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

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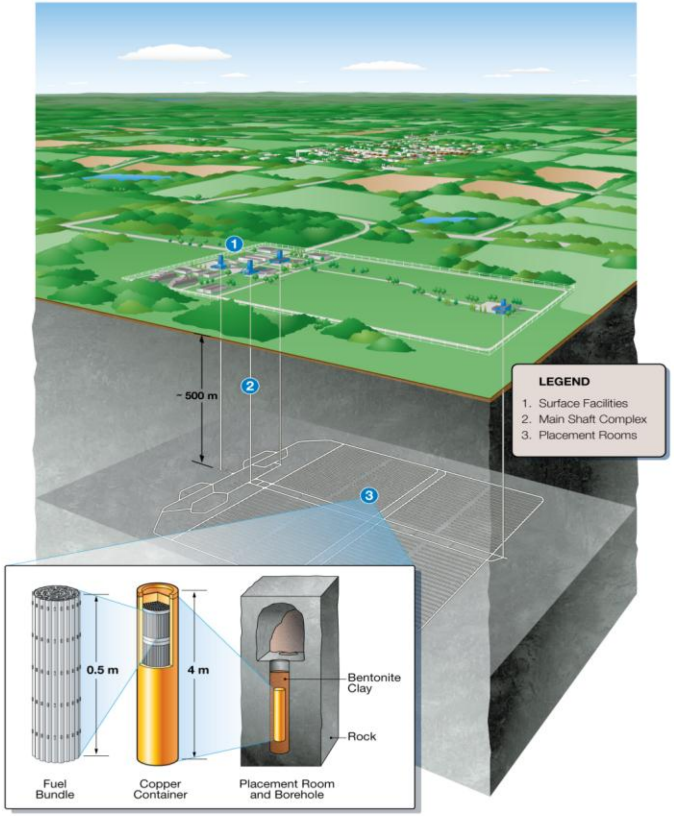
Concept for the long term management of Canadian spent nuclear fuel

- 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



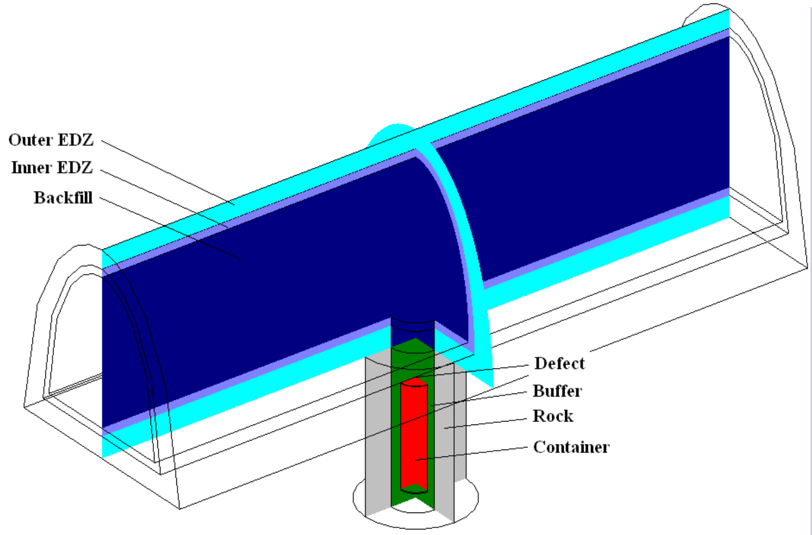
Garisto, F., T. Kempe and P. Gierszewski. Technical Summary of the Safety Aspects of the Deep Geological Repository Concept for Used Nuclear Fuel. Nuclear Waste Management Organization, report: NWMO TR-2009-12, (2009)

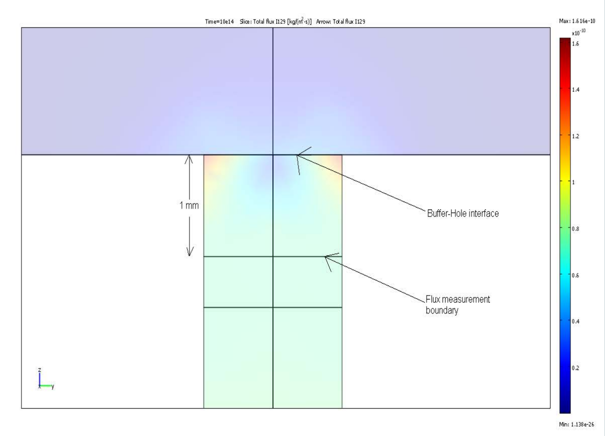
COMSOL Model

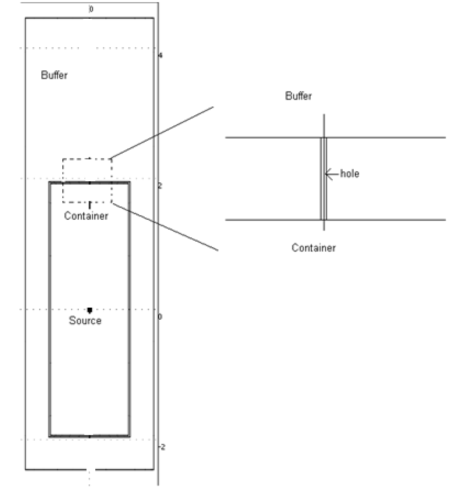
- Goal: Develop a model to calculate the release of radionuclides from a defective container and their subsequent transport through the vault
- Key aspects:
  - Accurate representation of the vault geometry
  - Vertical container emplacement
  - Pin-hole defect in the container
  - Time dependent radionuclide 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







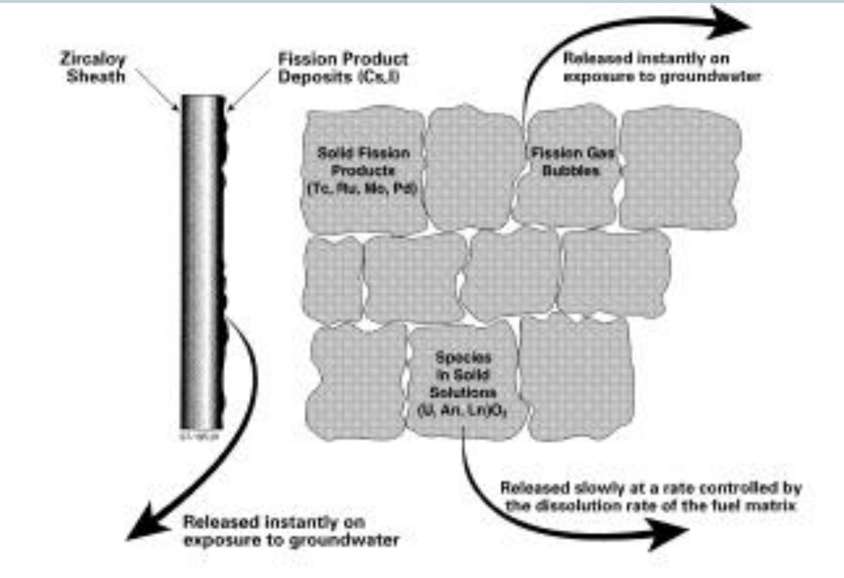
Governing Equations

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

- θ_s – porosity
- κ_d – sorption coefficient
- ρ_b – bulk density
- τ – 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



Shoesmith, D.W., Review – Fuel corrosion processes under waste disposal conditions, *Journal of Nuclear Materials*, **282,** 1-31(2000)

Congruent release - Fuel Dissolution

$$R_{uo2} = R_{\alpha} + R_{\beta} + R_{\gamma} + R_{diss}$$

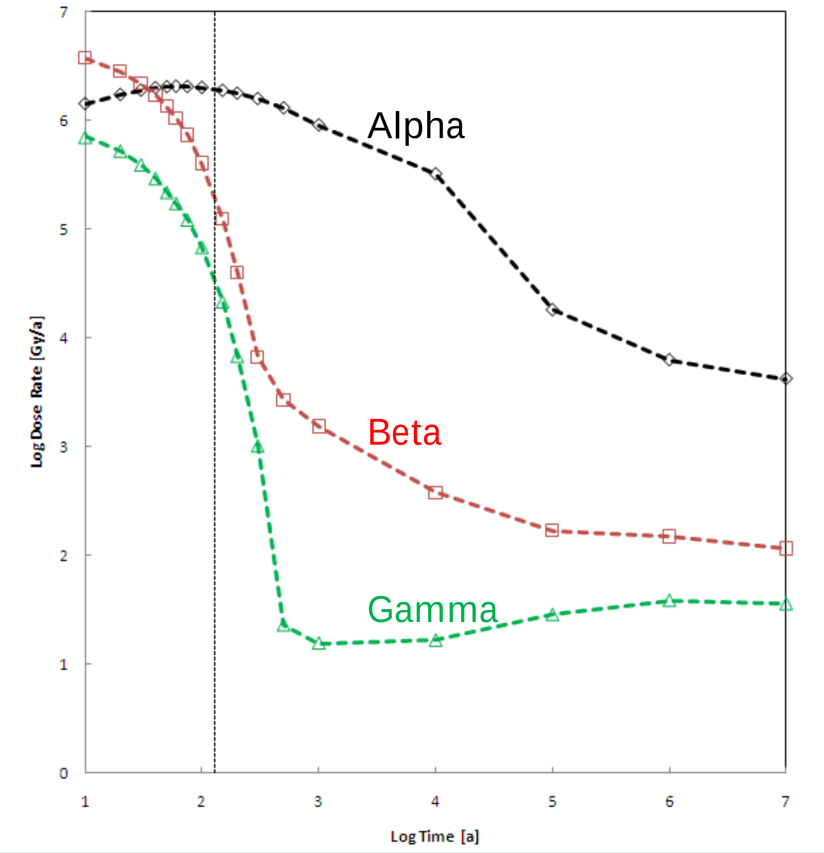
$$R_{\alpha} = A_{fuel} \cdot G_{\alpha} \cdot D_{\alpha}(t)$$

$$R_{\beta} = A_{fuel} \cdot G_{\beta} \cdot D_{\beta}(t)$$

$$R_{\gamma} = A_{fuel} \cdot G_{\gamma} \cdot D_{\gamma}(t)$$

$$R_{diss} = A_{fuel} \cdot R_{UChem}$$

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



Garisto, F., D.H. Barber, E. Chen, A. Inglot and C.A. Morrison. Alpha, Beta and Gamma Dose Rates in Water in Contact with Used CANDU Fuel. Nuclear Waste Management Organization, report: NWMO TR-2009-27, (2009)

Congruent Release (cont'd)

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

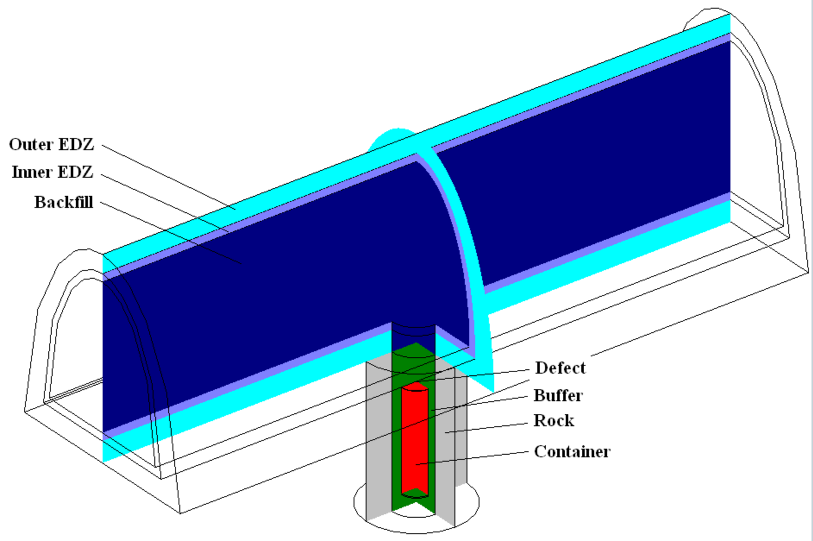
$$I_{UO2}^i(t) = I_{o,UO2}^i \cdot e^{(-\ln(2)/t_{1/2}^i \cdot t)} \cdot m_u$$

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

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

Boundary Conditions

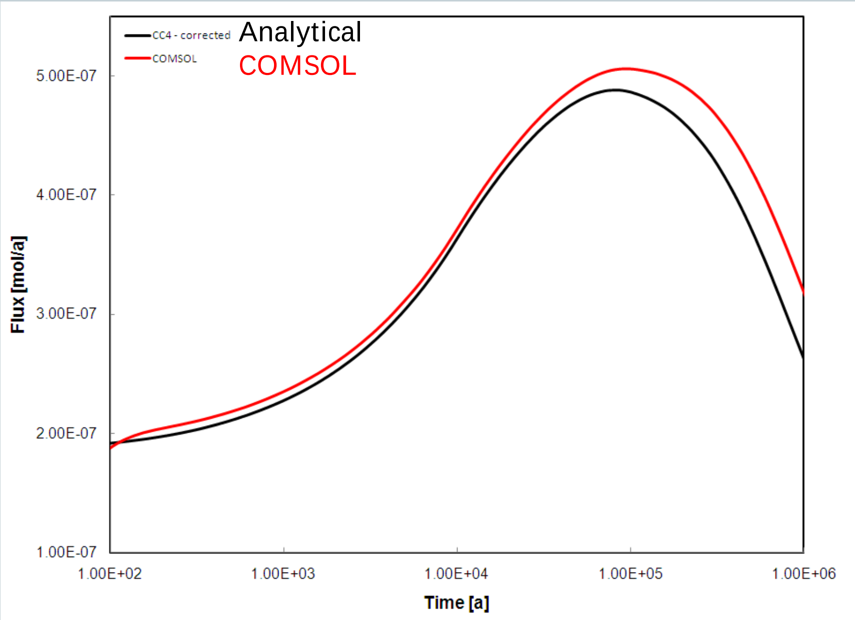
Boundary condition	**Boundary name**
No Flux $N_i = D_i \nabla C_i = 0$	Inner container walls Outer container walls Hole walls
Continuous $C_{i,1}=C_{i,2}$ $-n\left(N_{i,1} - N_{i,2}\right) = 0$	All internal boundaries
Constant concentration $C_i = 0$	Outer boundaries

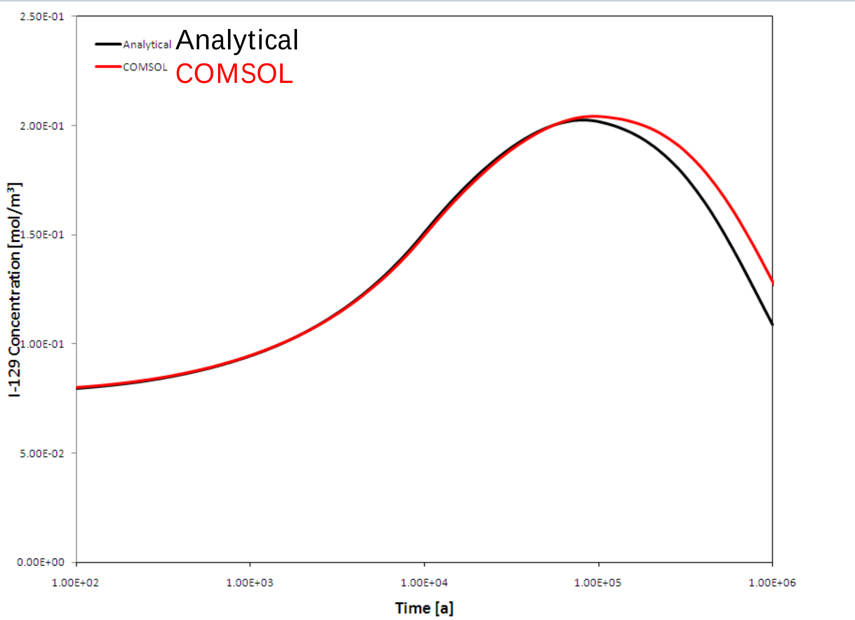


Simulations

- Geometry:
  - Container and pinhole
  - Container, pinhole and buffer
  - Complete vault
- Radionuclide source
  - Constant concentration in the container
  - Constant fractional dissolution rate (1×10^{-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

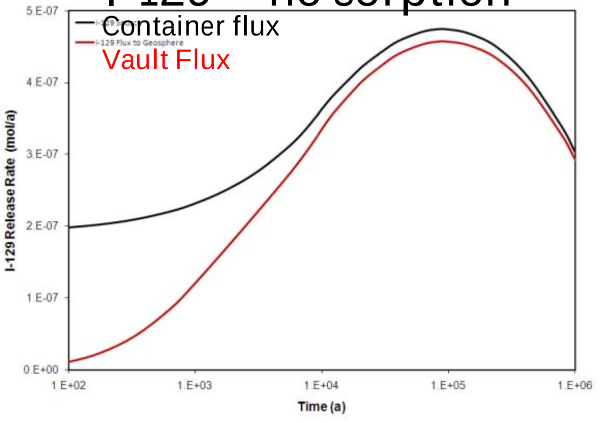




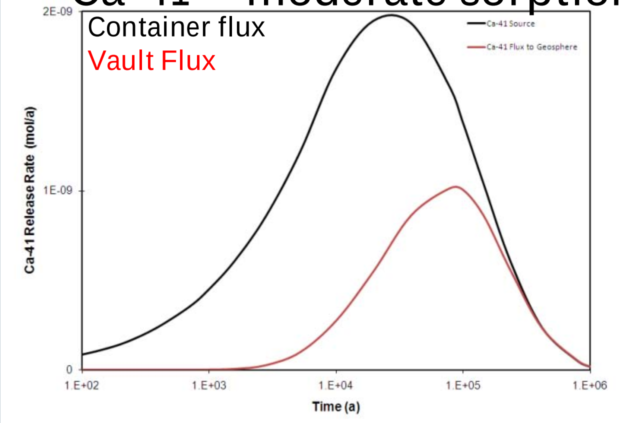
- Geometry: container, pinhole, buffer
- Peak container release rate occurs ~10^5 a
- Overall strong agreement

Results – Releases to the geosphere

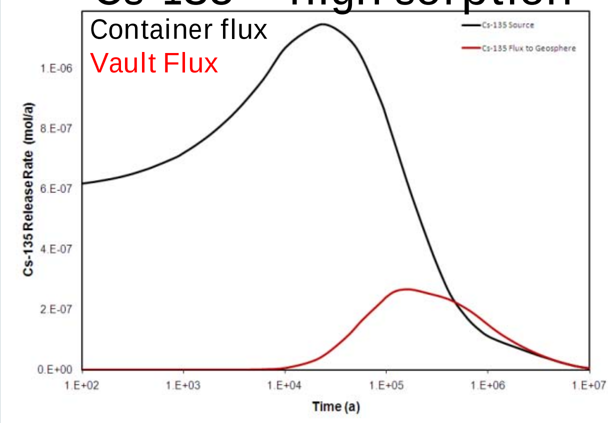
I-129 – no sorption


Container flux
Vault Flux

Ca-41 – moderate sorption


Container flux
Vault Flux

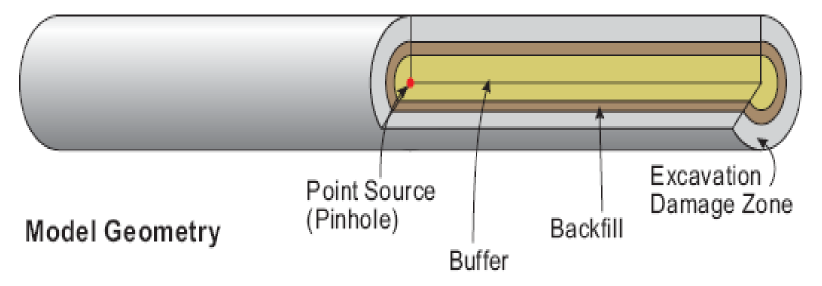
Cs-135 – high sorption


Container flux
Vault Flux

- 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 ($\kappa_d^{I} = 0$, $\kappa_d^{Ca} > \kappa_d^{Cs}$)

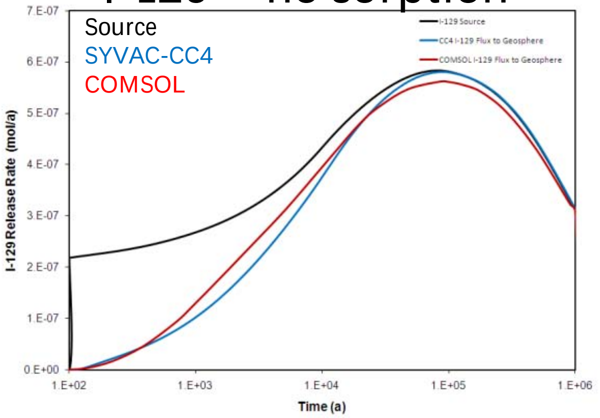
Verification of SYVAC-CC4 Near-Field Model

- 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

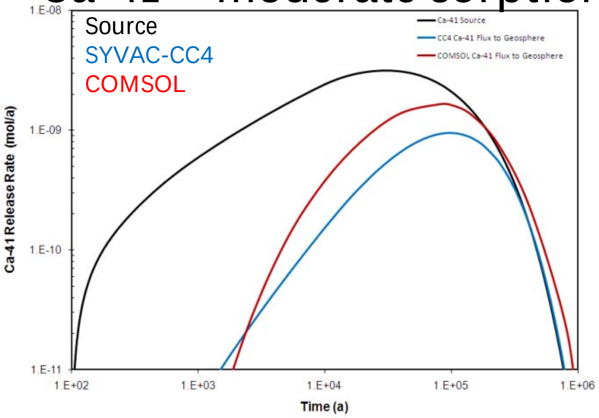


Results – Verification of SYVAC-CC4 Near Field Model

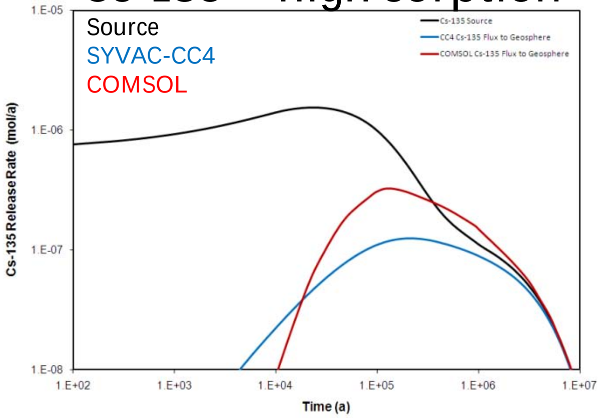
I-129 – no sorption



Ca-41 – moderate sorption



Cs-135 – high sorption



- 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 thourgh 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

nwmo

NUCLEAR WASTE MANAGEMENT ORGANIZATION

SOCIÉTÉ DE GESTION DES DÉCHETS NUCLÉAIRES

- MITACS Accelerate

MITACS

- Dave Shoesmith Group, University of Western Ontario

ShoesmithGroup

Electrochemistry and Corrosion

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

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

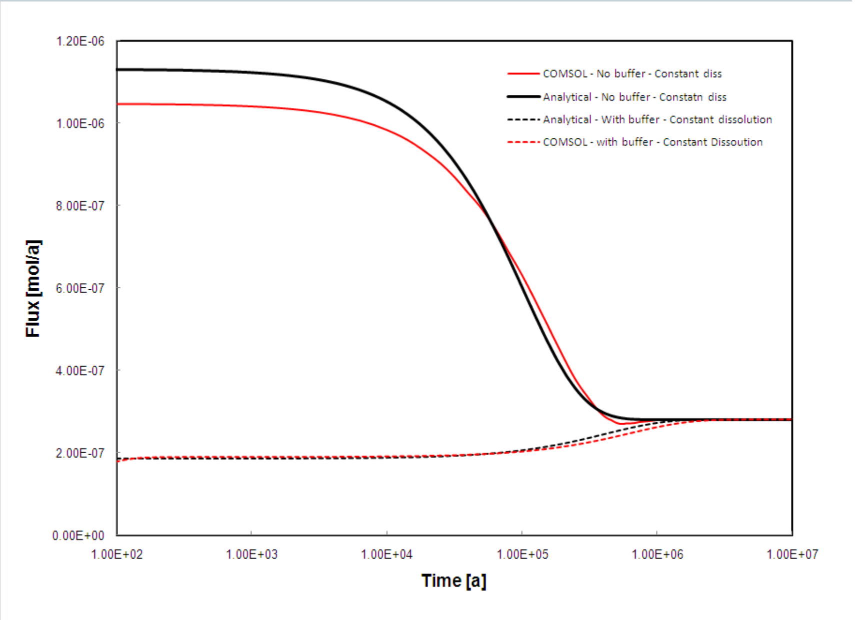
- V_{void} – Internal void volume
- $f^{I\text{-}129}_{IR}=0.04$
- $f^{Cs\text{-}135}_{IR}=0.04$
- $f^{Ca\text{-}41}_{IR}=0$
- All other subdomains, initial radionuclide concentration is zero

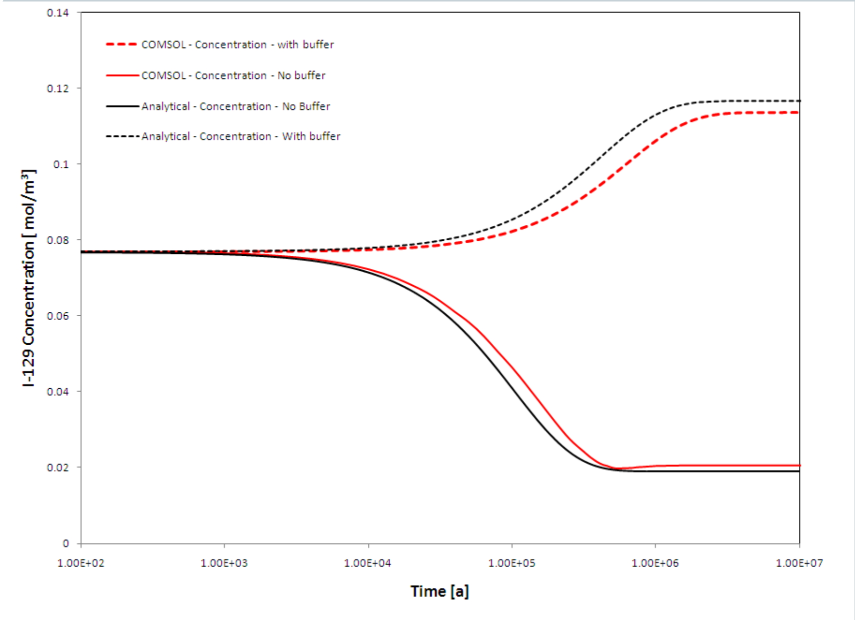
Results – constant concentration in container

	I-129 flux [mol/a]	
	No buffer	With buffer
COMSOL	1.05×10^{-6}	1.95×10^{-7}
Analytical	1.13×10^{-6}	1.85×10^{-7}

- 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

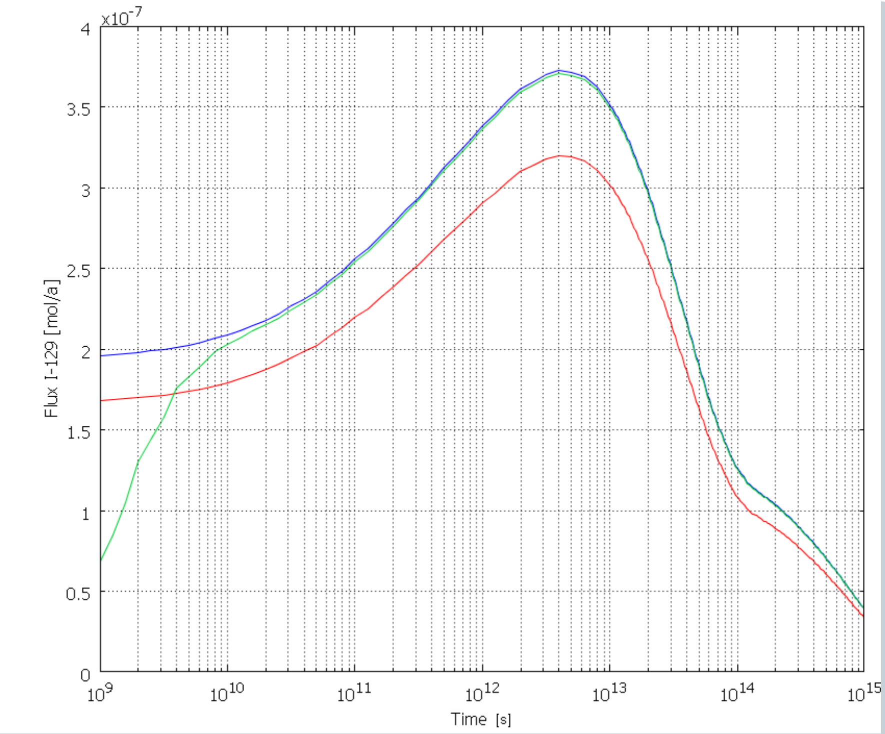
Results – Constant fuel dissolution rate

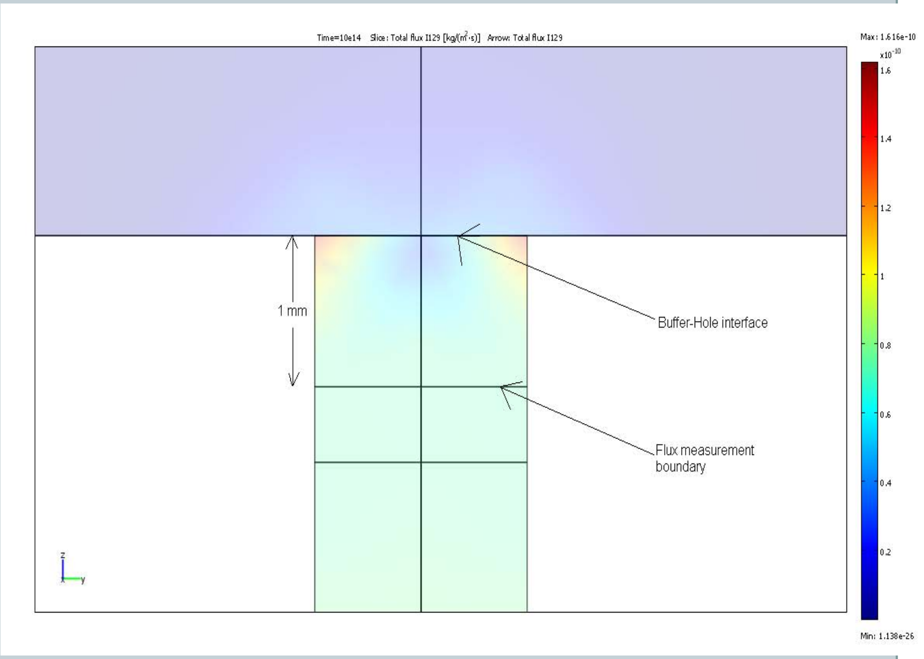




- Initial concentrations and final fluxes are the same
- Differences in initial flux and final concentration due to differences in resistance
- COMSOL solution is sensitive to solver tolerance

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

Analytical Solution

$$\text{Release Rate} = \frac{C_{cont}}{R_{buffer} + R_{pinhole}}$$

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

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

Radioactive Decay

$$R_{Li} = -\theta_s \cdot \frac{\ln(2)}{t^i_{1/2}} \cdot c_i$$

$$R_{Pi} = -\rho_b \cdot \kappa_d \cdot \frac{\ln(2)}{t^i_{1/2}} \cdot c_i$$

- $t^{i}_{1/2}$ – radionuclide half-life
- $t^{I\text{-}129}_{1/2}$: 1.57 x 10^7 a
- $t^{Ca\text{-}41}_{1/2}$: 1.02 x 10^5 a
- $t^{Cs\text{-}135}_{1/2}$: 2.30 x 10^6 a