DEVELOPMENT OF MINOR ACTINIDE TRANSMUTATION BY CRIEPI

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Metal Fuel FBR & Pyro-process

Innovative fuel cycle system is under development in CRIEPI

**U-Pu-Zr Fuel FBR**: Excellent nuclear performances & safety features

**Pyro-reprocessing**: Simultaneous recovery of MA with U and Pu

**Injection Casting Fuel Fabrication**: Simple remote-control operation

Goals:
- Security of the long-term energy supply,
- Reduction of the amount and the toxicity of radioactive waste,
- Improvement of the proliferation resistance.

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**LWR Fuel Cycle**

- U mining → Enrichment → Fuel Fabrication → LWR → Spent Oxide Fuel → Reprocessing (PUREX) → HLLW (MA, FP)

**FBR Metal Fuel Cycle**

- U mining → Enrichment → Fuel Fabrication → Reduction to Metal → Pyro-reprocessing → Fuel Fabrication → Metal Fuel FBR → Repository

HLLW: High-Level Liquid Waste
MA: Minor Actinides
FP: Fission Products
RE: Rare Earths

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MA-Containing Metal Fuel Development

• How much content of MA should be loaded in metal fuel FBR?

  **Evaluation of expected fuel compositions**
  Burnup and recycle calculations of MA & RE in metal fuel FBR cycle
  Mass flow analysis based on the future fuel cycle scenario

• How about the effect of MA addition in metal fuel?

  **Development of MA- and RE-containing U-Pu-Zr alloys**
  Ex-reactor experiments
  Irradiation experiment
  postirradiation examinations
  Characterization of U-Pu-Zr-MA-RE

**Various experimental studies on U-Pu-Zr-MA(-RE) alloys are performed in cooperation with JRC-ITU.**
MA Burnup Performance in Metal Fuel FBR

Large-scale & high-burnup metal fuel core design is assumed as a model of commercial FBR.

<table>
<thead>
<tr>
<th>Output</th>
<th>1,500MWe / 3,900MWt</th>
</tr>
</thead>
<tbody>
<tr>
<td>Core residence time</td>
<td>6years</td>
</tr>
<tr>
<td>Coolant temp.</td>
<td></td>
</tr>
<tr>
<td>inlet / outlet</td>
<td>355 / 510ºC</td>
</tr>
<tr>
<td>Max. cladding temp.</td>
<td>650ºC</td>
</tr>
<tr>
<td>Max. linear power</td>
<td>500W/cm</td>
</tr>
<tr>
<td>Ave. discharge burnup</td>
<td>150GWD/t</td>
</tr>
</tbody>
</table>

Feed compositions and core performance parameters

<table>
<thead>
<tr>
<th></th>
<th>No-MA-makeup</th>
<th>MA-enriched</th>
</tr>
</thead>
<tbody>
<tr>
<td>MA content, wt%</td>
<td>0.8</td>
<td>2.0</td>
</tr>
<tr>
<td>RE content 1, wt%</td>
<td>0.5</td>
<td>1.5</td>
</tr>
<tr>
<td>Pu enrichment, wt%</td>
<td>16.6</td>
<td>16.4</td>
</tr>
<tr>
<td>Makeup MA ratio 2, %</td>
<td>-</td>
<td>37</td>
</tr>
<tr>
<td>Doppler const., Tdk/dT</td>
<td>-2.3×10⁻³</td>
<td>-2.2×10⁻³</td>
</tr>
<tr>
<td>Coolant coeff., Φ/ºC</td>
<td>0.254</td>
<td>0.273</td>
</tr>
</tbody>
</table>

1: D.F. = 10, 2: (MA from LWR) / (MA in FBR fuel)

Transition Scenario from LWRs to FBRs

Assumptions
- 50GWe of LWRs are operated for 50 years before FBR introduction,
- Reactor lifetime is 40 years,
- MA content in the feed is 2 or 5wt%.

Mass Balance of Pu & MA

MA content of 2wt%: MA & Pu are recycled at the almost same time,
5wt%: MA can be consumed in shorter-term.
Mass Flow Evaluation (2)

Transition Scenario from LWRs to FBRs (2)

Assumptions
- 50GWe of LWRs are operated for 75 years before FBR introduction,
- Reactor lifetime is 40 years,
- MA content in the feed is 2wt%.

Mass Balance of Pu & MA

MA content of 2wt%: MA & Pu are balanced.

Miscibilities among U-Pu-Zr-MA-RE

U-Pu-Zr-MA-RE alloys of different compositions were mixed by arc-melting.

→ U-Pu-Zr-MA alloys without RE can be blended homogeneously.

44U-18Pu-10Zr-9Np-5Am-3Ce-10Nd (wt%)

U-Pu-Zr-Np phase

Pu-Am-RE phase

In the alloys of high RE content, → Matrix segregates into upper and lower parts.

39U-22Pu-12Zr-15Np-10Am-0.6Ce-1.8Nd

Pu-Am-RE precipitates

U-Pu-Zr-Np phase

In the alloys of low RE content (≤5%), → RE-rich precipitates were uniformly dispersed.

RE ≤ 5% can be mixed in U-Pu-Zr-MA matrix.

Phase Structures of annealed U-Pu-Zr-MA-RE

U-Pu-Zr-MA-RE alloys were annealed and quenched.

Metallography of U-Pu-Zr-2MA-2RE.

Matrix phase

- ≤ 600°C: Two phase structures
  - \(\zeta + \delta\) at 500°C
  - \(\gamma + \delta\) (or \(\zeta + \delta\)) at 600°C
- ≥ 700°C: Single \(\gamma\)-phase

Am & RE-rich precipitates
- Uniformly dispersing
- Cohesion at grain boundary
  - (≥700°C)
- ~3µm (-2MA-2RE),
- ≥10µm (-5MA-5RE)

Metallography of U-Pu-Zr-5MA-5RE.
Phase transition temperature

Phase transition temperature of U-Pu-Zr(-MA-RE) were measured by dilatometry method.

Dilatometric curves

(a) U-Pu-Zr-2MA-2RE

\[ \zeta + \delta \leftrightarrow \gamma + \delta \]
\[ \gamma + \delta \leftrightarrow \gamma \]

(b) U-Pu-Zr-5MA-5RE

\[ \zeta + \delta \leftrightarrow \gamma + \delta \]
\[ \gamma + \delta \leftrightarrow \gamma \]

(c) U-Pu-Zr

\[ \zeta + \delta \leftrightarrow \gamma + \delta \]
\[ \sim 630^\circ C \quad \gamma + \delta \leftrightarrow \gamma \]
\[ \sim 580^\circ C \quad \zeta + \delta \leftrightarrow \gamma + \delta \]

For all samples,

two distinctive phase transition temperatures at \( \sim 580^\circ C \) & \( \sim 630^\circ C \)

→ Insignificant influence of MA and RE addition up to 5wt%
Other properties

<table>
<thead>
<tr>
<th></th>
<th>U-19Pu-10Zr-5MA-5RE</th>
<th>U-19Pu-10Zr</th>
<th>Reported U-19Pu-10Zr</th>
</tr>
</thead>
<tbody>
<tr>
<td>Melting point [°C]</td>
<td>1207±10</td>
<td>1217±10</td>
<td>1214±75 [2]</td>
</tr>
<tr>
<td>Elasticity</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Young’s modulus [GPa]</td>
<td>93.31</td>
<td>85.22</td>
<td></td>
</tr>
<tr>
<td>Shear modulus [GPa]</td>
<td>35.39</td>
<td>32.65</td>
<td></td>
</tr>
<tr>
<td>Poisson’s ratio</td>
<td>0.32</td>
<td>0.31</td>
<td></td>
</tr>
<tr>
<td>Compatibility with SS *</td>
<td>920-960</td>
<td>970-990</td>
<td></td>
</tr>
</tbody>
</table>

*: Metallurgical reaction temperature between the alloy and stainless steel.

Thermal conductivity

Influence of MA and RE addition ≤ 5wt% is not significant.

Fabrication of MA-containing Metal Fuel

Fuel Fabrication:
U-19Pu-10Zr-2MA-2RE, U-19Pu-10Zr-5MA,
U-19Pu-10Zr-5MA-5RE and U-19Pu-10Zr
MA=60Np-30Am-10Cm, RE=10Y-10Ce-70Nd-10Gd.

<table>
<thead>
<tr>
<th>Fuel Rod diameter</th>
<th>Density</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>U-19Pu-10Zr-2MA-2RE</td>
</tr>
<tr>
<td></td>
<td>U-19Pu-10Zr-5MA-5RE</td>
</tr>
<tr>
<td></td>
<td>U-19Pu-10Zr-5MA</td>
</tr>
<tr>
<td></td>
<td>U-19Pu-10Zr</td>
</tr>
<tr>
<td>4.9 mm</td>
<td>14.73 g/cm³</td>
</tr>
<tr>
<td>20-50 mm</td>
<td>14.66 g/cm³</td>
</tr>
<tr>
<td></td>
<td>15.31 g/cm³</td>
</tr>
<tr>
<td></td>
<td>15.77 g/cm³*</td>
</tr>
</tbody>
</table>

*: Reported value =15.8 g/cm³, J.H.Kittel, et al., N.E.D. 15 (1971)
MA-containing alloys were irradiated in Phénix. 3 metal fuel pins & 16 oxide fuel pins were arranged in an capsule.

- Pin No.1 : U-19Pu-10Zr
- Pin No.2 : U-19Pu-10Zr-2MA-2RE
- Pin No.3 : U-19Pu-10Zr-5MA / -5MA-5RE

Cladding material : 15-15Ti

Burnup goals ~2.5at.% (METAPHIX-1), ~ 7at.% (METAPHIX-2), ~10at.% (METAPHIX-3).

Schematic views of irradiated fuel pins.

Fuel pin arrangement in irradiation capsule.

- Irradiation experiments were carried out from Dec. 2003 to May 2008 in Phénix.
- After cooling, NDT were carried out.
  No excessive damage due to neutron irradiation was observed.
- Irradiated fuel pins are transported to ITU for nondestructive & destructive PIE.
- After the PIE, pyro-reprocessing experiment is planned.
Irradiation Conditions

Irradiation parameters were analyzed taking account of the operation diagram of the Phénix reactor.

Projected Irradiation Conditions for METAPHIX Experiment

<table>
<thead>
<tr>
<th></th>
<th>Pin No.1 U-19Pu-10Zr</th>
<th>Pin No.2 U-19Pu-10Zr +2MA +2RE</th>
<th>Pin No.3(lower) U-19Pu-10Zr +5MA</th>
<th>Pin No.3(upper) U-19Pu-10Zr +5MA +5RE</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Begin of Irradiation</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Max. Linear Power (^1) [W/cm]</td>
<td>350</td>
<td>327</td>
<td>343</td>
<td>332</td>
</tr>
<tr>
<td>Max. Cladding Temp. (^2) [°C]</td>
<td>581</td>
<td>581</td>
<td>581</td>
<td>←</td>
</tr>
<tr>
<td><strong>End of METAPHIX-1 (120EFPD (^3))</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Max. Linear Power (^1) [W/cm]</td>
<td>330</td>
<td>308</td>
<td>325</td>
<td>313</td>
</tr>
<tr>
<td>Max. Cladding Temp. (^2) [°C]</td>
<td>572</td>
<td>572</td>
<td>572</td>
<td>←</td>
</tr>
<tr>
<td>Max. Burnup [at.%]</td>
<td>2.4</td>
<td>2.5</td>
<td>2.4</td>
<td>2.6</td>
</tr>
<tr>
<td><strong>End of METAPHIX-2 (360EFPD (^3))</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Max. Linear Power (^1) [W/cm]</td>
<td>295</td>
<td>276</td>
<td>294</td>
<td>282</td>
</tr>
<tr>
<td>Max. Cladding Temp. (^2) [°C]</td>
<td>556</td>
<td>556</td>
<td>556</td>
<td>←</td>
</tr>
<tr>
<td>Max. Burnup [at.%]</td>
<td>6.9</td>
<td>7.1</td>
<td>7.0</td>
<td>7.5</td>
</tr>
<tr>
<td><strong>End of METAPHIX-3 (900EFPD (^3))</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Max. Linear Power (^1) [W/cm]</td>
<td>268</td>
<td>251</td>
<td>269</td>
<td>256</td>
</tr>
<tr>
<td>Max. Cladding Temp. (^2) [°C]</td>
<td>543</td>
<td>543</td>
<td>543</td>
<td>←</td>
</tr>
<tr>
<td>Max. Burnup [at.%]</td>
<td>10.9</td>
<td>11.2</td>
<td>11.2</td>
<td>11.9</td>
</tr>
</tbody>
</table>

\(^1\): Top of the test alloy, \(^2\): Top of the fuel stack, \(^3\): EFPD=Effective Full Power Days.

Three concentric regions are formed.
$\gamma \leftrightarrow \gamma + \zeta \leftrightarrow \zeta + \delta$
(center) \hspace{1cm} (periphery)

Two concentric regions are formed. ($\gamma$-phase is not observed.)
$\rightarrow$ Irradiation temperature $< 630^\circ C$
Matrix morphology is similar to that of U-Pu-Zr fuel (b). Some narrow layered phases (MA-RE inclusions) spread along grain boundaries in the $\gamma + \zeta$ zone. In low-temperature region, some dark spots (MA and RE inclusions) are visible.
Matrix morphology is similar to that of U-Pu-Zr fuel (a). Large precipitates (MA and RE inclusions) appear in γ phase zone. Some narrow layered phases (MA-RE inclusions) spread along grain boundaries in γ+ζ zone. In low-temperature region, small dark spots (MA and RE inclusions) are observed.
Characteristics of Irradiated MA-Containing Metal Fuel

1. The radial distribution of fuel matrix morphology is similar to that of U-Pu-Zr ternary fuels.

2. Some large precipitates (MA and RE inclusions) appear in the high-temperature phase.

3. In the dense matrix zone, some narrow layered phases (MA and RE inclusions) spread along grain boundaries.

4. In low-temperature region, some dark spots (MA and RE inclusions) are visible.
Mass flow of Pu and MA was analyzed for future LWR-FBR transition scenario. MA content in the FBR fuel was estimated to be 2wt%. With using 5wt% MA content fuel, MAs recycling from LWRs can be accelerated for several decades.

Relevant characteristics of U-Pu-Zr-MA-RE were examined. In the case of ≤5wt% MA and ≤5wt% RE additions,
- Am-RE-rich precipitates are dispersed almost uniformly in the alloy,
- Basic properties are practically unchanged.

MA-containing U-Pu-Zr alloys were irradiated in Phénix.
- Compositions: U-19Pu-10Zr, U-19Pu-10Zr-2MA-2RE, U-19Pu-10Zr-5MA(-5RE)
- Peak burnups: ~2.5at.%, ~7at.% and ~10at.%. 

NDT of the METAPHIX-1, -2 & -3 pins
- No critical damage had occurred during irradiation.

Metallography of METAPHIX-1
- Matrix structure is similar to that of U-Pu-Zr fuels.
- Large precipitates appear in γ-phase zone.
- Some layered phase spread along grain boundaries in γ+ζ phase region.

Quantitative analyses are being carried out.
- Fuel constituent redistribution,
- MA transmutation performance

Thank you for your attention!!
Compositions of Metal Fuel Alloys

4 types of metal fuel alloy were prepared.

Average Compositions of Fabricated Metal Fuel Alloys [wt%]

<table>
<thead>
<tr>
<th>Target</th>
<th>71U-19Pu-10Zr</th>
<th>67U-19Pu-10Zr +2MA+2RE</th>
<th>66U-19Pu-10Zr +5MA</th>
<th>61U-19Pu-10Zr +5MA+5RE</th>
</tr>
</thead>
<tbody>
<tr>
<td>U</td>
<td>71.00</td>
<td>66.85</td>
<td>66.30</td>
<td>63.50</td>
</tr>
<tr>
<td>Pu</td>
<td>18.93</td>
<td>19.80</td>
<td>19.35</td>
<td>19.75</td>
</tr>
<tr>
<td>Zr</td>
<td>10.19</td>
<td>9.46</td>
<td>8.97</td>
<td>8.19</td>
</tr>
<tr>
<td>MA</td>
<td>0.03</td>
<td>2.08</td>
<td>4.74</td>
<td>4.78</td>
</tr>
<tr>
<td>Np</td>
<td>-</td>
<td>1.23</td>
<td>2.97</td>
<td>3.04</td>
</tr>
<tr>
<td>Am</td>
<td>0.03</td>
<td>0.67</td>
<td>1.45</td>
<td>1.52</td>
</tr>
<tr>
<td>Cm</td>
<td>-</td>
<td>0.18</td>
<td>0.32</td>
<td>0.31</td>
</tr>
<tr>
<td>RE</td>
<td>-</td>
<td>1.73</td>
<td>-</td>
<td>3.40</td>
</tr>
<tr>
<td>Y</td>
<td>-</td>
<td>0.12</td>
<td>-</td>
<td>0.31</td>
</tr>
<tr>
<td>Ce</td>
<td>-</td>
<td>0.20</td>
<td>-</td>
<td>0.45</td>
</tr>
<tr>
<td>Nd</td>
<td>-</td>
<td>1.25</td>
<td>-</td>
<td>2.30</td>
</tr>
<tr>
<td>Gd</td>
<td>-</td>
<td>0.16</td>
<td>-</td>
<td>0.32</td>
</tr>
</tbody>
</table>

Impurities < 0.3wt%
Specifications of Metal Fuel Pins

Fuel pins were manufactured according to Phénix geometry.

### Fuel Pin Specifications in this Irradiation Experiments

<table>
<thead>
<tr>
<th>Specifications</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pin length [mm]</td>
<td>1,793</td>
</tr>
<tr>
<td>Outer cladding diameter [mm]</td>
<td>6.55</td>
</tr>
<tr>
<td>Cladding material</td>
<td>15-15 Ti</td>
</tr>
<tr>
<td>Fuel length [mm]</td>
<td>485</td>
</tr>
<tr>
<td>Fuel diameter [mm]</td>
<td>4.9</td>
</tr>
<tr>
<td>Initial fuel-cladding gap [mm]</td>
<td>0.375</td>
</tr>
<tr>
<td>Fuel smear density [%]</td>
<td>75.2</td>
</tr>
<tr>
<td>Sodium level above fuel* [mm]</td>
<td>~10</td>
</tr>
<tr>
<td>Plenum length [mm]</td>
<td>464</td>
</tr>
</tbody>
</table>

* : Sodium is filled into fuel-cladding gap as thermal bonding.
Axial Swelling of Fuel alloy

Fuel stack position was estimated by axial gamma-ray distribution from $^{106}$Ru.

Fuel elongation behavior is independent of MA and RE additions. Axial swelling of METAPHIX fuels is within the range of the prediction.
FP gas release fraction of MA & RE-containing fuel pins is the same level as that of U-Pu-Zr alloy fuel pins, and consistent with EBR-II ternary test fuel data.

Matrix morphology is similar to that of U-Pu-Zr fuel (b). Some narrow layered phases (MA-RE inclusions) spread along grain boundaries in $\gamma + \zeta$ zone. In low-temperature region, small dark spots (MA and RE inclusions) are dispersed.
Irradiation Behavior Analysis

-Fuel Temperature Distribution-

420°C < Temp. < 685°C

Fig. Evaluated irradiation temperature for METAPHIX-1 fuel pin at EOI (Pin No.1: U-19Pu-10Zr).
Discussion - Irradiation Temperature -

Fig. Relative γ-ray intensity emitted from $^{106}$Ru and axial power profile of Pin No. 1

Analyzed Linear Power ~275 W/cm (by $^{104}$Nd method)

- Linear Power [W/cm]
- Relative γ-ray intensity from $^{106}$Ru
- Evaluated Linear Power
- Uncertainty width of Linear Power

Axial position of fuel stack [mm]

Discussion - Irradiation Temperature -

Due to the uncertainty of linear power,

- Temperature fluctuation for each fuel rod reaches ~20°C at the fuel center,
  → Irradiation temperature at higher axial level can be lower than that at lower level,

- The highest temperature can be ~660°C.
  → High-temperature γ-phase appears at only limited fuel rods.

Fig. Evaluated irradiation temperature for METAPHIX-1 fuel pins, taking account of the uncertainties of linear power.