Hot Cell Gamma Scanning of the AGR TRISO Fuel Experiments for Fission Product Distribution, Inventory, and Burnup

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**VHTR Fuels Program**

- Tri-structural isotropic (TRISO) coated particle fuel
  - Coatings provide fission product containment during reactor operation and during high temperature accident scenarios
  - Fuel is fabricated with very low defect fraction (<10^{-5})

- A series of irradiation experiments are in progress to evaluate TRISO fuel performance

- Advanced Gas Reactor (AGR)-1 completed in November 2009
  - Peak calculated burnup of 19.5% FIMA
  - Zero particle failures observed during the irradiation
AGR-1 PIE

• Advanced Gas Reactor (AGR)-1 objectives
  – Shakedown
  – Provide early performance data
  – Support selection of reference fuel

• AGR-1 post-irradiation examination (PIE) objectives
  – Assess overall irradiation test train performance to verify simulations (i.e. burnup)
  – Evaluate fission product retention
  – Characterize compacts and particles with microscopy
**AGR-1 Compact Gamma Scanning**

- Gamma-ray scans of all 72 AGR-1 compacts have been completed.
- Non-destructive burnup evaluation using gamma spectrometry has been performed using two techniques:
  - Direct fission product inventory
  - Ratio of shielded to direct fission product inventories
- Destructive burnup by mass spectrometry:
  - Residual Heavy Atom method and ICP-MS
  - Four compacts spanning the burnup range
- Measured burnup values have been compared to detailed production/depletion simulations.
Gamma Scanning of AGR-1 Fuel

- Compact gamma scanning was performed using Idaho National Laboratory (INL) Precision Gamma Scanner (PGS) system
- Key features of the PGS system
  - Penetration of Hot Fuel Examination Facility (HFEF) main cell wall
  - Rectangular collimator 22.2 mm wide and adjustable from 0.025 to 2.5 mm tall
  - Collimator rotates between vertical and horizontal alignment
  - Control Software automatically adjusts sample position after a set live time and saves spectra
  - HPGe detector with Compton suppression
  - Can accept commercial LWR fuel (4.3m)
- AGR-1 compact scanning parameters
  - 2.5 mm collimator height
  - Collimator viewed axial “slices” of each compact
  - Ten minute live time per slice
  - All 72 Compacts were scanned
PGS Collimator Detail
Characterization of PGS System

- AGR-1 compacts were placed in individual aluminum containers and loaded into a larger tube for scanning along with three Eu-152 calibration sources.
- Gamma-ray spectra were analyzed using the INL PC-GAP software package to calculate an activity per slice.
- PC-GAP outputs were analyzed using a control script.
- Conversion of counts in a spectrum to activities requires:
  - Energy calibration (Eu-152 sources)
  - Efficiency calibration
    - Eu-152 sources
    - MCNP simulation for finer geometry and self-shielding corrections
    - Self-shielding was not significant above 300 keV.
**AGR-1 Fission Product Inventory Simulations**

- Detailed production/depletion calculations of the AGR-1 experiment fission product and actinide inventories were performed using as-run reactor conditions*

- Burnup for each calculation node (half compact) was calculated by:

\[
BU = \frac{N_{U-235,i} + N_{U-238,i} - N_{U-235,f} - N_{U-238,f} - N_{Act,f}}{N_{U-235,i} + N_{U-238,i}}
\]

- Fission product inventory for each node (half compact) was also tracked and the neutron cross sections of all important fission products were updated at each depletion step

- In addition to burnup, measured fission product inventories were compared to the predicted fission product inventories in the simulations

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**Measured Fission Products**

- Several isotopes were present in the PGS scans: Y-91, Zr-95, Ru-103, Rh-106 (Ru-106), Sb-125, Cs-134, Cs-137, Ce-141, Ce-144, Eu-154, Pr-144 (Ce-144)

- Some of these isotopes are of special interest for burnup evaluation
  - Direct longer lived, Cs-137, Sb-125
  - Shielded Cs-134, Eu-154

- Compact activities were estimated by summing the “slice” activity for the set of spectra that corresponded to a specific compact

- Calculated-to-Experimental (C/E) ratio for many isotopes (Cs-137, Cs-134, Ce-144) was very near to 1.0 for all compacts

- It appears that the simulation did over-predict some isotopes like Eu-154 and Sb-125
Measured Fission Products C/E

<table>
<thead>
<tr>
<th>Capsule 6</th>
<th>Capsule 5</th>
<th>Capsule 4</th>
<th>Capsule 3</th>
<th>Capsule 2</th>
<th>Capsule 1</th>
</tr>
</thead>
</table>

Cs-137
Cs-134
Sb-125
Eu-154
Ce-144 (Pr-144)
Ru-106 (Rh-106)
Conversion of Measured Activities to Burnup

- Non-destructive burnup evaluation using gamma spectrometry has been performed using two techniques
  - Direct fission product inventory
  - Ratio of shielded to direct fission product inventories
- Measured burnup values have been compared to detailed production/depletion simulations
- Relationships between final burnup and final fission product activity or activity ratio were developed to predict burnup from the PGS measurements
- Both gamma-ray spectrometry burnup analysis methods have challenges
  - Direct activity: Efficiency calibration, local variations not captured
  - Activity ratio: Neutron energy spectrum shifts, tracking cross sections
- C/E results suggest an optimum indicator for both methods
  - Direct activity: Cs-137
  - Activity ratio: Cs-134 / Cs-137 (shielded FP) / (direct FP)
Burnup from PGS Measurement and Simulation by Direct Cs-137 Activity

- **Peak Simulation Burnup**: 19.5% FIMA
- **Peak Measured Burnup**: 20.1% FIMA
- **Minimum Simulation Burnup**: 11.2% FIMA
- **Minimum Measured Burnup**: 10.1% FIMA

Axial Position of Fuel Compact in AGR-1 Test Train
Burnup from PGS Measurement and Simulation by Cs-134 / Cs-137 Activity Ratio

- **Capsule 6**
  - Peak Simulation Burnup: 19.6% FIMA
  - Minimum Simulation Burnup: 11.2% FIMA

- **Capsule 5**
  - Minimum Measured Burnup: 10.7% FIMA

- **Capsule 4**
  - Peak Measured Burnup: 20.0% FIMA

- **Capsule 3**

- **Capsule 2**

- **Capsule 1**

**Legend**:
- ECAR-958 Burnup Simulation Stack 1 and Stack 3
- ECAR-958 Burnup Calculation Stack 2
- Measured Burnup Stack 1 (Cs-134/137)
- Measured Burnup Stack 2 (Cs-134/137)
- Measured Burnup Stack 3 (Cs-134/137)
Slice Specific Burnup by Cs-134 / Cs-137 Activity Ratio

Experiment Axial Position

Burnup % FIMA

- Capsule 1
- Capsule 2
- Capsule 3
- Capsule 4
- Capsule 5
- Capsule 6
- ECAR 958 Burnup Simulation
Mass Spectrometry Burnup

- For each mass spectrometry burnup analysis
  - Compact is deconsolidated
  - Three sets of 20 randomly selected particles crushed
  - Burned and dissolved in nitric acid or modified nitric acid
  - Aliquots analyzed by ICP-MS
- Four Compacts Spanning the Burnup range
- Burnup by – “Fission Product Monitor – Residual Heavy Atom Technique”

\[
BU = \frac{\left( \frac{N_{fp}}{Y_{fp}} \right)}{\left( \frac{N_{fp}}{Y_{fp}} \right) + N_{Act}} \times 100
\]
Mass Spectrometry Burnup

- Fission products used in this method should have:
  - Small neutron absorption cross sections
  - High cumulative fission yields
  - Similar fission yield for the isobar (A) between U and Pu
  - Readily dissolve during leaching
- In this work La-139, Ce-140, Ce-142, Pr-141, Nd-145 and Nd-146 were used:
  - Nd-145, Nd-146 grouped together
  - Effective fission yields were used
  - Average from isotopes
  - Average of aliquots
- Separate Nd-148 measurement
Mass Spectrometry Burnup - Results

• Results available for four compacts
  – Low burnup 6-3-2
  – High burnup 3-2-1
  – 5-3-1
  – 1-3-1

• Agreement for all three methods is within 6%

• Significantly better than historical data

<table>
<thead>
<tr>
<th>Compact</th>
<th>Mass spectrometry</th>
<th>Gamma spectrometry</th>
<th>Simulation</th>
</tr>
</thead>
<tbody>
<tr>
<td>6-3-2</td>
<td>10.7(±0.5) %</td>
<td>11.0(±0.3) %</td>
<td>11.31 %</td>
</tr>
<tr>
<td>3-2-1</td>
<td>19.3(±1.0) %</td>
<td>18.6(±0.6) %</td>
<td>18.98 %</td>
</tr>
<tr>
<td>5-3-1</td>
<td>16.3(±0.8) %</td>
<td>15.9(±0.5) %</td>
<td>16.88 %</td>
</tr>
<tr>
<td>1-3-1</td>
<td>16.3(±0.8) %</td>
<td>15.6(±0.5) %</td>
<td>15.98 %</td>
</tr>
</tbody>
</table>
Overview of Capsule Fission Product Inventory

- Fission product migration in tri-structural isotropic (TRISO) fuel is a critical metric of fuel performance.
- Various AGR-1 capsule components were examined for condensable fission products released from the fuel compacts:
  - Metallic capsule components, spacers, gas lines, graphite fuel holders, fuel compacts
  - Ag, Ce, Cs, Eu, Sr
- The graphite holders were scanned using a semi-tomographic technique.
- Measured activities are decay corrected and compared to predicted fission product inventory.
- Available compacts were gamma scanned for Ag-110m content.
- Ag-110m capsule mass balance calculated.
Capsule Fission Product Inventory

Measurement methodology:

• Capsule shells
  – Leach with nitric acid and measure radionuclide inventory

• Graphite holders
  – Two off-axis scans
    • Generate axial profile
    • Select locations for horizontal scans
  – Gamma scan in-cell
    • Transverse scans at select levels in two azimuthal directions to generate activity concentration maps
  – Crush and gamma count in Analytical Lab Hot cell

• Graphite spacers
  – Gamma count out-of-cell

• Gas exit lines
  – Gamma counted forty 25 mm segments out-of-cell; no significant amounts of fission products were found
Graphite Holder Ag-110m Location

- High variability in Ag-110m distribution
- Ag-110m concentrated at:
  - Ends of Capsule 1 and 6 graphite holders
  - Middle of Capsule 2, 3, and 5 graphite holders
- Concentration maps:
  - Usually concentrated near Stacks 1 and 3
  - Cooler regions near Mo through tubes and at outer edge of holders
- Absolute Ag-110m activity from the two measurement methods agrees within 6%
  - Efficiency for absolute activity was determined from Monte Carlo modeling
Elevated Cs-134 and Cs-137 activity found at Level 2 in Holder 5

Transverse mapping indicates activity is concentrated near Stacks 1 and 3

Compacts 5-2-3 and 5-2-1 were sent to Oak Ridge National Laboratory (ORNL) for complete irradiated microsphere gamma analysis (IMGA) to locate any particles with defective SiC

Two defective SiC TRISO were found by ORNL in Compact 5-2-3
**Capsule Cs Inventory**

- Cs release from intact fuel is <3E-6
- Elevated release of ~1.2E-5 found in:
  - **Capsule 6** (confirmed one defective SiC in Compact 6-3-2 by DLBL)
  - **Capsule 5**
    - Cs hot spot in graphite suggests one or more defective SiC
    - Confirmed by ORNL advanced IMGA

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### Cs-134

- One particle ~2E-5
- Cross-hatched columns indicate activity below detection limit

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1 defective SiC Compact 6-3-2

Cs hot spot in graphite holder
Capsule Eu Inventory

- Total Eu release less than ~5E-4
  - Indicates release through intact particles
- Baseline fuel has slightly higher release (Capsules 3, 6)
  - Very different time average volume average temperatures
  - Very different Ag-110m release
- Most Eu released from particles remains in fuel compact matrix (HTR2012-3-021)

- One particle ~2E-5
- Cross-hatched columns indicate activity below detection limit
Capsule Ag Inventory

- Capsule Ag-110m inventory ranges from 1 to 38%.
  - End capsules have majority of Ag on capsule shells
    - Possibly related to increased graphite-capsule gap?
  - Middle capsules have majority of Ag in graphite holders
- No clear trends with fuel type
- No clear trends with capsule average temperatures
Compact Gamma Re-Scanning for Ag-110m

- Selected compacts have been re-scanned with live time >1 h per slice to quantify Ag-110m
- Ag-110m activity of slices summed to get total Ag-110m activity per compact
- Compare measured Ag-110m activity to predicted activity to estimate in-pile silver loss and determine capsule mass balance
Compact Ag-110m Inventory

- 56 of 72 compacts have been scanned (16 unavailable)
- Ag-110m retention varies from 8% to 108%
- Ag retention higher at top and bottom of capsules
- Compacts at same level in Stacks 1 and 3 have very similar Ag retention
  - Can use one compact in Stack 1 or 3 to “fill in” data where the other compact is missing
- Cannot determine if Ag may have migrated from one compact to another
  - Usually very little Ag in compact matrix (HTR2012-3-021)
Capsule Ag-110m Mass Balance

- Release fraction for unscanned compacts assumed to be average of those scanned to estimate totals

- Near complete mass balance for all capsules

- Capsule 6:
  - Simulations over-predict Pu
  - Simulations over-predict Burnup
  - Implies Ag-110m over prediction
  - Largest potential error from estimating unscanned compacts
  - Largest potential for loss of silver in chemical analysis
Summary

• Gamma-ray spectrometry scans of irradiated TRISO fuel from the AGR-1 experiment have been completed using the PGS system located at HFEF.

• The burnup results confirm the accuracy of the nondestructive burnup evaluation from gamma spectrometry for TRISO fuel compacts across a burnup range of approximately 10 to 20 % FIMA and also validate the approach used in the physics simulation of the AGR-1 experiment.

• Fission product inventory has been extensively surveyed in the AGR-1 capsule components and compacts:
  – Spatial distribution of Ag and Cs studied
  – Locating Cs activity in graphite fuel holder has lead to identification of particles with defective SiC for further study

• Fractional releases from compacts:
  – Very low Cs release from intact SiC (<3E-6)
  – Eu released from intact SiC (1.3 – 4.8E-4)
  – Ce release very low (<1E-5)

• Significant and varied fractional releases of Ag-110m from compacts:
  – Capsule average release: 1 – 38%
  – Individual compact release: 0 – 92%