An Overview of CRIEPI Pyroprocessing Activities

Tadashi Inoue
CRIEPI
2-11-1, Iwato-kita, Komae-shi
Tokyo
E-mail: inouet@criepi.denken.or.jp

presented by Tadafumi Koyama, CRIEPI
**Objective of R & D on Pyroprocessing in CRIEPI**

**Objective:**

To propose effective scenario and method for SF treatment for establishment of a fuel cycle including fast reactor cycle

**Target of R & D on pyroprocessing:**

1. To establish the compact method for treating various kinds of oxides fuel

2. To establish the effective fuel cycle for fast reactor

3. To establish the technology for recovering TRUs for HLLW from aqueous reprocessing
Metal Fuel Cycle Integrated with Oxide Fuel Treatment

LWR Fuel Cycle

Metal Fuel FR Cycle

Mining

Oxide fuel fabrication

U, Pu

Pyro-reprocessing

U, Pu, MA, FP

Metal Fuel FR

U, Pu, MA

Metal Fuel Fabrication

Pyro-partitioning

(UO2 recovery), Electrochemical Reduction

MA, (U, Pu)

Spent MOX, etc.

Purex Reprocessing

LWR

U, Pu

MOX

Hlw

E

Enrichment

U

U, Pu, MA, FP

U, Pu, MA

U, Pu
Process Flow of Pyroprocessing

- Spent metal fuel
- U-Pu-Zr / FP
- Electro refining
- U, U-Pu / salt, Cd
- Cathode process
- Fuel fabrication
- Metal Fuel FR
- Fast Reactor
- Metal Fuel
- Spent Oxide fuel
- UO2 separation
- Electro reduction
- Chlorination
- denitration
- Waste treatment
- MA / FP
- Pre-treatment
- fresh MOX fuel
- U-Pu
- TRU Extract
- Fuel fabrication
- U-Zr, U-Pu-Zr
- U-Pu / FP
- U-Pu-Zr / FP
- HLW
Overview of Collaboration with domestic and overseas organizations

CRIEPI
Role: Developing process technology
Extending to engineering scale development
- Process technology by use of molten salt and liquid metal
- Installation of experimental facility/equipment by use of actinides and genuine material
- Design study of facility/equipment for commercial base

JAEA(JAERI)
Plutonium
(Electrorefining, Fuel fabrication For irradiation)
(1996 - )

JAEA(JNC)
Plutonium
(Series demonstration of process)
(2000 - )
at CPF

INL
Mass tracking in ER

Government support program (Contract basis)
- Metal fuel cycle: 2 subjects
- Electrochemical reduction of oxide fuel: 2 subjects

ITU(EU-Germany)
TRU/Genuine material
(1989 - )
Contribution to EUROPART

Kyoto University Research Reactor
Uranium
(1990 - )

Other collaborations
- Toshiba (1987 - )
- AEAT (1997-2000)

Participation to cooperation research: EUROPART
Bi-lateral information exchange agreements: BNFL, CEA
Recovery rate into Cd cathode:
1.2 t-An/y (per one electrorefiner)

Equivalent cathode current density:
125 mA/cm²

Electrorefiner for Pu separation (U,Pu:100g)

Joint study with JAEA: Basics of Electrorefining with Pu
Joint study with JAEA: Series verification of pyroprocess with Pu

Evaluation of material balance through reduction to casting

(Oxide reduction) (Electrorefiner) (Distillation) (Injection casting) (Re-oxidation)

UO₂ (U,Pu)O₂ → U-Pu

Li Reduction / Electroreduction

LiCl/LiClKCl

(650°C)

Electrorefineing (500°C)

Cd Cathode

U³⁺ & Pu³⁺

Metal fuel cycle

Distillation (500–1500°C)

Sampling (1500°C)

Oxidation (500°C)

Oxide fuel cycle

Salt/Cd

U³⁺ & Pu³⁺

Zr
Joint Study with JRC-ITU: Test with HLLW and Metal Fuels

Caisson with high purity Ar atmosphere and manipulating operating system

Dismantling and transfer to hot cell after experience with non irradiated TRU-containing material

Set up of caisson in hot cell

Cross section after anodic dissolution of U-Pu-Zr-MA

Model of anodic dissolution and anode potential change

Uranium deposit

Pu,MA,U in cadmium

Concentration of U,Pu,Zr

Slurry layer with salt and Zr

Diffusion layer in salt
Engineering Technology development: High-throughput Electrode

U collection rate based on the solid cathode ca. 8.8g-U/h/L (at unit volume of electrode)

0.73kg-U

After the improvement with clearance, scraper, etc., more than 40 g-U/h/L was achieved

Uranium metal recovered (ca. 5kg)
Engineering Technology development: Transport technology & Equipments

PYRO-STATION: Pyroprocess Scaled-up Test Apparatus for Industrialization

Salt Transport Test Rig
Metal Transport Test Rig
Electrorefiner

Salt Transport Rig
Metal Transport & Distillation Rig
Ar atmosphere Large Glove Box
The separation experiments were carried out with Ce, Gd, Y as substitutes of U, Pu(MA), REs, respectively.

The flow rates of the experiments were about 3 liters of both salt and Cd in 1-3 hr.

The concentration of each solute was found to reach steady state immediately. As expected, effective extraction were achieved at high agitation speed and low flow rate.

In the case of low flow rate experiment, more than 97% of Ce and Gd (substituted for actinides) were recovered in one stage.

Further experiments are now under way with using 3-staged extractor.
Development of Oxide Fuel Treatment: flowsheet development

Decladded spent fuel

Cs, Sr, I, etc. dissolution

Anode

~ 0.5 V

UO₂ transfer

Cathode

LiCl or LiCl/KCl

Residual oxide

~ 3 V

O₂⁻ ion transfer

Storage

~ 0.5 V

Metal reduced

Fuel Fabrication

Cathode

U³⁺, TRUⁿ⁺

In Cd

Anode

Solid

Liquid

UO₂ transfer

Cathode

LiCl or CaCl₂

Salt

Waste treatment by zeolite

Salt waste

Waste with AL, ALE

Salt

Actinide recovery by multi-stage extraction

Waste treatment by zeolite

Salt waste

LiCl or LiCl/KCl

Fuel Fabrication

9th OECD/NEA P&T IEM, Nimes, France
**Development of Oxide Fuel Treatment: experimental achievements**

**Electrowinning of UO₂ in LiCl-KCl**

**SEM image of UO₂ deposit**
*(Radial cross section)*

**Magnified**

**Electrochemical reduction**

Reduction of MOX (55U-5Np-40Pu) was confirmed with small scale.

Reduction of UO₂ in larger scale was confirmed in ~100g scale.
## Objectives of irradiation of metal fuel

**Fuel performance to be verified for commercial use**

<table>
<thead>
<tr>
<th>Item</th>
<th>Objective</th>
<th>Target value</th>
<th>Target for oxide</th>
</tr>
</thead>
<tbody>
<tr>
<td>Burn up</td>
<td>Cost-effective</td>
<td>ca. 200GWd/t</td>
<td>ca. 200GWd/t</td>
</tr>
<tr>
<td>Linear heat rate</td>
<td>Core-compactness</td>
<td>ca. 500W/cm</td>
<td>ca. 450W/cm</td>
</tr>
<tr>
<td>High temperature</td>
<td>Heat efficiency &gt;40%</td>
<td>Cladding temp. ≥650°C</td>
<td>Cladding temp. ≥675°C</td>
</tr>
<tr>
<td>MA-transmutation</td>
<td>Environmental friendly</td>
<td>Several %/year</td>
<td>70-80% compared with metal fuel</td>
</tr>
</tbody>
</table>

**Linear heat rate**
- ca. 500 W/cm
- ca. 350 W/cm

**Fabrication**
- Injection casting
- Arc-melting

**Number of pins**
- ca. 600 pins
- ca. 10 pins
- 9 pins

**Max. cladding temp.**
- Less than 600 °C
- More than 650°C

**Max. burn-up**
- ca. 200GWd/t
- ca. 200GWd/t
- ca. 100GWd/t

**Linear heat rate**
- 450 ~ 500 W/cm
- 500 W/cm
- ca. 350 W/cm

**Schedule**
- Finished( ~’95)
- Fab.:’03 ~’06
- Irrad.:’07 ~’15
- Fab. finished( ~’94)
- Irrad.:’03 ~’10

**List of irradiation test of metal fuel**

<table>
<thead>
<tr>
<th>Item of verification</th>
<th>ANL record</th>
<th>Objective</th>
<th>JOYO. planned</th>
<th>Phenix-METAPHIX-</th>
</tr>
</thead>
<tbody>
<tr>
<td>Item of verification</td>
<td>High B.U., High LHT</td>
<td>High temp. operation</td>
<td>MA transmutation</td>
<td>Phenix-METAPHIX-</td>
</tr>
<tr>
<td>Fuel composition</td>
<td>U-Pu-Zr</td>
<td>U-Pu-Zr</td>
<td>U-Pu-MA(RE)-Zr</td>
<td>Phenix-METAPHIX-</td>
</tr>
<tr>
<td>Fabrication</td>
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<td>Injection casting</td>
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<td></td>
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<td>ca. 600 pins</td>
<td>ca. 10 pins</td>
<td>9 pins</td>
<td></td>
</tr>
<tr>
<td>Max. cladding temp.</td>
<td>Less than 600 °C</td>
<td>More than 650°C</td>
<td>Less than 580°C</td>
<td></td>
</tr>
<tr>
<td>Max. burn-up</td>
<td>ca. 200GWd/t</td>
<td>ca. 200GWd/t</td>
<td>ca. 100GWd/t</td>
<td></td>
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</tr>
<tr>
<td></td>
<td></td>
<td>Irrad.:’07 ~’15</td>
<td>Irrad.:’03 ~’10</td>
<td></td>
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</table>
Phenix irradiation of MA-metal fuel - METAPHIX -

- Irradiation of metal fuel pin with U-Pu-Zr contained minor actinides (Np, Am, Cm) and rare earths
- Fuel fabrication, PIE and recycling with pyro-process in ITU

<table>
<thead>
<tr>
<th>Irradiation</th>
<th>2003</th>
<th>2004</th>
<th>2005</th>
<th>2006</th>
<th>2007</th>
<th>2008</th>
<th>2009</th>
<th>2010</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.4 at.%</td>
<td>Start</td>
<td>←</td>
<td></td>
<td>Trans. PIE</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>7.0 at.%</td>
<td>Start</td>
<td>←</td>
<td>←</td>
<td>←</td>
<td>Trans. PIE</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>11.0 at.%</td>
<td>Start</td>
<td>←</td>
<td>←</td>
<td>←</td>
<td>←</td>
<td>Trans</td>
<td>PIE</td>
<td></td>
</tr>
</tbody>
</table>

- Finished the non-destructive analysis on pins with 2.4 at.% burnup at PHENIX

\[\text{Composition of region B, wt\%} \]

- Pin 1: U-19Pu-10Zr
- Pin 2: U-19Pu-10Zr-2MA -2RE
- Pin 3: U-19Pu-10Zr-5MA -5RE

- Irradiation of 3 pins in the same capsule at different burnup

<Configuration of capsule>

Irradiation of 3 pins in the same capsule at different burnup

<Appearance after irradiation>
No failure and no visual deformation

Transport to ITU for destructive analysis etc.

Collaboration with ITU
**Metal fuel development** - Fabrication of fuel pins for JOYO irradiation -

- Installed electrorefiner for reduction and separation, Injection casting in former JAERI
- Fabricated U-Pu-Zr rod
- Fabricate 10 pins until 2007, and put into the core of JOYO at 2008, low, medium and high B.U.

**Cathode product (Cd-Pu-U)**  ⇒  **Cd distillation**  ⇒  **U-Pu ingot**

**Collaboration with JAEA (JAERI)**
Engineering scale model of injection casting for U-Zr

- 20kg/batch : ~400mm length, 50~60 slugs/batch
Future study

Outline:
- Verification of process by use of genuine material
- Development of engineering scale model
- Safeguard and material accounting
- Irradiation of metal fuel

R&D item:
- Verification of pyro-process by use of irradiated metal alloy at PHENIX
- Verification of reduction process by use of spent oxide fuel
- Development of engineering scale model of electrorefiner, TRU extractor, waste treatment & solidification
- Safeguard and material accounting measures
- Design study of reprocessing facility and economic analysis
Process Flow of Pyroprocessing of Metal Fuel Cycle

Spent metal fuel

Decladding & Chopping

(U-Pu-Zr) +MA, FP

Electrorefining

Pu, U, MA, AEM, RE

Distillation

U, Pu, MA

Fuel fabrication (Injection casting)

Fresh metal fuel

Spent salt (LiCl-KCl + U, Pu, MA, FP)

Multistage counter current extraction (Actinide recovery)

Pu, MA recovery into Cd from salt

Salt with FP

Zeolite column (FP decontamination)

Zeolite absorbed alkali and alkaline earth FP in salt

Salt waste solidification

Boric acid, Al2O3

Sodalite

Recycle

Recycled salt

Spent salt (LiCl-KCl + FP)

Zeolite + FP + LiCl-KCl

Spent U metal with salt

U metal

U, Pu, MA

Cd-Li

Cd-U, Pu, MA

Salt purified

Spent salt purified

Zeolite contained FP

Recycle

Cd-U, Pu, MA

Salt with FP

Zr

Fresh U, Zr

Quartz mold used

Fuel fabrication

Injection casting

Fresh metal fuel

Umet with salt

U metal

U, Pu, MA

Electrorefining

Distillation

Salt waste solidification

Sodalite

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