AGR-3/4 Modeling and Transport Parameters

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AGR-3/4 Modeling and Transport Parameters

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Outline

• AGR-3/4 experiment and objectives
• Description of AGR-3/4 modeling approach and inputs
  ▪ Source term (release from particles)
  ▪ Temperature profiles
  ▪ Diffusion through concentric rings
  ▪ Adsorption across gaps between rings
• Simplified Source Model and updated model results
• Reconstruction of fission product distributions from gamma tomography data
The AGR-3/4 Experiment

- An in-pile experiment “designed to provide data on fission product diffusivities in fuel kernels and sorptivities and diffusivities in compact matrix and graphite materials”
- 20 “designed to fail” (DTF) particles placed at center of fuel stack to provide source of fission products
- Irradiated to ~370 EFPD in the Advanced Test Reactor
Modeling approach and objectives

- Basic assumption: fission products (Ag, Cs, Sr, Eu) migrate radially through the rings via Fickian diffusion:

\[
\frac{\partial C(r,t)}{\partial t} = \frac{1}{r} \frac{\partial}{\partial r} \left( rD(T(r,t)) \frac{\partial C(r,t)}{\partial r} \right) + S(r,t,T(r,t))
\]

- Modeling objective: predict releases to within +/- 1 order of magnitude

- Necessary inputs:
  - Diffusion coefficients (GA data)
  - Source term
    - (PARFUME or measured values)
  - Temperature distribution (ABAQUS model)
  - Boundary conditions (sorption isotherms)

ATR Physics Calculations  \(\rightarrow\)  ABAQUS Temperatures, \(T(r,t)\)  \(\rightarrow\)  Diffusion Coefficient and Sorption Isotherm Data

PARFUME or PIE: Mass Sources, \(f(t)\)  \(\rightarrow\)  COMSOL Multiphysics FP Transport, \(f(r,t)\)
Simplified source model

• Pre-test predictions made use of a release from particles (both DTF and intact) that was predicted by a PARFUME model

• We now have reasonably complete PIE measurements of what was actually released from particles during the course of the irradiation (for capsules 3, 5, and 7)

• We have used this data to create a simplified source model

• This assumes that fission products were released at a constant rate from DTF particles (i.e. near the centerline of the compacts)

• The rate is fixed to as to match measured inventories at the end of irradiation, minus any measured on foils and spacers
  - There is no representation of these in a 1D model

• This assumption is clearly inapplicable to silver, which will have a significant contribution from intact driver fuel at high temperature

• Eu models developed based on legacy Sr transport parameters
Thermal Analysis

- 3D, time-dependent ABAQUS analysis (INL/EXT-15-35550)
- Based on detailed ATR operating history
- Includes variable gap conductivity due to changing gas composition
- The input to COMSOL is the radial profile at the center (axially) of the capsule, as a function of time
Diffusion Coefficients

- From TECDOC-978, GA report 911200
- Arrhenius law with temperature:

\[ D = D_0 \exp\left(-\frac{Q}{RT}\right) \]

<table>
<thead>
<tr>
<th>Fission Product</th>
<th>Material</th>
<th>( D_0 ) (m(^2)/s)</th>
<th>( Q ) (kJ/mol)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ag</td>
<td>Matrix</td>
<td>1.6</td>
<td>258</td>
</tr>
<tr>
<td></td>
<td>Graphite</td>
<td>1.38e-2</td>
<td>226</td>
</tr>
<tr>
<td>Cs</td>
<td>Matrix</td>
<td>3.60e-4</td>
<td>189</td>
</tr>
<tr>
<td></td>
<td>Graphite</td>
<td>1.70e-6</td>
<td>149</td>
</tr>
<tr>
<td>Sr</td>
<td>Matrix</td>
<td>1.00e-2</td>
<td>303</td>
</tr>
<tr>
<td></td>
<td>Graphite</td>
<td>1.70e-2</td>
<td>268</td>
</tr>
</tbody>
</table>

- Direct analytical estimation of Sr, Eu diffusivity from inner ring PIE data suggests these are similar
- Eu model initiated using Sr diffusivity
Boundary Conditions - Sorption Isotherms

• The partial pressure of an FP in the gap between rings is related to its concentration at the surface by a sorption isotherm, consisting of Henry (linear) and Freundlich (power law) regimes:

\[
\frac{P}{P_{\text{ref}}} = \exp\left( A + \frac{B}{T} \right) \left( \frac{C}{C_{\text{ref}}} \right)^{d_1 + \frac{E}{T}} + \exp\left[ \left( A + \frac{B}{T} \right) + \left( D - 1 + \frac{E}{T} \right)(d_1 - d_2 T) \right] \frac{C}{C_{\text{ref}}}
\]

• It is assumed that both surfaces are in equilibrium with this pressure

• Different temperatures (and different materials) on either side of the gap result in a discontinuity in concentration across it; fluxes balance

• No Ag data available; Cs values used

• Sr values used for Eu model
Cesium-134

- Capsules 3,5 approximately meet +/- 10x objective
- Capsule 7 displays unexpected trend from compacts to matrix ring
Cesium-137

- Similar trends as Cs-134
Strontium-90

- Low, flat concentration profile (like fast diffusion) measured across rings inconsistent with high gradient in near-surface region (slow diffusion)
Europium-154

• Trends very similar to Sr-90
Tomographic image reconstruction

- AGR-3/4 rings were subject to gamma scanning
- Initial attempts at image reconstruction using filtered backprojection produced images with artifacts and some unphysical attributes
  - “Measured” activity in the void outside the ring
- Method was relied on to do two different tasks:
  - Reconstruct activity profile within the ring (presumably smooth)
  - Identify the boundaries of the ring itself (edge detection; discontinuous)
- Solution: reconstruct image in a polar coordinate system
  - Ring dimensions are known in detail from PIE metrology
  - Annular region can be defined in polar coordinates that coincides exactly with the ring geometry
  - It is impossible by definition for activity to be outside the ring in this case- the edge detection problem is eliminated
  - Iterative reconstruction methods are then employed that can enforce a degree of smoothness on the profiles
  - Can be benchmarked against physically sampled data
## Example: Cs-134, Capsule 7 Inner Ring

### Physically sampled data

<table>
<thead>
<tr>
<th>Radius of Inner Ring (mm)</th>
<th>Concentration [Ci/m³]</th>
</tr>
</thead>
<tbody>
<tr>
<td>5.0</td>
<td>4.5E+01</td>
</tr>
<tr>
<td>7.0</td>
<td>4.0E+01</td>
</tr>
<tr>
<td>9.0</td>
<td>3.5E+01</td>
</tr>
<tr>
<td>11.0</td>
<td>3.0E+01</td>
</tr>
<tr>
<td>13.0</td>
<td>2.5E+01</td>
</tr>
</tbody>
</table>

### Old Method

- Plot showing concentration distribution vs. radius.
- Graph showing sampled data points.

### New Method

- Graph showing improved concentration distribution.
- Additional graph comparing old and new methods.

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**Note:** The diagrams illustrate concentration distributions and sampled data points for Cs-134 in Capsule 7 Inner Ring, comparing old and new methods.
Summary

• AGR-3/4 models are now being updated based on PIE data
• A simplified source model based on measured inventories is a first step in model adjustment
• Some expected and some unexpected trends have been observed
• Diffusion coefficient and sorption isotherm parameters will be adjusted in further iterations, with an objective of a predictive model for all capsules to within +/- 1 order of magnitude
• A new image reconstruction technique has been developed and is successfully reproducing fission product distributions measured by physical sampling
  ▪ Minor modification needed to account for off-center rings during scanning
• Tomography data produced by validated reconstruction methods supplements destructive measurements from PIE to better inform parameter estimation