Post-irradiation characterization of zirconia- and spinel-based inert matrix fuels – the OTTO experiment (abstract & presentation slides)

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Abstract

The OTTO experiment is a joint project between JAERI, PSI and NRG, initiated to study the incineration of plutonium by means of non-reprocessable inert matrix fuels (once-through mode). The sample set comprises i) spinel-based targets with micro-dispersed (d<25 µm) and macro-dispersed (d~250 µm) Pu-bearing zirconia microspheres and ii) zirconia-based solid solutions. Irradiation was performed in the High Flux Reactor (HFR) in Petten for 548 full power days, and was completed in December 2002. Gamma spectrometry and tomography reveal the effect of the temperature during irradiation on the spatial distribution of the fission products. Fission gas releases appear to be very low for the micro-dispersed targets; the other targets show significantly higher values, 5-10%. On the other hand the macro-dispersed target shows unacceptable swelling. The macro-dispersed targets do not exhibit swelling; the fission product damage is confined within the fissile phase. Shrinkage of the zirconia-based fissile inclusions has been observed in both types of the spinel-based targets, concurrent with increased porosity and increased Xe-concentration around fissile inclusions. Shrinkage of the zirconia-based solid solution targets has resulted in severe cracking, but their overall behaviour compares to that of UO2.

Keywords: post-irradiation examination techniques, inert matrix fuels, plutonium incineration, zirconia, spinel
Post-irradiation characterization of zirconia- and spinel-based inert matrix fuels - the OTTO experiment

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OTTO project

- Collaboration between JAERI, PSI, and NRG, started in 1998
- Objective:

To study Pu incineration in a Once Through Then Out mode

- Both spinel-based and-zirconia based targets
- Fabrication at PSI and NRG
- Irradiation HFR started october 2000
- Irradiation HFR (548 FPD) completed 30 december 2002
- Post Irradiation Examinations completed march 2004
OTTO = Once Through incineration

- Geochemically stable inert matrices
  - insoluble in nitric acid
  - suitable for final storage

A. Zirconia (ZrO$_2$)
   - cubic phase, i.e. single phase system (Zr,Y,Pu,Er)O$_{2-x}$
   - excellent irradiation performance
   - low thermal conductivity

B. Spinel (MgAl$_2$O$_4$)
   - two phase system with (Zr,Y,Pu,Er)O$_{2-x}$
   - high thermal conductivity compared to UO$_2$ and ZrO$_2$
   - amorphisation / swelling due to fission product damage

Two-phase system: macro- vs. microdispersion

Macro: fission product damage confined to the fissile particle

Micro: fission product damage in the inert matrix as well.
Macro vs. microdispersion

 Macrodispersion

 Microdispersion

 OTTO test matrix

<table>
<thead>
<tr>
<th>Capsule</th>
<th>Composition</th>
<th>Dispersion</th>
<th>Fissile phase vol%</th>
<th>Pu_{88} (BOI) g/cm³</th>
<th>Supplier</th>
<th>Instrumentation</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>(Zr,Y,Pu,Er)O₂</td>
<td>Solid solution</td>
<td>single phase</td>
<td>0.37</td>
<td>PSI</td>
<td>-</td>
</tr>
<tr>
<td>2</td>
<td>(Zr,Y,Pu,U)O₂</td>
<td>Solid solution</td>
<td>single phase</td>
<td>0.34</td>
<td>JAERI</td>
<td>-</td>
</tr>
<tr>
<td>3</td>
<td>(Zr,Y,Pu,Er)O₂+MgAl₂O₄</td>
<td>&lt;25 μm</td>
<td>20.8</td>
<td>0.32</td>
<td>NRG</td>
<td>TC</td>
</tr>
<tr>
<td>4</td>
<td>(Zr,Y,Pu,U)O₂+MgAl₂O₄</td>
<td>&lt;25 μm</td>
<td>20.7</td>
<td>0.31</td>
<td>JAERI</td>
<td>TC</td>
</tr>
<tr>
<td>5</td>
<td>(Zr,Y,Pu,Er)O₂+MgAl₂O₄</td>
<td>200-250 μm</td>
<td>19.7</td>
<td>0.31</td>
<td>NRG</td>
<td>TC</td>
</tr>
<tr>
<td>6</td>
<td>(Zr,Y,Pu,U)O₂+MgAl₂O₄</td>
<td>200-250 μm</td>
<td>19.7</td>
<td>0.30</td>
<td>JAERI</td>
<td>-</td>
</tr>
<tr>
<td>7</td>
<td>(Pu,U)O₂</td>
<td>Solid solution</td>
<td>single phase</td>
<td>0.39</td>
<td>PSI</td>
<td>TC</td>
</tr>
</tbody>
</table>
OTTO irradiation characteristics

- Irradiation was performed in HFR in-core position H8:
  - start of irradiation: 7 November 2000
  - end of irradiation: 30 December 2002
  - irradiation time: 548 full power days (45 MWth)
  - thermal flux: $\sim 7 \cdot 10^{17} \text{ m}^2\text{s}^{-1}$
  - gamma heating: $\sim 2.0 \text{ W/g}$
  - burn-up of Pu: $\sim 30-35 \%$ (IMF), $13 \%$ (MOX)
  - linear power: $\sim 200 / \sim 150 \text{ W/cm (BOI / EOI)}$
    $\sim 135 / \sim 115 \text{ W/cm (BOI / EOI)}$

Post Irradiation Examination

- Non-destructive PIE:
  - visual inspection
  - gamma spectrometry
  - tomography (3 capsules)
  - profilometry
  - X-ray imaging
  - puncturing and fission gas analysis

* Destructive PIE:
  - ceramography
  - microprobe analysis
Capsule 4 cladding failure

X-ray images after irradiation

1
2
3
4
5
6
7
Gamma spectrometry - macrodispersion

- RU-106
- CS-137
- ZR-95
- NB-65

No Cs-diffusion ==> lower irradiation temperatures

Large gap between pellets

Counts (per minute)

Axial position from the bottom of the capsule (mm)

Tomography

Tomography confirms conclusions of the gamma spectrometry:

- Cs diffusion towards the cladding in capsule 2 (zirconia)
- Swelling and distortion of pellets in capsule 4 (microdispersion)

- no Cs diffusion towards the cladding in capsule 6 (macrodispersion) - lower temperatures during irradiation and confinement of fission products
Gas puncturing

<table>
<thead>
<tr>
<th>Capsule</th>
<th>Kr</th>
<th>Xe</th>
<th>Xe / Kr ratio</th>
<th>Remark</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>10.37%</td>
<td>9.94%</td>
<td>17.0</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>5.55%</td>
<td>6.74%</td>
<td>17.0</td>
<td>no Kr detected</td>
</tr>
<tr>
<td>3</td>
<td>0.00%</td>
<td>0.05%</td>
<td></td>
<td>capsule failed</td>
</tr>
<tr>
<td>4</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>5.19%</td>
<td>4.55%</td>
<td>15.8</td>
<td></td>
</tr>
<tr>
<td>6</td>
<td>5.35%</td>
<td>6.67%</td>
<td>16.5</td>
<td></td>
</tr>
<tr>
<td>7</td>
<td>0.00%</td>
<td>0.20%</td>
<td></td>
<td>no Kr detected</td>
</tr>
</tbody>
</table>

- Very low gas release in micro-dispersed fuel and MOX
- Gas release about 5% in macro-dispersed fuel
- Gas release 5 – 10 % in zirconia-based IMF

Destructive PIE: ceramography capsule 1

Solid solution of (Zr,Y,Pu,Er)O₂ (homogeneous)
Destructive PIE: ceramography capsule 1

Solid solution of (Zr,Y,Pu,Er)O₂ (homogeneous)

Ceramography capsule 1
Ceramography capsule 3

Microdispersed zirconia in spinel

Ceramography capsule 5
Ceramography capsule 5

Zircon radiation damage in rocks

1.5 Ga old zircons in mica (Uvdal, Telemark)
Ceramography conclusions

Ceramography shows satisfactory irradiation behaviour:

- macro-dispersed zirconia in spinel:
  - no swelling (pellet was taken out from the cladding)
  - confinement of fission product damage to immediate surroundings of fissile phase

- micro / macro-dispersed zirconia in spinel:
  - shrinkage of zirconia-based fissile inclusions
  - porosity and increased Xe-concentration around fissile inclusions

- Solid solution of (Zr,Y,Pu,Er)O₂
  - shrinkage of the pellet ==> cracks
  - behaviour comparable to UO₂
Suitability of OTTO matrices

- Are the OTTO matrices suitable for once-through Pu incineration?

  Zirconia-based inert matrix fuels (solid solution) ➔ +

  Spinel-based inert matrix fuels (microdispersive) ➔ -

  Spinel-based inert matrix fuels (macrodispersive) ➔ +/-

Axial overview of the OTTO sample holder

24 thermocouples, including 4 central thermocouples, monitor the irradiation
X-ray images prior to irradiation

Central temperatures in the OTTO irradiation

- Sample holder shifted vertically
- Thermocouple of Capsule 4 failed

Graph: Central temperature / °C vs Irradiation time / days
# Swelling

The axial swelling of the fuel stack from the neutron radiographs

<table>
<thead>
<tr>
<th>Capsule</th>
<th>Description</th>
<th>Fissile stack length at BOI</th>
<th>Axial swelling after 7 cycles</th>
<th>Axial swelling after 14 cycles</th>
<th>Axial swelling (EOI), $\gamma$-spect.</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$(Zr,Ti,Pu,Er)O_2$ (Solid Solution)</td>
<td>67.260 mm</td>
<td>-0.10 ± 0.19 %</td>
<td>0.05 ± 0.32 %</td>
<td>1.8 ± 0.6 % (?)</td>
</tr>
<tr>
<td>2</td>
<td>$(Zr,Y,Pu,U)O_2$ (Solid Solution)</td>
<td>66.833 mm</td>
<td>-0.24 ± 0.06 %</td>
<td>0.12 ± 0.14 %</td>
<td>0.2 ± 0.6 %</td>
</tr>
<tr>
<td>3</td>
<td>$(Zr,Y,Pu,Er)O_2$ + spinel (&lt; 25 μm)</td>
<td>66.687 mm</td>
<td>1.72 ± 0.14 %</td>
<td>-</td>
<td>2.7 ± 0.6 %</td>
</tr>
<tr>
<td>4</td>
<td>$(Zr,Y,Pu,U)O_2$ + spinel (&lt; 25 μm)</td>
<td>67.190 mm</td>
<td>3.75 ± 0.49 %</td>
<td>4.13 ± 0.33 %</td>
<td>5.1 ± 0.6 %</td>
</tr>
<tr>
<td>5</td>
<td>$(Zr,Y,Pu,Er)O_2$ + spinel (250 μm)</td>
<td>66.111 mm</td>
<td>0.57 ± 0.27 %</td>
<td>1.94 ± 0.32 %</td>
<td>1.0 ± 0.6 %</td>
</tr>
<tr>
<td>6</td>
<td>$(Zr,Y,Pu,U)O_2$ + spinel (250 μm)</td>
<td>65.547 mm</td>
<td>0.75 ± 0.19 %</td>
<td>0.39 ± 0.31 %</td>
<td>1.3 ± 0.6 %</td>
</tr>
<tr>
<td>7</td>
<td>MOX</td>
<td>66.427 mm</td>
<td>1.30 ± 0.20 %</td>
<td>1.49 ± 0.25 %</td>
<td>1.7 ± 0.6 %</td>
</tr>
</tbody>
</table>

# Profilometry results

![Graph showing Profilometry results](image)

Curves shifted for clarity; nominal outer diameter = 9.50 mm
Gamma scan device

Gas puncturing (II)

<table>
<thead>
<tr>
<th>Capsule</th>
<th>Released amount (mole)</th>
<th>Computed amount (mole)</th>
<th>Computed amount (mole) II</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Kr</td>
<td>Xe</td>
<td>Kr</td>
</tr>
<tr>
<td>1</td>
<td>3.68E-06</td>
<td>6.47E-05</td>
<td>6.32E-05</td>
</tr>
<tr>
<td>2</td>
<td>2.65E-06</td>
<td>4.28E-05</td>
<td>6.32E-05</td>
</tr>
<tr>
<td>3</td>
<td>0.00E+00</td>
<td>2.53E-07</td>
<td>6.32E-05</td>
</tr>
<tr>
<td>4</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>1.44E-06</td>
<td>2.28E-05</td>
<td>6.32E-05</td>
</tr>
<tr>
<td>6</td>
<td>1.90E-06</td>
<td>3.13E-05</td>
<td>6.32E-05</td>
</tr>
<tr>
<td>7</td>
<td>0.00E+00</td>
<td>9.10E-07</td>
<td>6.32E-05</td>
</tr>
</tbody>
</table>

Some differences in pre- and post-irradiation computations of the produced gas amounts.
Ceramography capsule 4