parameter range chosen



Char formation, oxidation, and combustion was clearly important in the experimental effort, but was intentionally ignored in the model to simplify vetting of the new release mechanism

- Future efforts should consider the effects of



Notional Char Layering

While the new release model was found to result in release, some modifications to the model are desirable to achieve quantitative accuracy.

Summary

Limited data exist for contaminant release

New model developments are enabling predictions of increasingly relevant physics

- Some model tuning still needed for the more complex physical behaviors
- ARF is modeled to be functional with the behavior of the fuel
- Simulations are able to bracket experimental data to varying degrees

Main Findings:

- Contaminants migrate into PMMA plastics as they burn, deploying at present simple linear models for release and diffusion
- The MS-1973 dataset is complex in unexpected ways (beaker absorptivity, fuel height), and probably not reliable to use as bounding for ARF estimates as per HDBK 3010
- Cellulose burning using array of Lagrangian particles is challenging, matching burn rate is difficult
    - Release temperature model assumption abandoned, overestimates release
    - Release due to high heating rate can explain the ARF data, suggests early release of contaminant

Value:

- Simulations can when appropriately validated augment the range of existing scenarios for release
- Contaminant releases can be tested with surrogates, but can be modeled with