Design, development and qualification of advanced fuels for an industrial ADS prototype


OUTLINE:
• Objectives & Background
  • Addressed topics
  • Some results:
    Core configuration and performances
    Thermomechanical behaviour of the pins
    FUTURIX-FTA, HELIOS, BODEX tests
    Thermo-chemical compatibility tests
  • Conclusion
Objectives & background for fuel developments

• Objectives:
  - Ranking of fuel concepts according to in-pile behaviour, out-of-pile properties, predicted behaviour in normal operating conditions and safety performance.
  - Recommendations for the most promising fuel.

• Background:
  - Emphasis in Europe on oxide-based fuels
    - CERCER (Pu, MA)O$_2$ + MgO and CERMET (Pu, MA)O$_2$ + $^{92}$Mo
    - First development in the frame of the FP5 - FUTURE program: best candidates according to performance, safety and fabricability criteria, synthesis of oxide compounds, out-of-pile characterisation.
    - Strong synergy with transmutation target programs
    - Large industrial experience on oxide fuel fabrication for critical reactors
  - Nitride-based fuels: (Pu,MA,Zr)N
    - Development in the frame of the FP5 - CONFIRM program: (Am,Zr)N synthesis, irradiation of (Pu,Zr)N pellets in HFR, out-of-pile measurements
    - Development by JAEA

backup solution

reference fuels
Topics addressed within the project

• **TRU-fuel design and performance assessment:**
  – Neutronic design of CERCER and CERMET cores
  – Neutronic and thermo-mechanical behaviour from BOL to EOL

• **Safety Analysis:** transients conditions (ULOF, UTOP, …) and accidents

• **In-pile experiments:**
  – PIE on an irradiated CONFIRM pin: (Pu,Zr)N fuel
  – FUTURIX-FTA test in PHENIX
  – HELIOS test in HFR
  – BODEX test in HFR and Post Irradiation Examinations

• **Out of pile experiments:**
  – Thermal and mechanical properties of CERMET, CERCER fuels
  – Chemical compatibility: fuels/clad, fuels/coolant, TRU compounds/Inert Matrices
  – Oxygen potential measurements
  – Phase diagrams: Pu-Am-O, Pu-Am-Zr-O
92Mo-CERMET core configuration and performances

EFIT design specifications:
• 400MWth
• proton beam: 800MeV - 20mA
• Pb target: 11MW - Φ 782mm
• $k_{\text{eff}} \approx 0.97$
• fuel vector
• inlet-outlet Pb $T^\circ$: 400-480°C
• clad and wrapper: T91
• efficiency: ~42kg MA/TWh$_{th}$

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Inner</th>
<th>Medium</th>
<th>Outer</th>
</tr>
</thead>
<tbody>
<tr>
<td>Assembly number</td>
<td>42</td>
<td>90</td>
<td>80</td>
</tr>
<tr>
<td>Wrapper inner width (mm)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pin number/assembly</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Clad outer diameter (mm)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fuel pellet diameter (mm)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fuel/clad gap (mm)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fuel/matrix ratio</td>
<td>35/65</td>
<td>43/57</td>
<td>50/50</td>
</tr>
<tr>
<td>Pu/MA ratio</td>
<td>45/54</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Av. fuel power density (W.cm$^{-3}$)</td>
<td>270</td>
<td>262</td>
<td>211</td>
</tr>
<tr>
<td>Peak pellet linear power (W.cm$^{-1}$)</td>
<td>190</td>
<td>172</td>
<td>154</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>$k_{\text{eff}}$</th>
<th>$k_{\text{source}}$</th>
<th>Void worth</th>
<th>Beta eff.</th>
<th>Doppler Contant</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.97336</td>
<td>0.93337</td>
<td>7335 pcm</td>
<td>192 pcm</td>
<td>-68 pcm</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Initial mass (kg)</th>
<th>Variation (3 year cycle + 3 year cooling)</th>
</tr>
</thead>
<tbody>
<tr>
<td>MA</td>
<td>3610</td>
</tr>
<tr>
<td>Pu</td>
<td>3055</td>
</tr>
<tr>
<td>Total</td>
<td>6665</td>
</tr>
</tbody>
</table>
Thermo-mechanical behaviour at BOL

- fuel, clad and coolant temperatures for the hottest pin in the inner zone
  - 24 hours after start:

- CERCER fuel ($T_{\text{limit}}$: 1860°C)
- CERMET fuel ($T_{\text{limit}}$: 2180°C)
- $T_{\text{clad}}$ ($T_{\text{limit}}$: 550°C)
- $T_{\text{coolant}}$
FUTURIX-FTA test in PHENIX

- In-pile behaviour comparison of 3 fuel types: oxide, nitride, metallic
- Collaboration DOE-JAEA-ITU-CEA
- CERMET and CERCER studies under EUROTRANS project

<table>
<thead>
<tr>
<th>Fuel composition</th>
<th>Max. linear power (W/cm)</th>
<th>T° max. estimated (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\text{Pu}<em>{0.80}\text{Am}</em>{0.20}\text{O}_{2-x} + 86 \text{ vol}%\text{Mo}$</td>
<td>140</td>
<td>1590</td>
</tr>
<tr>
<td>$\text{Pu}<em>{0.23}\text{Am}</em>{0.24}\text{Zr}<em>{0.53}\text{O}</em>{2-x} + 60 \text{ vol}%\text{Mo}$</td>
<td>130</td>
<td>1510</td>
</tr>
<tr>
<td>$\text{Pu}<em>{0.5}\text{Am}</em>{0.5}\text{O}_{2-x} + 80 \text{ vol}%\text{MgO}$</td>
<td>100</td>
<td>1420</td>
</tr>
<tr>
<td>$\text{Pu}<em>{0.8}\text{Am}</em>{0.2}\text{O}_{2-x} + 75 \text{ vol}%\text{MgO}$</td>
<td>80</td>
<td>1260</td>
</tr>
</tbody>
</table>

CERCER and CERMET fuels in pile since may 2007 for ~240 EFPD
- CERCER: 5th ring. Flux: $4.4 \times 10^{15}$ n.cm$^{-2}$s$^{-1}$
- CERMET: 1st ring. Flux: $3.2 \times 10^{15}$ n.cm$^{-2}$s$^{-1}$

Sept. 08: 153 EFPD achieved
**HELIOS test in HFR**

- Influence of microstructure and temperature on gas release and fuel swelling.

<table>
<thead>
<tr>
<th>Fuel composition</th>
<th>$T^\circ$ max. estimated (°C) (Ne+He in gap)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\text{Am}_2\text{Zr}_2\text{O}_7 + 80 \text{ vol} % \text{MgO}$</td>
<td>800</td>
</tr>
<tr>
<td>$\text{Zr}<em>{0.80} \text{Y}</em>{0.13} \text{Am}<em>{0.07} \text{O}</em>{2-x}$</td>
<td>720</td>
</tr>
<tr>
<td>$\text{Pu}<em>{0.04} \text{Am}</em>{0.07} \text{Zr}<em>{0.76} \text{Y}</em>{0.13} \text{O}_{2-x}$</td>
<td>1470</td>
</tr>
<tr>
<td>$\text{Am}<em>{0.22} \text{Zr}</em>{0.67} \text{Y}<em>{0.11} \text{O}</em>{2-x} + 71 \text{ vol} % \text{Mo}$</td>
<td>750</td>
</tr>
<tr>
<td>$\text{Pu}<em>{0.80} \text{Am}</em>{0.20} \text{O}_{2-x} + 84 \text{ vol} % \text{Mo}$</td>
<td>1240</td>
</tr>
</tbody>
</table>

Beginning of the irradiation expected by Nov. 08 for 200 EFPD

0.7g Am/cm³

F. Delage

10-IEMPT
CERCER and CERMET fabrication processes

ITU flowsheet / CERMET:

- Pu, Zr Nitrate Solution
- Droplet to Particle Conversion
- Pu$_x$Zr$_{1-x}$O$_{2.4}$ Microspheres
- Calcination 800°C, air, 2 h
- Actinide Solution
- Solution Infiltration
- Thermal Treatment 800°C
- Actinide Content Gravimetric control
- Metal Powder Mo
- Mixing
- Pressing
- Sintering

CEA flowsheet / CERCER:

- Pu nitrate
- Am nitrate
- H$_2$C$_2$O$_4$
- H$_2$O
- Calcination
- Pu$_x$Am$_y$O$_2$
- calcined MgO
- Molding/grinding
- sieving
- pressing
- Sintering
BODEX test in HFR

- Study of helium build-up and release mechanism study on inert matrices

- $^{10}\text{B}$ surrogate of $^{241}\text{Am}$ to simulate He production: $\frac{10}{5}\text{B} + \frac{1}{0}\text{n} \rightarrow \frac{7}{3}\text{Li} + \frac{4}{2}\alpha$

- Advantages: no Am handling & short irradiation time (~1-2 months)

| ✓ 3 matrices: Mo, MgO, ZrO$_2$ | ✓ 2 T$^\circ$: 800-1200$^\circ$C | ✓ 1.5 mmole B/cm$^3$
|---------------------------------|---------------------------------|-----------------
| ✓ 3 boron compounds: Mo$_2$B / Mo | ZrB$_2$ / ZrO$_2$ | Mg$_3$B$_2$O$_6$ / MgO
| 60-70MPa, 1600$^\circ$C / 5h / Ar, D: 97% | 600MPa, 1600$^\circ$C / 5h / Ar, D: 92% | 800MPa, 1300$^\circ$C / 5h / Ar, D: 78%

✓ 2x3 capsules: 3 pellets doped with $^{10}\text{B}$ + 1 pellet doped with $^{11}\text{B}$ + 1 undoped pellet

Irradiation achieved – PIE on-going

F. Delage

10-IEMPT
TRU-oxides/Inert Matrices compatibility tests

- **Experimental grid:**
  - powder blend
  - 1800K or 1300K - 2x24 h
  - Air/Ar/Ar-H₂ 5%
  - XRD analysis:

<table>
<thead>
<tr>
<th>Atm</th>
<th>PuO₂+MgO</th>
<th>AmO₂+MgO</th>
<th>PuO₂+Mo</th>
<th>AmO₂+Mo</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Air</strong></td>
<td>MgO PuO₂</td>
<td>AmO₂ MgO</td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Ar</strong></td>
<td>MgO PuO₂</td>
<td>Am₂O₃ h &amp; c AmO₂₋ₓ c MgO Other peaks</td>
<td>PuO₂ Mo</td>
<td>Mo Am-Mo-O m?</td>
</tr>
<tr>
<td><strong>Ar/H₂ 5%</strong></td>
<td>PuO₂ MgO</td>
<td>Am₂O₃ h &amp; c MgO</td>
<td>PuO₂₋ₓ c Mo</td>
<td>Am₂O₃ h Mo</td>
</tr>
</tbody>
</table>

[Belin & al., ARWIF 2008]

- no reaction between PuO₂ and Inert Matrices
- minor interactions between AmO₂ and Inert Matrices
Conclusion

• Major results:
  – Reference designs of $^{92}$Mo-CERMET and MgO-CERCER Cores:
    • $\text{MA/(Pu+MA)} \sim 54\%$ - MgO and Mo content $\geq 50\%$
    • transmutation efficiency (1st cycle): 42 kg MA/TWhth - $\Delta \text{Pu} \sim 0$
    • safety under analysis
  – CERCER and CERMET fabrication (20%Am) demonstrated at lab. scale
  – Thermal properties of CERCER, CERMET fuels and (Pu,MA)O$_2$ phases: accurate and reliable data available
  – In-pile fuel behaviour investigation on-going
  – Fuel thermomechanical behaviour modeling under development

• Additional information:
  – Fernandez-Carretero et al. (Oct.8 – 9:00): fuel fabrication
  – Maschek et al. (Oct. 9 – 14:15): Core design and safety analysis
  – Chen et al. (Poster - section IV): Safety studies on the EFIT with CERMET fuel