The Dissolution and Transport of Radionuclides from Used Nuclear Fuel in an Underground Repository

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COMSOL CONFERENCE, BOSTON, MA  
OCTOBER 7\textsuperscript{TH} 2010

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Concept for the long term management of Canadian spent nuclear fuel

- Used fuel bundles are placed in durable containers
- Containers are emplaced within vaults excavated in a stable geological formation
- Containers are surrounded by self-sealing clay material

Goal: Develop a model to calculate the release of radionuclides from a defective container and their subsequent transport through the vault

Key aspects:
- Accurate representation of the vault geometry
- Vertical container emplacement
- Pin-hole defect in the container
- Time dependent radionuclide source term (function of the dose rate and spent fuel dissolution)
- Non-adsorbed (I-129), moderately adsorbed (Ca-41), strongly adsorbed (Cs-135) radionuclides
Model Geometry

- **Container**
  - “Empty” region representing the container walls

- **Pinhole**
  - $r = 8.25 \times 10^{-4}$ m
  - Flux measurement boundary

- **Buffer**
  - compacted bentonite

- **Backfill**
  - bentonite, clay, granite

- **Inner EDZ**

- **Outer EDZ**

- **Rock**
Governing Equations

\[ \theta_s \frac{\partial c_i}{\partial t} + \rho_b \kappa_d \frac{\partial c_i}{\partial t} + \nabla \cdot \left[ - \theta_s \tau D_o \nabla c_i \right] = R_{Li} + R_{Pi} + S_{Ci} \]

- \( \theta_s \) – porosity
- \( \kappa_d \) – sorption coefficient
- \( \rho_b \) – bulk density
- \( \tau \) – tortuosity
- \( D_o \) – free water diffusivity
- \( R_{Li}, R_{Pi} \) – liquid and solid reaction terms (radioactive decay)
- \( S_{Ci} \) – Source term
Radionuclide release

- **Instant release fraction**
  - Radionuclides present at the fuel cladding gap and in the grain boundaries
  - Released immediately upon contact with groundwater
  - Initial concentration of radionuclides in the container

- **Congruent release**
  - ~95% of radionuclides are present within the fuel grains
  - Release is dependent on the dissolution of the fuel matrix

Congruent release - Fuel Dissolution

\[ R_{uo2} = R_{\alpha} + R_{\beta} + R_{\gamma} + R_{diss} \]
\[ R_{\alpha} = A_{fuel} \cdot G_{\alpha} \cdot D_{\alpha}(t) \]
\[ R_{\beta} = A_{fuel} \cdot G_{\beta} \cdot D_{\beta}(t) \]
\[ R_{\gamma} = A_{fuel} \cdot G_{\gamma} \cdot D_{\gamma}(t) \]
\[ R_{diss} = A_{fuel} \cdot R_{UChem} \]

- \( A_{fuel} \) - Fuel surface area
- \( G_{\alpha, \beta, \gamma} \) – Empirical fuel dissolution rate constant
- \( D_{\alpha, \beta, \gamma}(t) \) – Time dependent dose rates

Congruent Release (cont’d)

$$R_i(t) = \frac{(1 - f_{ir}^i) \cdot I_{UO2}^i(t)}{I_{o,UO2}} R_{uo2}(t)$$

$$I_{UO2}^i(t) = I_{o,UO2}^i \cdot e^{(-\ln(2)/t_{1/2}^i \cdot t)} \cdot m_u$$

$$S_{ci} = \frac{R_i(t)}{V_{container}}$$

- $f_{IR}^i$ - Instant release fraction
- $I_{UO2}^i(t)$ – Inventory of radionuclide $i$ at time $t$
- $I_{o,UO2}$ – Initial inventory of UO$_2$
- $I_{o,UO2}^i$ – Initial inventory of radionuclide $i$
- $m_u$ – Mass of uranium in the container
### Boundary Conditions

<table>
<thead>
<tr>
<th>Boundary condition</th>
<th>Boundary name</th>
</tr>
</thead>
<tbody>
<tr>
<td>No Flux</td>
<td>Inner container walls</td>
</tr>
<tr>
<td>( N_i = D_i \nabla C_i = 0 )</td>
<td>Outer container walls</td>
</tr>
<tr>
<td></td>
<td>Hole walls</td>
</tr>
<tr>
<td>Continuous</td>
<td>All internal boundaries</td>
</tr>
<tr>
<td>( C_{i,1} = C_{i,2} )</td>
<td>All internal boundaries</td>
</tr>
<tr>
<td>( -n(N_{i,1} - N_{i,2}) = 0 )</td>
<td>All internal boundaries</td>
</tr>
<tr>
<td>Constant concentration</td>
<td>Outer boundaries</td>
</tr>
<tr>
<td>( \bar{C}_i = 0 )</td>
<td>Outer boundaries</td>
</tr>
</tbody>
</table>

**Diagram:**
- Outer EDZ
- Inner EDZ
- Backfill
- Defect
- Buffer
- Rock
- Container
Simulations

- **Geometry:**
  - Container and pinhole
  - Container, pinhole and buffer
  - Complete vault

- **Radionuclide source**
  - Constant concentration in the container
  - Constant fractional dissolution rate (1 x 10^{-7} a^{-1})
  - Dose dependent dissolution rate

- Compared COMSOL results to analytical calculations

- Used COMSOL vault model to verify SYVAC-CC4
Results – Dose dependent source term

- Geometry: container, pinhole, buffer
- Peak container release rate occurs $\sim 10^5$ a
- Overall strong agreement
**Results – Releases to the geosphere**

- **I-129** – no sorption
- **Ca-41** – moderate sorption
- **Cs-135** – high sorption

- **Geometry**: Complete vault
- **Cs-135** source term highest due to highest inventory and higher IRF than Ca-41
- **Sorption** causes a time delay in peak flux to the geosphere and a reduction in its magnitude compared to the source ($\kappa_d^{I} = 0$, $\kappa_d^{Ca} > \kappa_d^{Cs}$)
Verification of SYVAC-CC4 Near-Field Model

- Engineered barrier system represented by a series of concentric cylinders
- Developed for horizontal in-room container emplacement
- The vault portion of the COMSOL model was used to calibrate SYVAC-CC4 for vertical container emplacement
The buffer, backfill and EDZ layer thickness were selected so that the agreement between COMSOL and SYVAC-CC4 is strong for low and non-sorbing elements (I-129), which are the highest dose contributors.

Preferential pathway for lower sorbing elements is up through the buffer and into the tunnel. A large buffer thickness is required in SYVAC-CC4.

Preferential pathway for higher sorbing elements is through the sides of the borehole and into the rock due to the higher transport resistance in the buffer. Therefore SYVAC-CC4 underpredicts Ca-41 and Cs-135 releases from the vault.

Differences in peak fluxes of approximately 3%, 40% and 60% for I-129, Ca-41 and Cs-135 respectively.
Model Conclusions and future work

• Developed a COMSOL model to account for a dose dependent radionuclide source term, radionuclide release from a pinhole defect in a vertically emplaced container and transport through the buffer, backfill and EDZ

• Model was built in a series of increasingly complex steps

• Vault portion of the model used to calibrate SYVAC-CC4

• Future work can include examining expanding pin-hole size, multiple defective containers, advective flow and geosphere transport
Acknowledgements

• Nuclear Waste Management Organization

• MITACS Accelerate

• Dave Shoesmith Group, University of Western Ontario
Assumptions

- Water enters the container after the buffer saturates with water, which corresponds to a model time of zero (fuel age = 130 a.)
- The groundwater is reducing and neutral
- Transport is diffusion dominated
- All materials are fully saturated
- Steel canister insert and fuel cladding are not considered transport barriers
Initial Conditions – Instant release fraction

\[ C_{0,i} = \frac{f_{IR}^i \cdot I_{UO2}^i(t_f)}{V_{void}} \]

- \( V_{void} \) – Internal void volume
- \( f^{129}_{IR} = 0.04 \)
- \( f^{Cs-135}_{IR} = 0.04 \)
- \( f^{Ca-41}_{IR} = 0 \)
- All other subdomains, initial radionuclide concentration is zero
Results – constant concentration in container

<table>
<thead>
<tr>
<th>I-129 flux [mol/a]</th>
<th>No buffer</th>
<th>With buffer</th>
</tr>
</thead>
<tbody>
<tr>
<td>COMSOL</td>
<td>$1.05 \times 10^{-6}$</td>
<td>$1.95 \times 10^{-7}$</td>
</tr>
<tr>
<td>Analytical</td>
<td>$1.13 \times 10^{-6}$</td>
<td>$1.85 \times 10^{-7}$</td>
</tr>
</tbody>
</table>

- **Case without buffer:**
  - COMSOL flux is lower due to local concentration depression at the entrance of the pinhole
  - A simulation without the container yielded a flux that is exactly as predicted analytically

- **Case with buffer:**
  - COMSOL flux is higher, possibly due to the fact that the analytical solution is applicable to a semi infinite geometry whereas COMSOL uses a $C=0$ boundary condition, which would result in larger concentration gradients and higher fluxes
Results – Constant fuel dissolution rate

- Initial concentrations and final fluxes are the same
- Differences in initial flux and final concentration due to differences in resistance
- COMSOL solution is sensitive to solver tolerance
Differences in fluxes calculated at the “buffer-hole” boundary and “flux-measurement” boundary

Red: I-129 flux calculated at the buffer-hole boundary
Blue: I-129 flux calculated at the 1 mm below the buffer hole boundary (flux measurement boundary)
Green: I-129 flux calculated at the outer buffer boundary
Release Rate = \frac{C_{cont}}{R_{buffer} + R_{pinhole}}

R_{buffer} = \frac{1}{4 \cdot r \cdot \tau \cdot \theta_s \cdot D_o}

R_{pinhole} = \frac{1}{\left( \frac{\pi \cdot r^2 \cdot D_o}{L} \right)}
Radioactive Decay

\[ R_{Li} = -\theta_s \cdot \frac{\ln(2)}{t_{1/2}} \cdot c_i \]

\[ R_{Pi} = -\rho_b \cdot \kappa_d \cdot \frac{\ln(2)}{t_{1/2}} \cdot c_i \]

- \( t_{1/2}^i \) – radionuclide half-life
- \( t_{1/2}^{I-129} \): 1.57 \times 10^7 a
- \( t_{1/2}^{Ca-41} \): 1.02 \times 10^5 a
- \( t_{1/2}^{Cs-135} \): 2.30 \times 10^6 a