

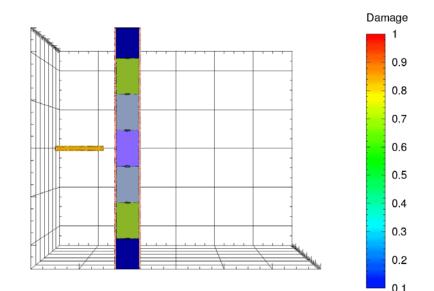
Nuclear Energy

Numerical Estimation of the Spent Fuel Ratio

Samuel Durbin Eric Lindgren Jason Wilke Sandia National Laboratories

Jon Margraf and Tim Dunn Lawrence Livermore National Laboratory

SAND2016-5418 C





Motivation

Nuclear Energy

- Releases of spent nuclear fuel (SNF) by sabotage could have significant impacts to the public health and nuclear industry
 - Need to quantify the amount released (source term)
 - Subject of research for almost 40 years in US
 - Early studies were overly conservative due to lack of data
 - Model refinements as a result of testing

5.6 > 3 > 1.2

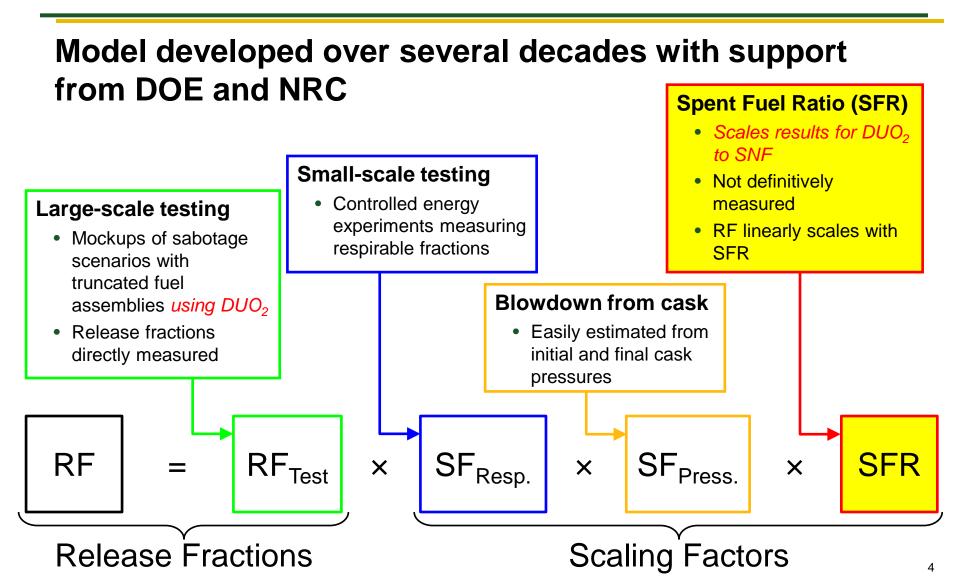


Spent Fuel Ratio (SFR)

- Concern that surrogate fuel pellets may aerosolize differently than actual spent fuel
 - Spent fuel pellets undergo changes to bulk material properties such as density and porosity due to irradiation
- Data needed to scale release fractions determined from previous large-scale tests conducted with surrogate (DUO₂)
- SFR quantifies the respirable aerosols produced by an high energy device (HED) acting on spent fuel compared to a surrogate material
 - SFR = $\frac{\text{RF}_{\text{Spent Fuel}}}{\text{RF}_{\text{Surrogate}}}$, Aerodynamic Equivalent Diameter (AED) < 10 µm
 - Comparisons must be made under identical conditions
 - Statistically significant number of experiments are required
 - Or modeling using acceptable, simplifying assumptions
- Underlying physics highly complex



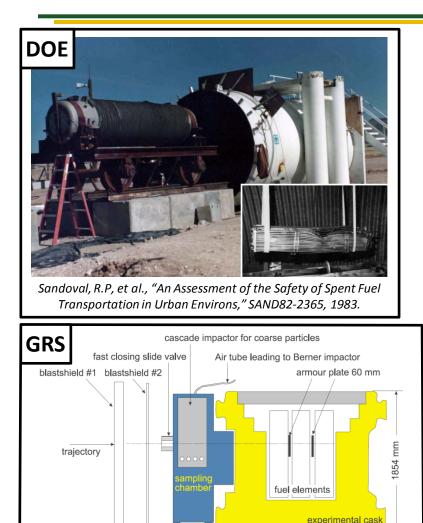
Current Source Term Evaluation





Large-Scale Cask Sabotage Testing

Nuclear Energy



DOE sponsored full-scale test of obsolete truck cask (SAND82-2365)

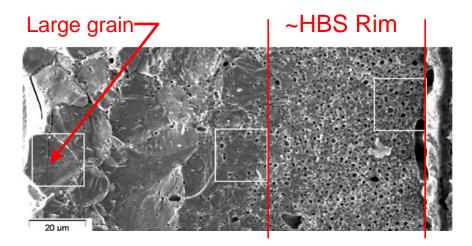
- High energy density device (HED) directed at cask
- 15×15 PWR truncated assembly with DUO₂
 - Cask and fuel unpressurized
 - ~3 g released in "respirable" range
- GRS sponsored full-scale test mimicking CASTOR (Lange, et al.)
 - 17×17 PWR assemblies with DUO₂ pressurized to 40 bar
 - First two tests (1 bar) released ~1 g
 - Third test (0.8 bar) 0.35 g

Lange, F., et al., "Experiments to Quantify Potential Releases and Consequences from Sabotage Attack on Spent Fuel Casks," 13th Int. Sym. on Packaging and Transportation of Radioactive Materials, Chicago, IL, 2001.



Significant Differences between DUO₂ and SNF

Nuclear Energy



Small grains -

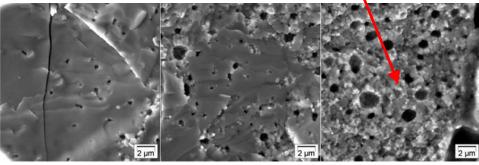


Fig. 11. SEM Fractograph of the 73 GWd/tU Sample Periphery

NOIROT et al., High Burnup Changes in UO2 Fuels Irradiated up to 83 GWd/t in M5® Claddings NUCLEAR ENGINEERING AND TECHNOLOGY, VOL.41 NO.2 MARCH 2009 - SPECIAL ISSUE ON THE WATER REACTOR FUEL PERFORMANCE MEETING 2008

Bulk changes from irradiation

- Density decreases
 - Porosity increases
 - Pellet swells

Grain size decreases

- ~20 μm grains in fresh fuel
- ~0.5 μm grains in high burnup structure

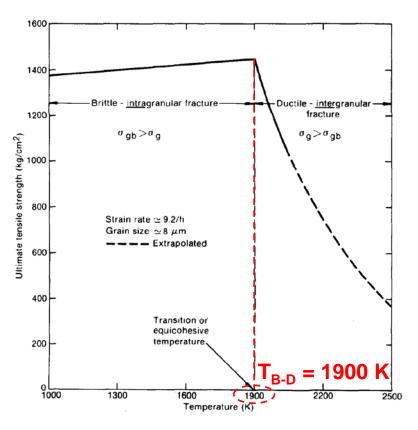
High Burnup Structure (HBS)

- ~60 μm thick rim
 - Small volume fraction
- Rim burnup ~2x bulk burnup
- Possible to simulate properties as f(r) with current modeling tools



Importance of the Transition Temperature

Nuclear Energy



A.W. Cronenberg, T.R. Yackle "Intergranular fracture of unrestructured fuel," Journal of Nuclear Materials 84 (1979) 295-318.

- Brittle-ductile transition T_{B-D} = 1900 K
- Brittle fracture if T_{Fracture} ≤ T_{B-D}
 - Fractures through the ceramic grains (intragranular)
 - Argument for fractures independent of grain size
 - Respirable generation for SNF and DUO₂ should be similar for same energy density (i.e. SFR ≈1)

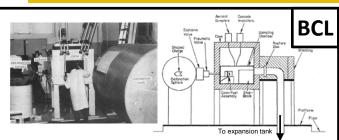
Ductile fracture if T_{Fracture} > T_{B-D}

- Fractures along grain boundaries (intergranular)
- Size distribution of particles would be similar to grain size distribution
 - SNF would produce more respirable aerosols than DUO₂ (i.e. SFR > 1)

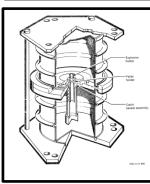


Previous SFR Measurement Attempts

Nuclear Energy



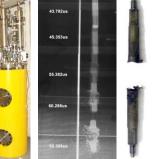
Schmidt, E.W., et al., "Final Report on Shipping Cask Sabotage Source Term Investigation," NUREG/CR-2472, 1981.



Alvarez, J.L. and Kaiser, B.B., "Waste Forms Response Project Correlation Testing," EGG-PR-5590, 1982.

INL

SNL



Molecke, M.A., et al., "Spent Fuel Sabotage Test Program, Characterization of Aerosol Dispersal: Interim Final Report," SAND2007-8070.

No definitive value to date

- Large degree of experimental scatter
- Battelle Columbus Laboratories
 - <u>SFR = 0.42 to 0.71</u>
 - Analysis of BCL results by Sandoval (SAND82-2365)
 - <u>SFR = 2.5 to 12</u>
 - Subsequent review by Luna (SAND99-0963)
 - Current RF calculations assume <u>SFR = 3</u>
- Idaho National Laboratory
 - <u>SFR = 5.6</u>
 - Based on questionable extrapolation of wet sieve data
 - Value used in previous analyses
 - <u>SFR = 0.53</u>
 - Bulk aerosol measurements

Sandia National Laboratories

- Testing on different surrogate materials resulted in similar respirable release fractions
 - Provided confidence in using lower SFR estimate
 - No SNF testing



Current Modeling Approach

Nuclear Energy

- Model DUO₂ and SNF as continuum in shock physics code
 - Interactions at the grain level not explicitly modeled
- Same equation of state for DUO₂ and SNF
 - Mie-Grüneisen

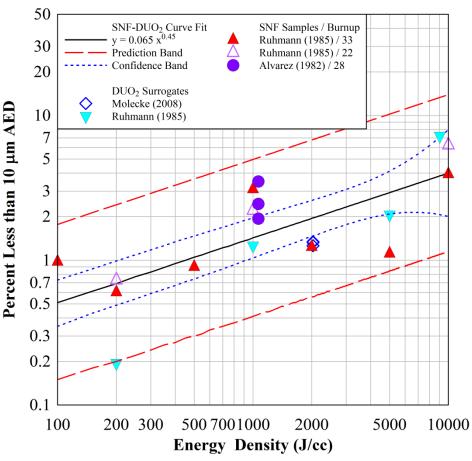
Differences in SNF explored by:

- Decreasing density (density ↓ as burnup ↑) along with the P-Alpha porous material model
- Quantify the average, internal energy density rise in the target material
- Aerosol generation estimated from empirical fit of DUO₂ and SNF data
 - Quantifies mass fraction less than 10 µm AED as a function of internal energy density
 - Low energy density and non-UO₂ samples discarded for these analyses



Energy Density Determines Release

Nuclear Energy



Alvarez, J.L. and Kaiser, B.B., EGG-PR-5590, 1982.

Molecke, M.A., et al., SAND2007-8070, 2008.

Ruhmann, H., et al., "Research Program on the Behavior of Burnt-Up Fuel under Strong Mechanical Impacts," Kraftwerk Union, Report R 917/85/002, (1985).

Empirical aerosol model

- Percent of sample smaller than 10 µm AED after subjected to sudden energy input
- Additional surrogate data ignored for these analyses (CeO₂, SYNROC, concrete, and various glasses)
- Respirable fraction 1 as energy density 1
 - Roughly square root dependence
- All SNF data for relatively low burnup
 - Authors unaware of any high burnup data



Y (cm)

-2

X (cm)

0

-2

0

X (cm)

-2

0

X (cm)

2

-2

0

2

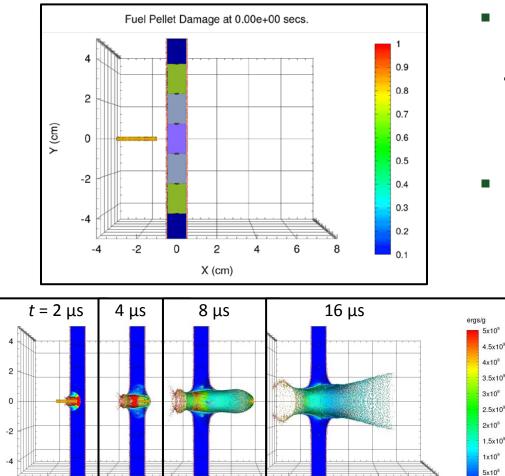
X (cm)

4

0

Shock Physics Modeling

Nuclear Energy



- High velocity copper jet impacts perpendicularly into fuel segment
 - 7 pellet segment of a 15x15 PWR fuel rod

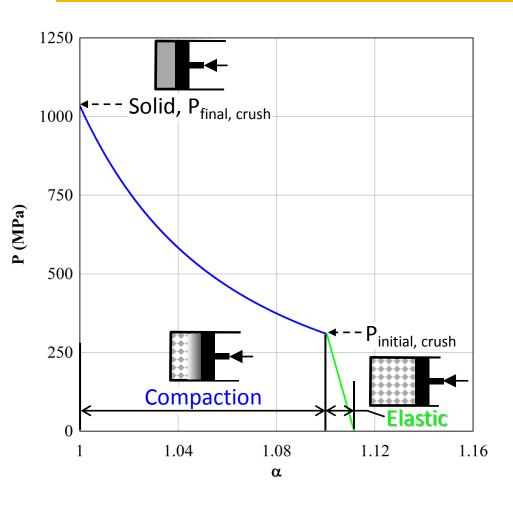
Modeling with CTH

- Shock physics code developed at SNL
 - Explicit Eulerian code developed for solving high strain transient dynamics problems
 - Explosions and high velocity impact problems
 - Mie-Grüneisen EOS
 - P-Alpha crush model for porous media



Porous Material Modeling (P-α)

Nuclear Energy



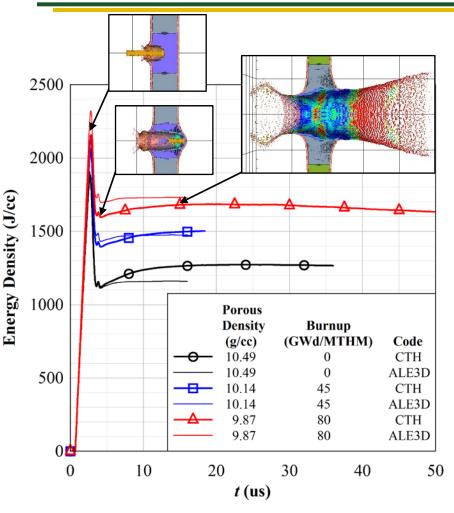
 P-Alpha used to model porous material behavior

$$\alpha = \frac{\rho_{\text{solid}}}{\rho}$$

- Initially elastic when stress is applied
- Pores are crushed as stress is increased
 - Irreversible process
 - Plastic compression
- Eventually all pores are eliminated
 - Material behaves as solid and follows solid Hugoniot curve (Mie Gruneisen)



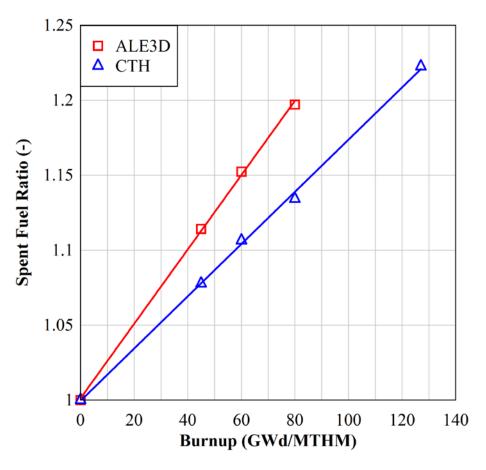
Internal Energy Density Results



- Internal energy density for central fuel pellet only
- More energy absorbed as porosity increases
 - Additional work to compact material to solid density
 - 6% \downarrow density \Rightarrow 33% \uparrow energy density
- Simulations insensitive to choices in P-Alpha model
 - Varied initial and final crush pressures by 7.5× and $3\times$ from baseline values, respectively
 - Less than 1% change to energy density
- Aerosol model is valid based on T_{Fracture} < 1900 K
 - Results assume T_o = 300 K
 - Max. energy density = 1680 J/cc
 - For storage T_{Fuel} < 700 K
 - Max. energy density = 1970 J/cc
 - Energy density = 3700 J/cc to reach T_{Fuel} = 1900 K



Spent Fuel Ratio Results

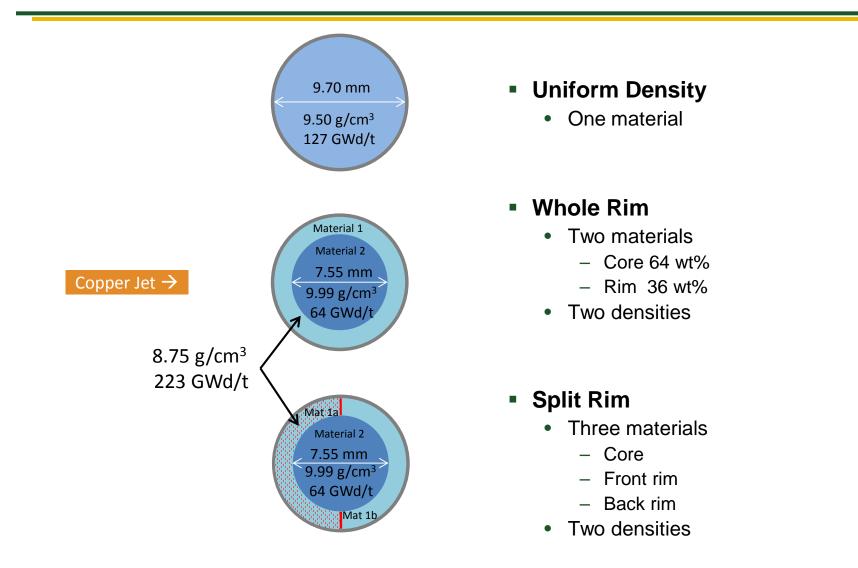


- CTH results trend lower than ALE3D
- **SFR ≈ 1.20** for 80 GWd/MTHM
- **SFR** ≈ **1.15** for 60 GWd/MTHM
- SFR effectively linear with burnup (and density)
- Calculated SFR at least 2.5× smaller than previously assumed

Density	Burnup	CTH	ALE3D
(g/cc)	(GWd/MTHM)	SFR	SFR
10.49	0	1.00	1.00
10.14	45	1.08	1.11
10.02	60	1.11	1.15
9.87	80	1.13	1.20
9.50	127	1.22	



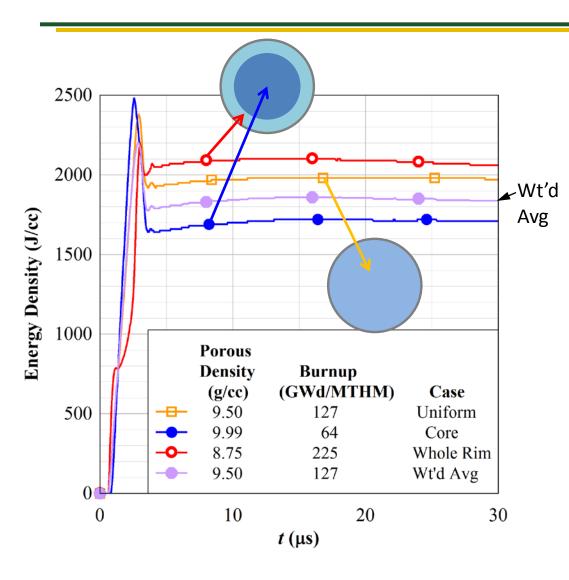
Rim Case Domains





Whole Rim Case

Nuclear Energy



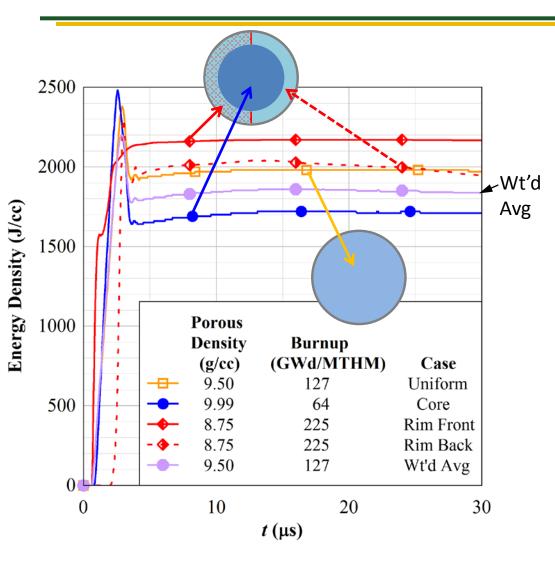
Deposited energy density

- Highest in rim
- Lowest in core
 - Uniform in between
- Weighted average slightly lower than the uniform case



Split Rim Case

Nuclear Energy



Partitioning of rim gives more spatial detail

- Overall similar response to single rim case
- Highest in front rim
- Lower in back rim
- Lowest in core
- Weighted average slightly lower than the uniform case



Rim Case Summary

Nuclear Energy

CTH Porous Rim Cases (9.50 g/cc average density)

- Extreme case (average burnup 127 GWd/MTHM)
- Front rim gives max SFR=1.27
- Wt'd avg rim and uniform cases similar
 - SFR = ~1.2 (by CTH, ALE3D probably higher)
 - Rim inclusion did not increase SFR

	Case	Pellet Mass (g)	Density (g/cc)	Burnup (GWd/MTHM	Energy Density (J/cc)	Resp. (%)	SFR (-)	
			No]	Rim				
	Uniform	10.7	9.50	127	1985	1.98	1.22	
	Whole Rim							
	Core	6.8	9.99	64	1717	1.86	1.15	
└──	Rim	3.9	8.75	223	2105	2.03	1.25	
	Wt avg	10.7	9.50	127	1858	1.92	1.19	\rightarrow
			Split	Rim				
	Core	6.8	9.99	64	1717	1.86	1.15	7
	Front rim	1.9	8.75	223	2170	2.06	1.27	
	Back rim	1.9	8.75	223	2039	2.01	1.24	
	Wt avg	10.7	9.50	127	1858	1.92	1.19	





Nuclear Energy

Large-scale sabotage testing scaled by Spent Fuel Ratio (SFR)

- All tests used DUO₂ surrogate
- Need SFR for source term analyses

Previous testing efforts to define SFR were indeterminate

• Large uncertainties in SFR

Modeling alternative to additional testing demonstrated

- Shock physics codes excellent for providing insight into SFR
- Preliminary numerical investigations indicate SFR ≈ 1—
 - Well within values defined by SFR test data
 - Not confirmed by new test data
- Simulations of high burnup fuel (80 GWd/MTHM)
 - Model also used for even higher porosity and radius dependent calculations
- Reducing SFR decreases calculated release
 - Significant impact possible 🗲

1.2 < 3 < 5.6