PREIRRADIATION MICROSTRUCTURAL CHARACTERIZATION OF FFTF MIXED-OXIDE FUEL

DE Rasmussen
PS Schaus

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OBJECTIVES

- Evaluate potential for fuel to undergo densification during irradiation
- Assure PuO₂ homogeneity in mixed oxide fuel
- Provide data base for pre-/post-irradiation comparisons
- Evaluate effect of fuel fabrication conditions
PROTOTYPIC PuO$_2$-UO$_2$ FUEL PELLETS
PuO₂ & UO₂ POWDERS

MECHANICALLY BLEND PuO₂ & UO₂

ADD ORGANIC BINDER/LUBRICANT

PRESLUG

GRANULATE

ADD ORGANIC BINDER/LUBRICANT

FINAL PRESS

REMOVE ORGANIC

SINTER ADJUST O/M

CENTERLESS GRIND

Pu & UNITRATE SOLUTION

COPRECIPITATED PuO₂-UO₂

Fuel Fabrication Process Flow Diagram
LOW PRESSURE PRESLUGGED FUEL

- MECHANICALLY BLENDED PuO₂ AND UO₂ POWDERS

- FOUR COMPOSITIONS

<table>
<thead>
<tr>
<th>TYPE</th>
<th>PuO₂ (WT %)</th>
<th>NUMBER OF LOTS SAMPLED</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.2</td>
<td>22.5</td>
<td>5</td>
</tr>
<tr>
<td>4.2</td>
<td>25.3</td>
<td>3</td>
</tr>
<tr>
<td>3.1</td>
<td>27.5</td>
<td>4</td>
</tr>
<tr>
<td>4.1</td>
<td>29.4</td>
<td>1</td>
</tr>
</tbody>
</table>

HIGH PRESSURE PRESLUGGED FUEL

- COPRECIPITATED MIXED OXIDE (PuO₂-UO₂) FROM MIXED Pu, U NITRATE SOLUTION

- TWO COMPOSITIONS

<table>
<thead>
<tr>
<th>TYPE</th>
<th>PuO₂ (WT %)</th>
<th>NUMBER OF LOTS SAMPLED</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.2</td>
<td>22.5</td>
<td>2</td>
</tr>
<tr>
<td>3.1</td>
<td>27.5</td>
<td>3</td>
</tr>
</tbody>
</table>
22.5% PuO₂ PELLET
LOW PRESSURE PRESLUGGED
25.3% PuO₂ PELLET
LOW PRESSURE PRESHLUGGED
29.4% PuO₂ PELLET

LOW PRESSURE PRESLUGGED
22.5% PuO₂ PELLET

HIGH PRESSURE PRESLUGGED
27.5% PuO₂ PELLET

HIGH PRESSURE PRESLUGGED
ASSESSMENT OF POTENTIAL FOR DENSIFICATION DURING IRRADIATION

- QUANTITATIVE MICROSCOPY
- OPTICAL MICROGRAPHS
- SCANNING ELECTRON MICROGRAPHS
- TRANSMISSION ELECTRON MICROGRAPHS OF REPLICA
- IMAGE ANALYSIS
- DATA REDUCTION/STATISTICAL UNFOLDING
TRACE LOCATIONS ON LONGITUDINAL PELLET CROSS SECTIONS
MINIMUM POROSITY $\leq 2 \mu m$

MAXIMUM POROSITY $\leq 2 \mu m$

(ORIGINAL 6000X)

HEDL TEST FUELS OF REFERENCE DENSITY 90.4 $\pm$ 2 %TD
"TRUE" SIZE CALCULATED BY STATISTICAL UNFOLDING

A = APPARENT PORE SIZE FOR AREA PERCENT PORE SIZE ANALYSIS

CONCEPTS OF STATISTICALLY UNFOLDED AND AREA PERCENT PORE SIZE DISTRIBUTIONS
LOW PRESSURE PRESLUGGED (27.5% PuO$_2$)

HIGH PRESSURE PRESLUGGED (27.5% PuO$_2$)

OPTICAL MICROGRAPHS (ORIGINALS 600X)
LOW PRESSURE PRESLUGGED (27.5% PuO$_2$)

HIGH PRESSURE PRESLUGGED (27.5% PuO$_2$)

SCANNING ELECTRON MICROGRAPHS OF CATHODIC VACUUM ETCHED CROSS SECTION

(ORIGINALS 6000X)
LOW PRESSURE PRESLUGGED (27.5% PuO₂)

HIGH PRESSURE PRESLUGGED (27.5% PuO₂)

TRANSMISSION ELECTRON MICROGRAPHS OF REPLICAS OF CATHODIC VACUUM ETCHED CROSS SECTIONS (ORIGINALS 8000X)
LOW PRESSURE PRESLUGGED (27.5% PuO$_2$)

HIGH PRESSURE PRESLUGGED (27.5% PuO$_2$)

TRANSMISSION ELECTRON MICROGRAPHS OF REPLICAS OF CATHODIC VACUUM ETCHED CROSS SECTIONS (ORIGINALS 40,000X)
FIGURE 1. Envelopes of cumulative spherical pore volume distributions for FFTF fuel lot pellets characterized by statistical unfolding and regression fitting shown with respect to range of experience for HEDL irradiation test fuels of density 90.4 ± 2.0% of theoretical density that have been successfully irradiated.
FIGURE 2. ENVELOPES OF CUMULATIVE SPHERICAL PORE VOLUME DISTRIBUTIONS FOR FFTF FUEL PELLETS CHARACTERIZED BY AREA PERCENT POROSITY SHOWN WITH RESPECT TO RANGE OF EXPERIENCE FOR HEDL-FABRICATED TEST FUELS OF DENSITY 90.4 ± 2.0% TD THAT HAVE BEEN SUCCESSFULLY IRRADIATED.
CONCLUSIONS

POTENTIAL FOR DENSIFICATION DURING IRRADIATION

• FAVORABLE PORE SIZE DISTRIBUTIONS

• LESS SUBMICRON POROSITY THAN PREVIOUS HEDL TEST FUELS WHICH WERE SUCCESSFULLY IRRADIATED IN EBR-II

• EXPECT ACCEPTABLE DENSIFICATION BEHAVIOR
FUEL QUALITY - PuO₂ HOMOGENEITY

- PuO₂ HOMOGENEITY FIGURE OF MERIT
- AUTORADIOGRAPHIC ANALYSIS (QUALITATIVE)
- ELECTRON MICROPROBE ANALYSIS (QUANTITATIVE)
DETERMINATION OF FUEL PELLET HOMOGENEITY
USING FIGURE OF MERIT REFERENCE PELLETS

(FIGURES OF MERIT DETERMINED FROM ELECTRON MICROPROBE
ANALYSIS, ALL ALPHA-AUTORADIOGRAPHS ENLARGED TO 30x)

FIGURE OF MERIT (M)

REFERENCE FUEL PELLET
CONTAINING PuO₂ ENRICHED
CONCENTRATION ZONES OF KNOWN
SIZES AND CONCENTRATIONS

30 WT.% PuO₂ CONCENTRATION ZONES
A = 480 MICRONS
B = 690 MICRONS
C = 560 MICRONS

25 WT.% PuO₂ - 75 WT.%
MATRIX MATERIAL

100 WT.% PuO₂ CONCENTRATIONS ZONES
D = 160 MICRONS
E = 115 MICRONS
AUTORADIOGRAPH OF 27.5% PuO₂ PELLET

LOW PRESSURE PRESLUGGED

MECHANICALLY BLENDED FEED POWDER

30X
AUTORADIOGRAPH OF 27.5% PuO₂ PELLET
HIGH PRESSURE PRESLUGGED
COPRECIPITATED FEED POWDER
MICROPROBE TRACE OF 27.5% PuO$_2$ LOW PRESSURE PRESLUGGED PELLET
MECHANICALLY BLENDED FEED POWDER

MICROPROBE TRACE OF 27.5% PuO$_2$ HIGH PRESSURE PRESLUGGED PELLET
COPRECIPITATED FEED POWDER
PuO₂ HOMOGENEITY FIGURE OF MERIT

- **AVERAGE OF ALL LOCAL FIGURES OF MERIT**

- **LOCAL FIGURE OF MERIT** - QUOTIENT OF CALCULATED RATE OF ENERGY DEPOSITION AT THE LOCAL POINT IN THE FUEL DIVIDED BY THE DEPOSITION RATE WHICH WOULD EXIST IN A COMPLETELY HOMOGENEOUS FUEL OF AVERAGE COMPOSITION

- RELATES TO THE HOMOGENEITY OF THE MIXED OXIDE FUEL BY APPROXIMATING THE PORTION OF THE FERTILE MATERIAL WHICH INSTANTANEOUSLY INCREASES IN TEMPERATURE DUE TO AN INSTANTANEOUS REACTOR TRANSIENT

- A VALUE OF 0.96 INDICATES THAT APPROXIMATELY 96 PERCENT OF THE FERTILE MATERIAL IN THE FUEL IMMEDIATELY INFLUENCES THE TEMPERATURE COEFFICIENT IN AN INSTANTANEOUS REACTOR TRANSIENT

- A COMPLETELY HOMOGENEOUS FUEL HAS A VALUE OF 1.00
PuO₂ HOMOGENEITY FIGURE OF MERIT OF FFTF MIXED OXIDE FUELS

MINIMUM ACCEPTABLE LEVEL

FIGURE OF MERIT

A B C E J X L N I H MIN. MAX.

Wt% PuO₂

22.5 25.3 27.5 29.4 HEDL FUELS

LOW PRESSURE PRESLUGGED FUELS (MECHANICALLY BLENDED POWDER)

MINIMUM ACCEPTABLE LEVEL

FIGURE OF MERIT

T

Wt% PuO₂

22.5 HEDL FUELS

HIGH PRESSURE PRESLUGGED FUELS (COPRECIPITATED POWDER)
SIMULATED PuO$_2$ DISTRIBUTION (FROM MICROCOMPOSITION ANALYSIS)

EQUIVALENT PuO$_2$ PARTICLE:
A "CALCULATED" PARTICLE COMPOSED OF 100% PuO$_2$, CONTAINING THE AMOUNT OF PuO$_2$ IN A RICH REGION IN EXCESS OF THE OVERALL SAMPLE MEAN

CONCEPT OF 100% PuO$_2$ EQUIVALENT PARTICLE
EXPERIENCE ENVELOPE

- HEDL TEST FUELS
- LOW PRESSURE PRESLUGGED (MECHANICALLY BLENDED)
- HIGH PRESSURE PRESLUGGED (COPRECIPITATED)

100% PuO₂ EQUIVALENT PARTICLE DIAMETER ———→

100% PuO₂ EQUIVALENT PARTICLE EXPERIENCE ENVELOPES
CONCLUSIONS

FUEL QUALITY

- VERY GOOD PuO₂ HOMOGENEITY

- OBSERVED PuO₂ HOMOGENEITY FIGURE OF MERIT VALUES OF 0.99 TO 1.00, COMPARED WITH A VALUE OF 1.00 FOR A COMPLETELY HOMOGENEOUS FUEL

- PuO₂ HOMOGENEITY EQUIVALENT TO OR BETTER THAN PREVIOUS HEDL TEST FUELS WHICH WERE SUCCESSFULLY IRRADIATED IN EBR-II
DATA BASE/FABRICATION EFFECTS ANALYSES

OPEN POROSITY

- PELLET DENSITY DETERMINATION
- PENETRATION OF MERCURY INTO SURFACE-CONNECTED PORES

GRAIN STRUCTURE

- PELLET SECTIONING/POLISHING/ETCHING
- GRAIN SIZE DETERMINATIONS
- EXAMINATION FOR POSSIBLE SECOND PHASE
OPEN POROSITY RESULTS FOR FFTF FUELS

LOW PRESSURE PRESLUGGED FUELS (MECHANICALLY BLENDED POWDER)

HEDL FUELS

HIGH PRESSURE PRESLUGGED FUELS (COPRECIPITATED POWDER)
HIGH PRESSURE PRESLUGGED (27.5% PuO₂)

ETCHED PELLET CROSS SECTIONS (ORIGINALS 500X)

LOW PRESSURE PRESLUGGED (27.5% PuO₂)

HIGH PRESSURE PRESLUGGED (27.5% PuO₂)
GRAIN SIZE RESULTS FOR FFTF FUELS

LOW PRESSURE PRESLUGGED FUELS (MECHANICALLY BLENDED POWDER)

HIGH PRESSURE PRESLUGGED FUELS (COPRECIPITATED POWDER)
CONCLUSIONS

DATA BASE/FABRICATION EFFECTS

• FABRICATION CONDITIONS AFFECTED PORE SIZE DISTRIBUTION

• FUTURE FUELS MADE USING DIFFERENT PROCESSING CONDITIONS WILL REQUIRE MICROSTRUCTURAL ANALYSIS TO ASSESS POTENTIAL FOR DENSIFICATION DURING IRRADIATION

• FABRICATION CONDITIONS AFFECTED OPEN POROSITY

• ALL CHARACTERIZED FUELS WERE A SINGLE PuO₂-UO₂ PHASE AFTER SINTERING