UO$_2$ interactions inside canister conditions


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Previous studies (IN-CAN processes, NF-PRO): Dissolution rates of U from 0, 5, 10% $^{233}$U-doped UO$_2$ samples in synthetic groundwaters (fraction/yr)

- Reducing conditions achieved by corroding iron in solution (Ar atmosphere)
- No indication of an effect of alpha radiolysis
- The rate decreased in 1M NaCl solution
- $2 \times 10^{-8}$ to $1 \times 10^{-7}$ fraction/yr
- U concentrations close to the detection limit of the analytical method (ICP-MS)
Previous studies (REDUPP):
Dissolution rates of U from 0, 5, 10% $^{233}$U-doped UO$_2$ samples in natural groundwaters (fraction/yr)

- Dissolution rates generally higher than in synthetic groundwaters, > $10^{-7}$ fraction/yr
- Lowest dissolution rates in saline OL-KR5 groundwater at higher SA/V
- The rate for the 10% $^{233}$U-doped UO$_2$ higher than the rate for 5% $^{233}$U-doped UO$_2$ at higher SA/V in brackish groundwater, suggesting the effect of alpha radiolysis
- High Resolution ICP-MS
SEM/EDS of Fe surface (a test with 5 % $^{233}$U-doped UO$_2$, SA/V= 15 m$^{-1}$)

- black coloured precipitates on the surface of iron strips, especially in the tests with higher SA/V in natural groundwaters
- SEM/EDS analyses ⇒ U, Si
- The amounts of precipitates too small for XRD
WG4 – Spent fuel dissolution & chemistry in container: UO₂ interactions inside canister conditions

➢ To gain improved understanding of the chemical and corrosion processes and their effects on UO₂ dissolution mechanisms and rates under in-canister conditions after water intrusion

➢ The role of iron and other metal surfaces (secondary phases) and of groundwater composition (e.g. Si and sulphide)

➢ Dissolution experiments with Gd-doped and alpha-doped UO₂ in simulated canister conditions
  ▪ magnetite, cast iron, copper, H₂
  ▪ batch experiments in Ar atmosphere
  ▪ an autoclave system to maintain gas phase composition (H₂)
  ▪ tracer methods

➢ Natural groundwaters

➢ Natural groundwater with elevated Si content
## Solid UO₂ samples

<table>
<thead>
<tr>
<th>UO₂ phase</th>
<th>[²³³U] (%)</th>
<th>[²³⁵U] (%)</th>
<th>[²³⁸U] (%)</th>
<th>²³⁵U/²³⁸U</th>
<th>Samples (g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Un-doped</td>
<td>0</td>
<td>2.82</td>
<td>97.18</td>
<td>0.029</td>
<td>1, 1, 3 g</td>
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<tr>
<td>5% ²³³U-doped</td>
<td>5.0</td>
<td>4.5</td>
<td>90.5</td>
<td>0.050</td>
<td>1, 1, 3 g</td>
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<tr>
<td>15.7 MBq/g</td>
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<tr>
<td>10% ²³³U-doped</td>
<td>10.0</td>
<td>4.5</td>
<td>85.5</td>
<td>0.053</td>
<td>1, 1, 2.5 g</td>
</tr>
<tr>
<td>31.4 MBq/g</td>
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</table>

UO₂ (10 % ²³³U) ↔ alpha activity of spent fuel 3000 years after disposal
UO₂ (5 % ²³³U) ↔ alpha activity of spent fuel 10 000 years after disposal

Gd-doped UO₂, 6% Gd₂O₃ (0.71% U-235)

The availability of alpha- and Cr₂O₃/Al₂O₃ - doped UO₂ is investigated
Effect of galvanic corrosion

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- In the case of water intrusion to the inside of canister the possible galvanic corrosion may have an effect to the dissolution of UO₂
- **Galvanic corrosion** is an **electrochemical** process in which one **metal corrodes** preferentially to another when both metals are in electrical contact, in the presence of an **electrolyte**
- Galvanic corrosion can accelerate the corrosion of less noble metal parts and thus change the chemistry of the ground water
- Besides copper and cast iron also materials in fuel assembly will produce galvanic contacts with several different metal types (zircalloy, stainless steels, inconel)
Thanks for your attention!
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