Development of fission release model accounting for UO₂ oxidation under air atmosphere

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In KAERI, **Nuclide management technology** (2021-) is being developed for reduction of disposal area required for spent fuel management. The technology focuses on the release of fission products in UO₂ spent fuel such as Tc, Se, Kr, Cs, I using **thermal treatment** at oxidization condition improving fission release.
However, the formation of CsTcO₄ should be avoided in thermal treatment since the compound is not easily volatilized leading to the reduced trapping efficiency of off-gas treatment process.

Therefore, thermal treatment should be divided into mid-temperature treatment mainly removing Tc, Se and Kr and high-temperature treatment removing Cs and I avoiding simultaneously release of Cs and Tc.

For the determination of such thermal treatment conditions, fission-release modeling is required, however, existing fission-release modeling cannot describe pulverization effect UO₂ → U₃O₈ in oxidizing condition.

Therefore, for the description of pulverization effect, fission-release modeling coupled to UO₂ oxidation model are developed and tested against Kr and Cs release data.
Model assumptions ::

- All fuel fragment are approximated to 1D sphere of mm size and grain to 1D sphere of μm size
- Fuel fragment is composed of equal shell representing grain and open pore at which fission products transport
- Fission products follows two stage diffusion, diffusion in solid spherical grain followed by diffusion in open pore
- UO₂ oxidation and fission product release separately occurs
Calculation Procedure

Input Conditions

- Gas composition (Inert, O\textsubscript{2} or Air)
- Size of fuel fragment and grain
- Heat treatment program

- Evaluation of average grain conversion from UO\textsubscript{2} to U\textsubscript{3}O\textsubscript{8} and oxygen concentrations

- Evaluation of FP concentration in grain/open pores and cumulative FP release amount through Finite-Volume method
### Governing equations

**UO₂ oxidation model**

**O₂ diffusion**
\[
\frac{\partial (\varepsilon_i C_{i,O_2})}{\partial t} = \frac{1}{r_i^2} \frac{\partial}{\partial r_i} \left( r_i^2 \varepsilon_i^0 D_p \frac{\partial C_{i,o_2}^*}{\partial r_i} \right) - (1 - \varepsilon_i) k(T)(1 - X_i)C_{i,O_2}^* \theta
\]

**UO₂ conversion**
\[
\frac{\partial (X_i)}{\partial t} = -3(1 - \varepsilon_i) k(T)(1 - X_i)C_{i,O_2}^* \theta
\]

**FP diffusion between open pores**
\[
\frac{\partial (\varepsilon_i C_{i,FP}^*)}{\partial t} = \frac{1}{R_i^2} \frac{\partial}{\partial R_i} \left( R_i^2 \varepsilon_i^0 D_{FP} \frac{\partial C_{i,FP}^*}{\partial R_i} \right) + (1 - \varepsilon_i) \frac{3}{a} \left( -D_s \frac{\partial C_{i,FP}^*}{\partial r} \right)_{r=a}
\]

**FP diffusion from grain to open pores**
\[
\frac{\partial (C_{i,FP}^*)}{\partial t} = \frac{1}{r_i^2} \frac{\partial}{\partial r_i} \left( r_i^2 D_s \frac{\partial C_{i,FP}^*}{\partial R_i} \right)
\]

**Solution strategy:** Finite Volume method (FVM)

**Coefficients**
\[
D_p = 0.001884 \sqrt{\frac{2(RT)^3}{\pi} \left( \frac{1}{M_1} + \frac{1}{M_2} \right) \frac{1}{N_A P \sigma_{12}^2 F}}
\]
\[
D_s = A_{FP} \exp \left( -\frac{B_{FP}}{RT} \right) (1 + CX + D_{pO_2})
\]
## Results – Tested systems

- Cumulative FP release data of AECL employed (DUPIC-AR-FT-06)

<table>
<thead>
<tr>
<th>Test</th>
<th>Previous Treatment (starting material)</th>
<th>Tested condition</th>
</tr>
</thead>
<tbody>
<tr>
<td>HOX1-HX1</td>
<td>None (UO$_2$ fuel fragment)</td>
<td>20°C → 440°C (air)</td>
</tr>
<tr>
<td>HOX1-HX2</td>
<td>HX1 (U$_3$O$_8$ powder)</td>
<td>50°C → 1320°C (air)</td>
</tr>
<tr>
<td>HOX1-HX4</td>
<td>HX1 (U$_3$O$_8$ powder)</td>
<td>125°C → 1400°C (air)</td>
</tr>
<tr>
<td>HOX1-HXA</td>
<td>None (UO$_2$ fuel fragment)</td>
<td>20°C → 400°C (Ar/4%H$_2$) → 400°C (air) → 1400°C (air)</td>
</tr>
<tr>
<td>HOX1-HR7</td>
<td>None (UO$_2$ fuel fragment)</td>
<td>40°C → 1400°C (air)</td>
</tr>
<tr>
<td>HOX1-AR1</td>
<td>None (UO$_2$ fuel fragment)</td>
<td>40°C → 400°C (air) → 1400°C (air)</td>
</tr>
<tr>
<td>HOX1-AR2</td>
<td>None (UO$_2$ fuel fragment)</td>
<td>240°C → 400°C (air) → 1400°C (air)</td>
</tr>
</tbody>
</table>
5 Results – Low Burn-up Fuel

- LWR fuel, 2.55 % enriched U-235, 28 Mwd/kgU
- FP dependent Two parameters fitted to experimental release data

〈Cesium〉
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#### Krypton

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<th>Graph</th>
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<tr>
<td>HOX1-HX1</td>
<td>Release fraction vs. time</td>
</tr>
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<td>HOX1-HXA</td>
<td>Temperature and release fraction vs. time</td>
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![Graphs of Krypton release](image)
Results – High Burn-up Fuel

- LWR fuel, 3.5 % enriched U-235, 58 Mwd/kgU

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<Krypton>
Two-stage diffusion release model coupled to UO$_2$ oxidation model was developed accounting for the effect of oxygen content and pulverization effect.

A good agreement between experimental data and calculated fission release data: Close agreement in high burn-up fuel and reasonable results for low-burn-up fuel was obtained.

The developed model is expected to estimate Cs and Krypton release behavior at given scenario, enabling the derivation of optimal thermal treatment condition for efficient fission product removal.

In subsequent study, the present modelling is being extended to the release modeling of Technetium and Iodine.
THANK YOU