Numerical Estimation of the Spent Fuel Ratio

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- 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 (DUO$_2$)
- SFR quantifies the respirable aerosols produced by a high energy device (HED) acting on spent fuel compared to a surrogate material
  - SFR = $\frac{RF_{\text{Spent Fuel}}}{RF_{\text{Surrogate}}}$, Aerodynamic Equivalent Diameter (AED) < 10 $\mu$m
  - Comparisons must be made under identical conditions
    - Statistically significant number of experiments are required
    - Or modeling using acceptable, simplifying assumptions
- Underlying physics highly complex
Model developed over several decades with support from DOE and NRC

**Spent Fuel Ratio (SFR)**
- Scales results for DUO₂ to SNF
- Not definitively measured
- RF linearly scales with SFR

**Small-scale testing**
- Controlled energy experiments measuring respirable fractions

**Large-scale testing**
- Mockups of sabotage scenarios with truncated fuel assemblies using DUO₂
- Release fractions directly measured

**Release Fractions**

\[
RF = RF_{Test} \times SF_{Resp.} \times SF_{Press.} \times SFR
\]

**Blowdown from cask**
- Easily estimated from initial and final cask pressures
Large-Scale Cask Sabotage Testing

- **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


Significant Differences between DUO₂ and SNF

- **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

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

- **Brittle-ductile transition** $T_{B-D} = 1900 \, \text{K}
- **Brittle fracture if** $T_{\text{Fracture}} \leq T_{B-D}$
  - Fractures through the ceramic grains (intragranular)
  - Argument for fractures independent of grain size
    - Respirable generation for SNF and DUO$_2$ should be similar for same energy density (i.e. $\text{SFR} \approx 1$)
- **Ductile fracture if** $T_{\text{Fracture}} > T_{B-D}$
  - Fractures along grain boundaries (intergranular)
  - Size distribution of particles would be similar to grain size distribution
    - SNF would produce more respirable aerosols than DUO$_2$ (i.e. $\text{SFR} > 1$)

Previous SFR Measurement Attempts

- No definitive value to date
  - Large degree of experimental scatter
- Battelle Columbus Laboratories
  - $SFR = 0.42$ to $0.71$
    - Analysis of BCL results by Sandoval (SAND82-2365)
  - $SFR = 2.5$ to $12$
    - Subsequent review by Luna (SAND99-0963)
    - Current RF calculations assume $SFR = 3$
- Idaho National Laboratory
  - $SFR = 5.6$
    - Based on questionable extrapolation of wet sieve data
    - Value used in previous analyses
  - $SFR = 0.53$
    - 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

- **Model DUO$_2$ and SNF as continuum in shock physics code**
  - Interactions at the grain level not explicitly modeled

- **Same equation of state for DUO$_2$ and SNF**
  - Mie-Grüneisen

- **Differences in SNF explored by:**
  - Decreasing density (density $\downarrow$ as burnup $\uparrow$) 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 DUO$_2$ and SNF data**
  - Quantifies mass fraction less than 10 $\mu$m AED as a function of internal energy density
  - Low energy density and non-UO$_2$ samples discarded for these analyses
Energy Density Determines Release

- **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 \( \uparrow \) as energy density \( \uparrow \)
  - Roughly square root dependence

- **All SNF data for relatively low burnup**
  - Authors unaware of any high burnup data

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Shock Physics Modeling

- 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)

- 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% ↓ density ⇒ 33% ↑ energy density
- Simulations insensitive to choices in P-Alpha model
  - Varied initial and final crush pressures by 7.5× and 3× 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
- $SFR \approx 1.20$ for 80 GWd/MTHM
- $SFR \approx 1.15$ for 60 GWd/MTHM
- SFR effectively linear with burnup (and density)
- Calculated SFR at least 2.5× smaller than previously assumed

<table>
<thead>
<tr>
<th>Density (g/cc)</th>
<th>Burnup (GWd/MTHM)</th>
<th>CTH SFR</th>
<th>ALE3D SFR</th>
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<td>10.49</td>
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<td>1.22</td>
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Rim Case Domains

- Uniform Density
  - One material

- Whole Rim
  - Two materials
    - Core 64 wt%
    - Rim 36 wt%
  - Two densities

- Split Rim
  - Three materials
    - Core
    - Front rim
    - Back rim
  - Two densities

Copper Jet →

- Material 1: 8.75 g/cm³, 223 GWd/t
- Material 2: 7.55 mm, 9.99 g/cm³, 64 GWd/t
Whole Rim Case

- Deposited energy density
  - Highest in rim
  - Lowest in core
    - Uniform in between

- Weighted average slightly lower than the uniform case
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**

- **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

<table>
<thead>
<tr>
<th>Case</th>
<th>Pellet Mass (g)</th>
<th>Density (g/cc)</th>
<th>Burnup (GWd/MTHM)</th>
<th>Energy Density (J/cc)</th>
<th>Resp. (%)</th>
<th>SFR (–)</th>
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<td>1.19</td>
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Summary

- **Large-scale sabotage testing scaled by Spent Fuel Ratio (SFR)**
  - All tests used DUO$_2$ 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 $SFR \approx 1$
    - 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**