Development of Np and Am bearing MOX fuels for Japan sodium cooled fast reactors

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Background — Development of MOX containing MA —

- Japan Atomic Energy Agency has developed homogeneous mixed oxide containing minor actinides (MA-MOX) for Japan sodium-cooled fast reactors.

Subjects to be carried out
1. Database and models for properties
2. Advanced fabrication process
3. Analysis code of irradiation behaviour of low O/M MA-MOX pellet

Pellets of \((\text{Np}_{0.02}\text{Am}_{0.02}\text{Pu}_{0.3}\text{U}_{0.64})\text{O}_2\)

Specification of the fuel pellet
- Type: Hollow type
- Pu content: 20 - 30%
- MA content: ~5% (Np+Am+Cm)
- Density: 95% TD
- O/M: <1.97
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3. Analysis code of irradiation behaviour of low O/M MA-MOX pellet

**Experimental evaluation**

1. Property Measurements
   - Thermodynamic data
   - Thermal properties
   - Chemical properties
2. Pellet Fabrication Test
   - Homogenization
   - O/M adjustment Technology
   - Sintering behavior
3. Irradiation test
   - Actinide redistribution
   - Microstructure change
   - FCCI
Japan Atomic Energy Agency has developed homogeneous mixed oxide containing minor actinides (MA-MOX) for Japan sodium-cooled fast reactors.

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**Purpose of this work**

To analysis PIE results by using the measured data to develop an analysis code.
Contents

1. Irradiation Test and PIE Results
   • Irradiation condition
   • Microstructure changes
   • Actinides redistribution

2. Properties of the fuel pellets
   • Evaluation of properties
     Lattice parameter, Melting temperature
     Thermal conductivity, Oxygen potential

3. Analysis of Irradiation behaviour
   • O/M redistribution
   • Relation between Pu redistribution and vapour pressure
   • Evaluation of microstructure change
### First Test

<table>
<thead>
<tr>
<th>Pin No.</th>
<th>Name</th>
<th>Composition</th>
<th>O/M</th>
</tr>
</thead>
<tbody>
<tr>
<td>No.1-1</td>
<td>MA-MOX</td>
<td>((\text{Np}<em>{0.016}\text{Am}</em>{0.016}\text{Pu}<em>{0.3}\text{U}</em>{0.668})\text{O}_y)</td>
<td>1.98</td>
</tr>
<tr>
<td></td>
<td>MOX</td>
<td>((\text{Pu}<em>{0.3}\text{U}</em>{0.7})\text{O}_y)</td>
<td>1.98</td>
</tr>
</tbody>
</table>

Held for 10min at 430W/cm

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**Fuel Pin**

O/M=1.98

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**Irradiation pattern**

**Power rise rate:** 12MWt/h

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**Irradiation test**

**Condition 1**

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**Plutonium Fuel Development Facility**

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**Pin No.1-1**

MA-MOX

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**MOX**

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**Reactor power (MWt)**

---

**Time**
### Second Test

<table>
<thead>
<tr>
<th>Pin No.</th>
<th>Name</th>
<th>Composition</th>
<th>O/M</th>
</tr>
</thead>
<tbody>
<tr>
<td>No.2-1</td>
<td>MA-MOX (Np₀.₀₁₆Am₀.₀₁₆Pu₀.₃U₀.₆₆₈)O₀ᵣ</td>
<td>1.98</td>
<td></td>
</tr>
<tr>
<td></td>
<td>MOX (Pu₀.₃U₀.₇)O₀ᵣ</td>
<td>1.98</td>
<td></td>
</tr>
<tr>
<td>No.2-2</td>
<td>MA-MOX (Np₀.₀₁₆Am₀.₀₁₆Pu₀.₃U₀.₆₆₈)O₀ᵣ</td>
<td>1.96</td>
<td></td>
</tr>
<tr>
<td></td>
<td>MOX (Pu₀.₃U₀.₇)O₀ᵣ</td>
<td>1.96</td>
<td></td>
</tr>
</tbody>
</table>

Held for 24h at 430W/cm

**Irradiation pattern**

**MAX. Burnup 0.0082 at%**
**Observation results**

1. The heat rate of the fuels attained about 430W/cm.
2. The pellets were unmelted.
3. Pores migrated to the center.
4. Columnar grain and central void were observed.
Observation results

1. The content of Pu and Am increased at pellet center.

2. The increment of Pu and Am decreased with decreasing O/M.

3. The contents of Np and U were flat.

No.2-1 MA-MOX, O/M=1.98, at 432W/cm for 24h

No.2-2 MA-MOX, O/M=1.96, at 429W/cm for 24h
Properties of fuel pellets (1/4)

Results

1. The model to represent the lattice parameter was derived.

2. MA content slightly caused the lattice parameters to decrease.

3. The difference of theoretical density is 0.01-0.02g/cm³.

\[
a = \frac{4}{\sqrt{3}} \left( r_c \left( 1 + 0.112x \right) + r_a \right)
\]

\[
r_c = (r_U (1 - z - y' - y'') + r_{Pu} z + r_{Am} y' + r_{Np} y'')
\]

Properties of fuel pellets (2/4)

**Melting Temperature**

Results

1. The data were measured and analyzed by the ideal solid solution model.

2. MA content caused to decrease by 2-3K /%MA.

3. The solidus temperature of MA-MOX is over 3000K.
Results

1. The data were measured as functions of MA content, density, O/M and temperature.

2. MA content caused to decrease slightly in temperatures of less than 1000K.

\[ \lambda_0 = \frac{1}{(2.713x + 3.583 \times 10^{-1} \times z_1 + 6.317 \times 10^{-2} \times z_2 + 1.595 \times 10^{-2}) + (-2.625x + 2.493) \times 10^{-4} T + \frac{1.541 \times 10^{11}}{T^{5/2}} \exp\left(\frac{1.522 \times 10^{12}}{T}\right)} \]

\( z_1 \): Am content  
\( z_2 \): Np content  
\( x \): Deviation x in \( \text{MO}_2\text{x} \)

M.Kato, et al., OECD/NEA, Oct. 6-10, 2008, Japan
Results

1. The data were measured by gas equilibrium method.

2. MA content caused the oxygen potential to increase slightly.
Results
1. The data were measured by gas equilibrium method.
2. MA content caused the oxygen potential to increase slightly.

Summary
The effect of MA addition on the properties is negligibly small in the operation temperature range of FR fuels.
Analysis of irradiation behaviour
Analysis Procedure

1. Comparison with Melting temperature
   - Vapour pressure
   - Oxygen potential
2. Relation between vapour pressure and Pu redistribution
3. Evaluation of microstructure change

Input
- Vapour pressure
- Thermal conductivity
- Lattice parameter

Temperature
- O/M ratio
- Linear heat rate
- Coolant temperature

O/M redistribution
- Dimension
- Density

Results
- Temperature
- O/M ratio

Migration rate of pore

Vapour pressure
Plutonium Fuel Development Facility

Analysis Procedure

INPUT

- Temperature
- O/M redistribution

RESULTS

- Temperature
- O/M ratio

VAPOUR PRESSURE

- Vapour pressure
- Migration rate of pore

LATTICE PARAMETER

- Dimension
- Density
- O/M ratio
- Linear heat rate
- Coolant temperature

THERMAL CONDUCTIVITY

1. Comparison with Melting temperature - Vapour pressure - Oxygen potential
2. Relation between vapour pressure and Pu redistribution
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Analysis Procedure

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Input

Temperature

O/M redistribution

Results
   - Temperature
   - O/M ratio

Vapour pressure

Migration rate of pore

Dimension
Density
O/M ratio
Linear heat rate
Coolant temperature

Lattice parameter

Thermal conductivity
Plutonium Fuel Development Facility

Analysis Procedure

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Dimension
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Lattice parameter

Thermal conductivity

1. Comparison with Melting temperature

- Vapour pressure
- Oxygen potential

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**Results**

1. Both models gave almost the same result in high O/M pellets.
2. The results of low O/M pellets differed according to models.
3. Sari’s model was employed.
Evaluation of vapour pressure

Variation of vapor pressure

Variation of Pu-/U- species

Result

1. Variation of vapor pressure ratio of Pu/U-species was evaluated using Rand –Markin model.
Result

1. Calculation result shows that U content increases at pellet center.
Evaluation of Pu redistribution

Composition of the pellet: Pu/U ~ 0.3/0.7

Sari's model
- O/M=1.96
- O/M=1.98

Pu increase

U increase

Amount of Pu increase (wt%)

Pu-species / Pu-species

Expectation
Increment of Pu should be increased with decreasing the $P_{Pu\text{-species}} / P_{U\text{-species}}$. 
**Evaluation of Pu redistribution**

**Results**

1. Calculation results show that Pu content increase at pellet centre.
2. Two lines were obtained depending on O/M ratio.
Reviewing of the model for O/M redistribution

Sari’s model for O/M redistribution

\[
\ln\left(\frac{x_1}{x_2}\right) = \frac{Q^*}{R} \left( \frac{1}{T_2} - \frac{1}{T_1} \right)
\]

\[
Q^* = -9.45 \times 10^5 + 5.66 \times 10^5 V_{Pu} - 8.5 \times 10^4 V_{Pu}^2
\]

Evaluation of equation for \( Q^* \)

Calculation of O/M Redistribution, temperature and vapour pressure

Evaluation of the relationship between the \( P_{\text{Pu-species}} / P_{\text{U-species}} \) and the Pu increase

Corrected Eq.

\[
Q^* = -7.01 \times 10^6 + 6.1012 \times 10^6 V_{Pu} - 1.7705 \times 10^6 V_{Pu}^2 - 1.7122 \times 10^5 V_{Pu}^3
\]

X: deviation in \((U,Pu)O_{2-x}\)
T: temperature
R : gas constant,
\( Q^* \): the molar effective heat of transport
\( V_{Pu} \): Pu valence
Sari’s model for O/M redistribution

\[
\ln\left(\frac{x_1}{x_2}\right) = \frac{Q^*}{R} \left(\frac{1}{T_2} - \frac{1}{T_1}\right)
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\]
Calculation results in MA-MOX pellets

(a) Temperature

(b) O/M ratio

(c) Thermal conductivity

(d) Vapour pressure ratio of Pu-/U-species
Results
1. The both region grew with temperature.
2. The central void of high O/M pellet is larger than that of low O/M pellets.
1. The physical properties of the Np/Am-MOX were evaluated, and the effect of Np/Am addition was negligibly small.
2. The relation between the Pu redistribution and the vapor pressure was described by correcting Sari’s model for O/M redistribution.
3. The low O/M pellet attained higher temperature, however, the diameter of the central void was small as compared with the high O/M pellet.
4. It is needed to measure experimental data and to derive advanced models for describing irradiation behavior of low O/M fuel.
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