Development of Numerical Model for Colloid-Facilitated Transport of Multiple Members of a Radionuclide Decay Chain

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Outline

1. Introduction
2. Mathematical model
3. Results and discussion
4. Conclusions
5. Future work

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Introduction

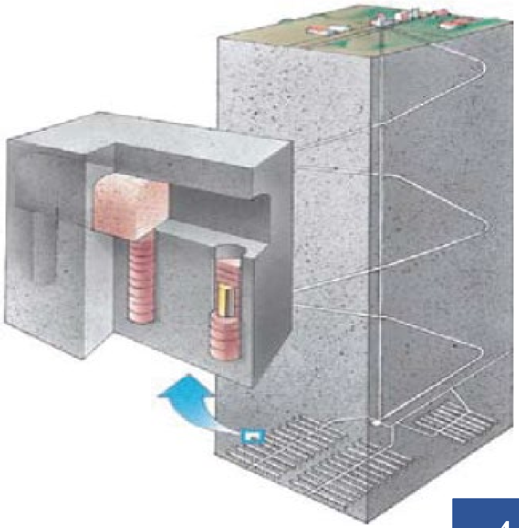
Introduction / Deep geological disposal for high-level waste

- Nuclear power generation has provided clean energy with less contaminant, but the disadvantage is that producing radioactive waste.

- Spent nuclear fuel has high level of radioactivity and a long half-life. This type of radioactive waste is called high-level waste (HLW).

- The concept of dealing with HLW is to isolate HLW from the biosphere until the concentration of HLW decays to a harmless level.

For the final disposal of HLW, the main method is to use deep geological disposal. The concept is a multi-barrier system, which is to bury HLW in a low-permeability hard rock formation 500 meters or more below the surface.

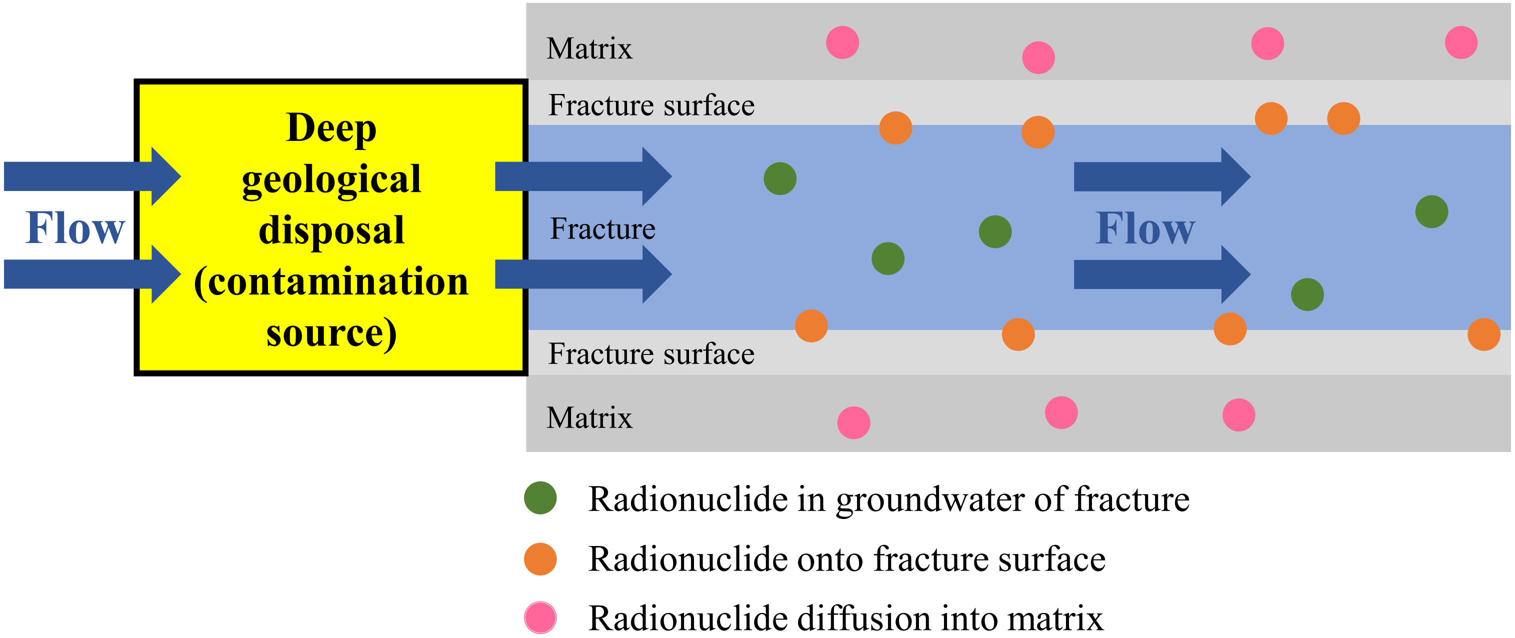


From Silveira et al., 1995

Introduction / Transport of radionuclide in the fracture-matrix system

- **The fractures may appear due to the mechanical stress created during the construction of deep geological disposal facilities and thermal and radiation effects due to the presence of HLW.**

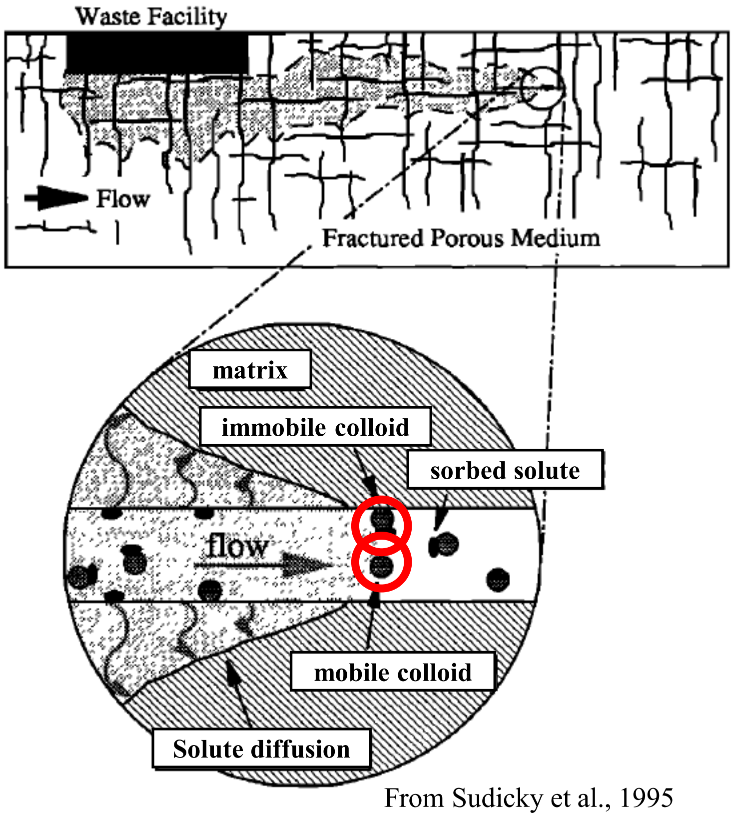
- **The mechanism by which radionuclides in a deep geological disposal could return to the biosphere is the movement of groundwater through the fractures.**



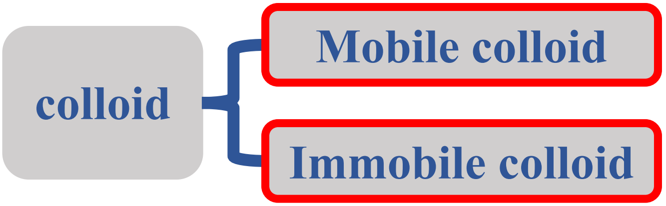
Introduction / Colloid-Facilitated the Transport of Radionuclide

- **Colloids are substances with sizes ranging from 1 nm to 10 μm**

- **Many substances can exist in colloidal form, including fine clay particles, humic substances, bacteria, radionuclides, etc.**

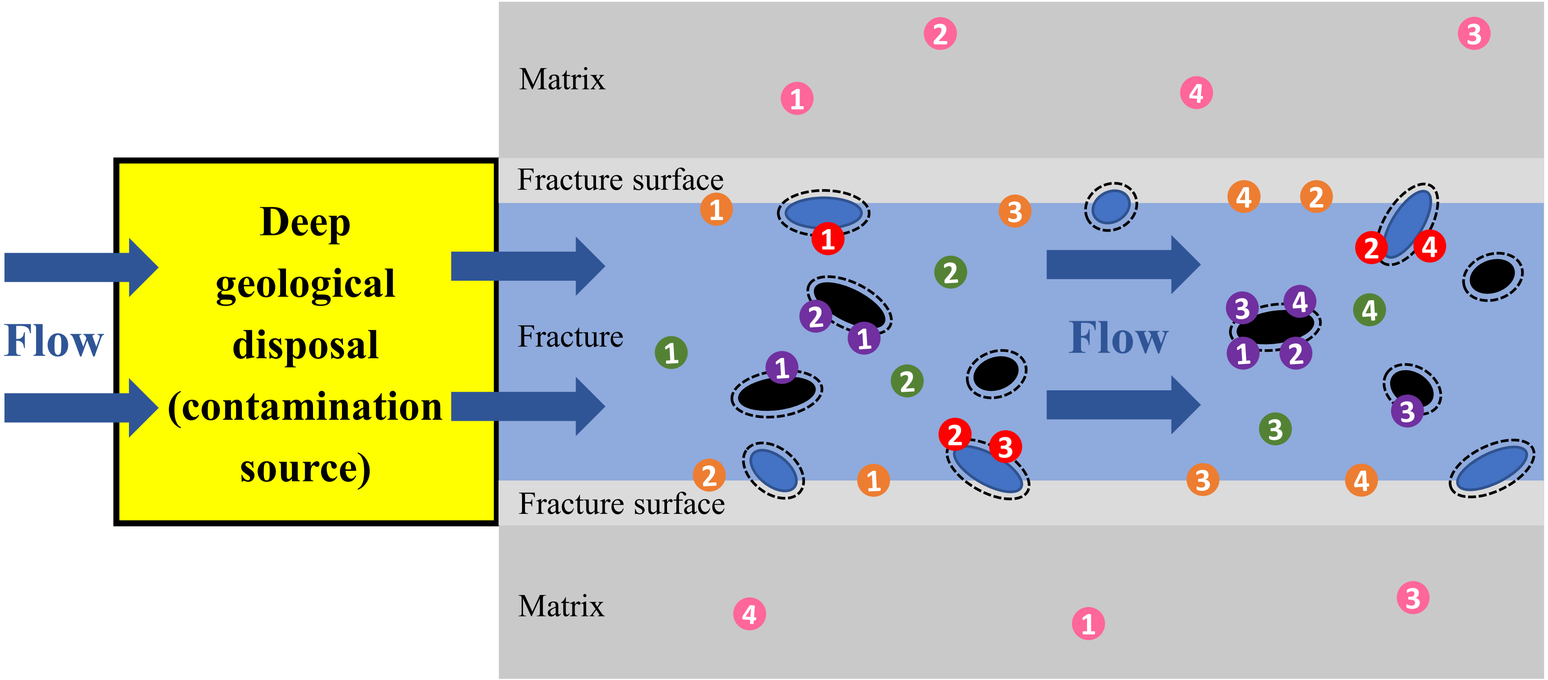


From Sudicky et al., 1995



Radionuclides are effectively prevented from diffusing into the matrix as long as it sorbed onto the colloids, and the attenuation effect of matrix diffusion is lost. Therefore, the distance that the radionuclides move will increase.

Introduction / Colloid-Facilitated the Transport of Radionuclide



Decay chain

- ① → ② → ③ → ④ Radionuclide in groundwater in the fracture
- ① → ② → ③ → ④ Radionuclide onto fracture surface
- ① → ② → ③ → ④ Radionuclide diffusion into matrix
- ① → ② → ③ → ④ Radionuclide sorption onto mobile colloid
- ① → ② → ③ → ④ Radionuclide sorption onto immobile colloid

● Mobile colloid

🔵 Immobile colloid

Introduction / Literature review

- Traditional two-phase approaches only contains an immobile solid phase and a mobile aqueous phase, which don't take into account the role of colloids, therefore have a tendency to underestimate the transport of contaminants.

- Field-scale and laboratory-scale observations have shown that colloids in groundwater can act as a third phase which can sorb contaminants, similar to the immobile solid phase. (McCarthy and Zachara, 1989)

- At a liquid waste disposal site in New Mexico, plutonium and americium should be retained in the top few millimeters of the soil profile. But in the end, it was found that it migrated over 30 m. Because the effect of colloids is ignored. (Nyhan et al., 1985)

Introduction / Literature review

	Numerical solution	Single radionuclide	Radionuclide decay chain	Colloid concentration
Baek and Pitt Jr (1996)	O	O		constant
Kheirabadi et al. (2016)	O	O		constant
Chopra et al. (2016)	O		O	constant
This study	O		O	Not constant

Ignoring the temporal and spatial variation of colloid concentration will affect the correctness of exploring the influence of colloids on the transport of multiple members of a radionuclide decay chain.

Objective

To develop a numerical model for colloid-facilitated transport of multiple members of a radionuclide decay chain which considering the change in the concentration of colloids.

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Mathematical model

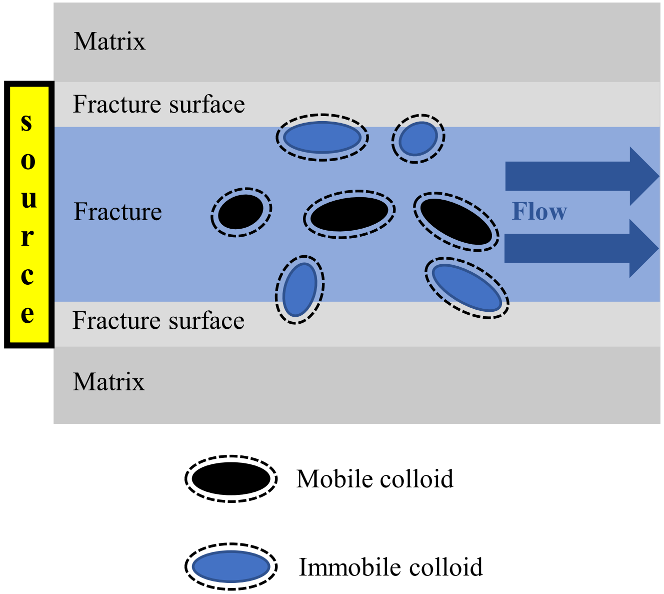
Mathematical model / Governing equation

1. Colloid transport in the fracture

$$\frac{\partial C_c(x,t)}{\partial t} = \boxed{D_f \frac{\partial^2 C_c(x,t)}{\partial x^2}} - \boxed{v_f \frac{\partial C_c(x,t)}{\partial x}} - \boxed{\frac{r_s}{b}}$$

Dispersion Advection Sorption

$C_c(x,t)$	The concentration of colloids in the water in the fracture (ML^{-3})
x	The distance along the fracture from the source (L)
t	The time (T)
D_f	The dispersion coefficient of water in the fracture (L^2T^{-1})
v_f	The velocity of water in the fracture (LT^{-1})
r_s	The rate of colloid capture on the fracture surfaces (ML^{-2})
b	The half fracture-aperture (L)



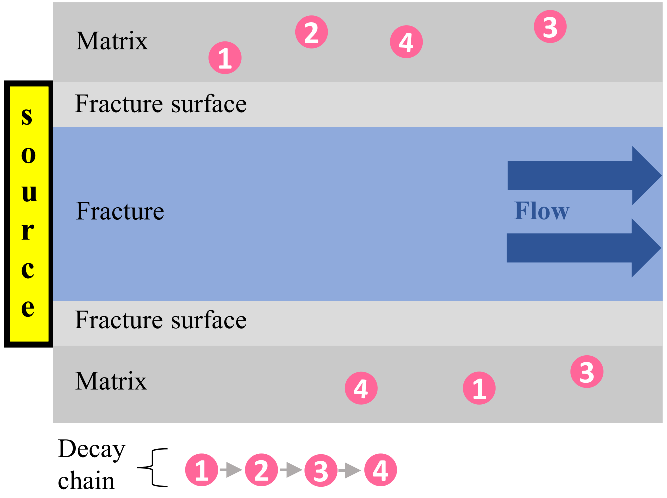
Mathematical model / Governing equation

2. Radionuclides in the rock matrix

$$\boxed{R_{m,k}\frac{\partial C_{m,k}(x,z,t)}{\partial t}} = \boxed{D_m\frac{\partial^2 C_{m,k}(x,z,t)}{\partial z^2}} - \boxed{\lambda_k R_{m,k} C_{m,k}(x,z,t) + \lambda_{k-1} R_{m,k-1} C_{m,k-1}(x,z,t)} \quad , R_{m,k} = 1 + \frac{S_m}{\phi_m} K_{m,k}$$

Retardation Dispersion First-order degradation

$C_{m,k}(x,z,t)$	The concentration of the kth radionuclide in water in rock pores (ML^{-3})
$R_{m,k}$	The retardation coefficient of the kth radionuclide in rock pores $(-)$
D_m	The diffusion coefficient in the pore water (L^2T^{-1})
λ_k	The decay constant of the kth radionuclide (T^{-1})
ϕ_m	The porosity of rock matrix $(-)$
$K_{m,k}$	The distribution coefficient of the kth radionuclide in the rock matrix (L)
S_m	The surface area per unit volume of rock matrix of pores (L^{-1})



Radionuclides diffusion into matrix

Mathematical model / Governing equation

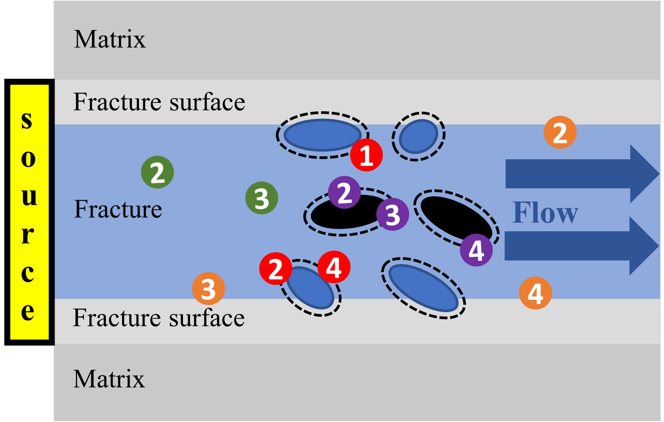
3. Colloid-facilitated radionuclides transport in the fracture

$$\frac{\partial C_{f,k}(x,t)}{\partial t} = \boxed{D_f \frac{\partial^2 C_{f,k}(x,t)}{\partial x^2}} - \boxed{v_f \frac{\partial C_{f,k}(x,t)}{\partial x}} - \boxed{\lambda_i C_{f,k}(x,t) + \lambda_{i-1} C_{f,k-1}(x,t)} - \boxed{\frac{r_{f,k}}{b} - \frac{r_{c,k}}{b} - r_{a,k} - \frac{q_k}{b}}$$

Dispersion Advection First-order degradation Sorption

4. The rate of diffusion from the fracture to the rock matrix

$$q_k(x,t) = -\phi D_m \left.\frac{\partial C_m(x,z,t)}{\partial z}\right|_{z=b}$$



$C_{f,k}(x,t)$	The concentration of the kth radionuclide in water in the fracture (ML^{-3})
D_f	The dispersion coefficient of water in the fracture (L^2T^{-1})
v_f	The velocity of water in the fracture (LT^{-1})
$r_{f,k}$	The sorption rate of the kth radionuclide from fracture water to fracture surface $(ML^{-2}T^{-1})$
$r_{c,k}$	The rate of the kth radionuclide sorption onto captured colloid surfaces $(ML^{-2}T^{-1})$
$r_{a,k}$	The rate of the kth radionuclide sorption onto mobile colloid surfaces $(ML^{-3}T^{-1})$
$q_k(x,t)$	The diffusion flux from fracture water into rock matrix of the kth radionuclide $(ML^{-2}T^{-1})$
b	The half fracture-aperture (L)

Decay chain
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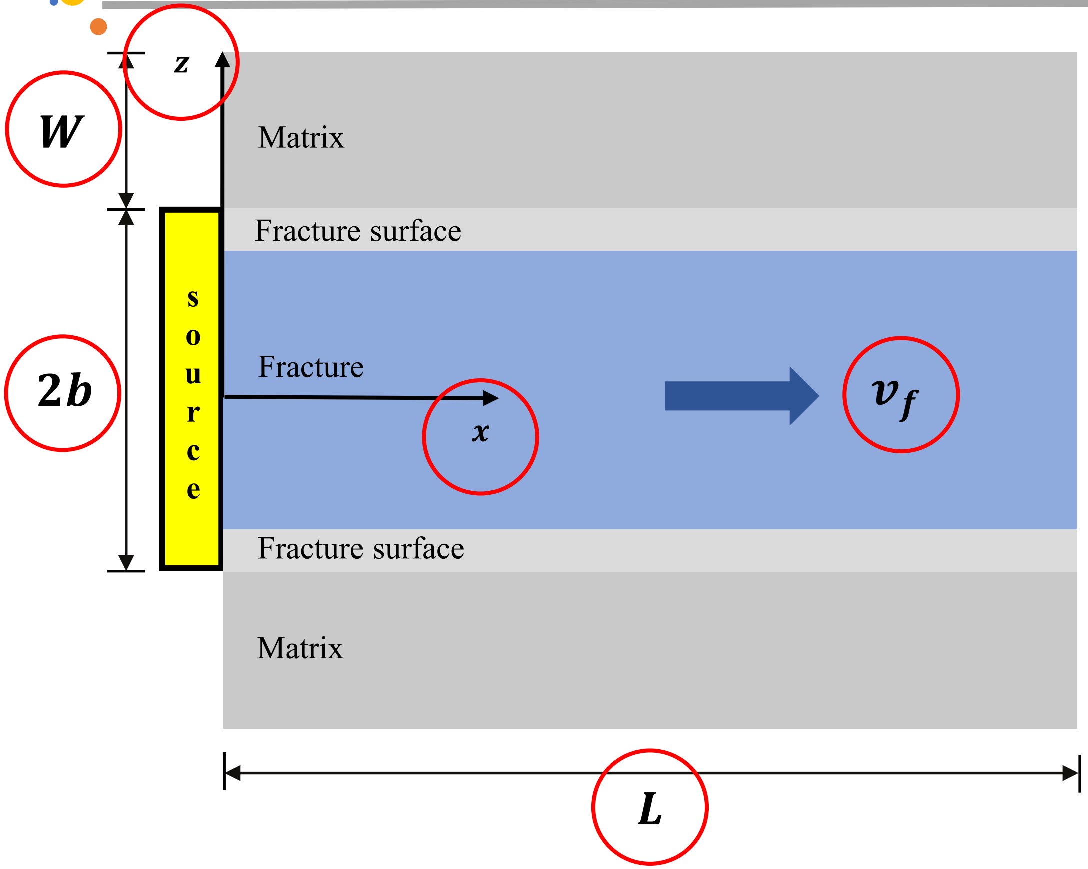
● Mobile colloid

● Immobile colloid

Mathematical model / Conceptual model



x	The distance along the fracture from the source (L)
z	The distance perpendicular to the fracture into the matrix (L)
v_f	The velocity of groundwater in the fracture (LT^{-1})
L	The length up to the fracture (L)
b	The half fracture-aperture (L)
W	The thickness of the matrix (L)

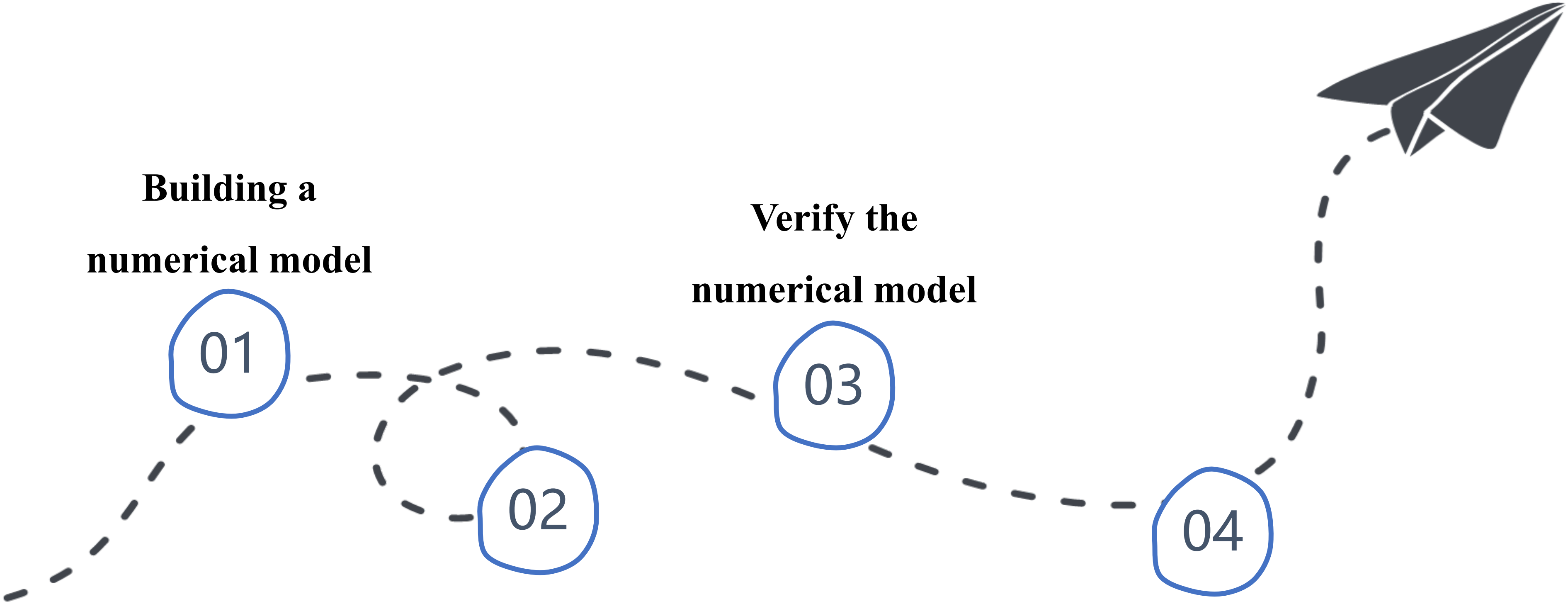
Boundary and initial conditions

$$C_{f,k}(x, t=0) = 0 \qquad C_c(x, t=0) = 0 \qquad C_{m,k}(x, z, t=0) = 0$$

$$C_{f,k}(x=0, t) = C_f e^{-\lambda t} \qquad C_c(x=0, t) = C_{c0} \qquad C_{m,k}(x, z=b, t) = C_{f,k}(x, t)$$

$$\frac{\partial C_{f,k}(x=L, t)}{\partial x} = 0 \qquad \frac{\partial C_c(x=L, t)}{\partial x} = 0 \qquad \frac{\partial C_{m,k}(x, z=w, t)}{\partial z} = 0$$

Mathematical model / Flow chart for this study



Building a numerical model

01

02

Solve governing equations using the finite difference method

Verify the numerical model

03

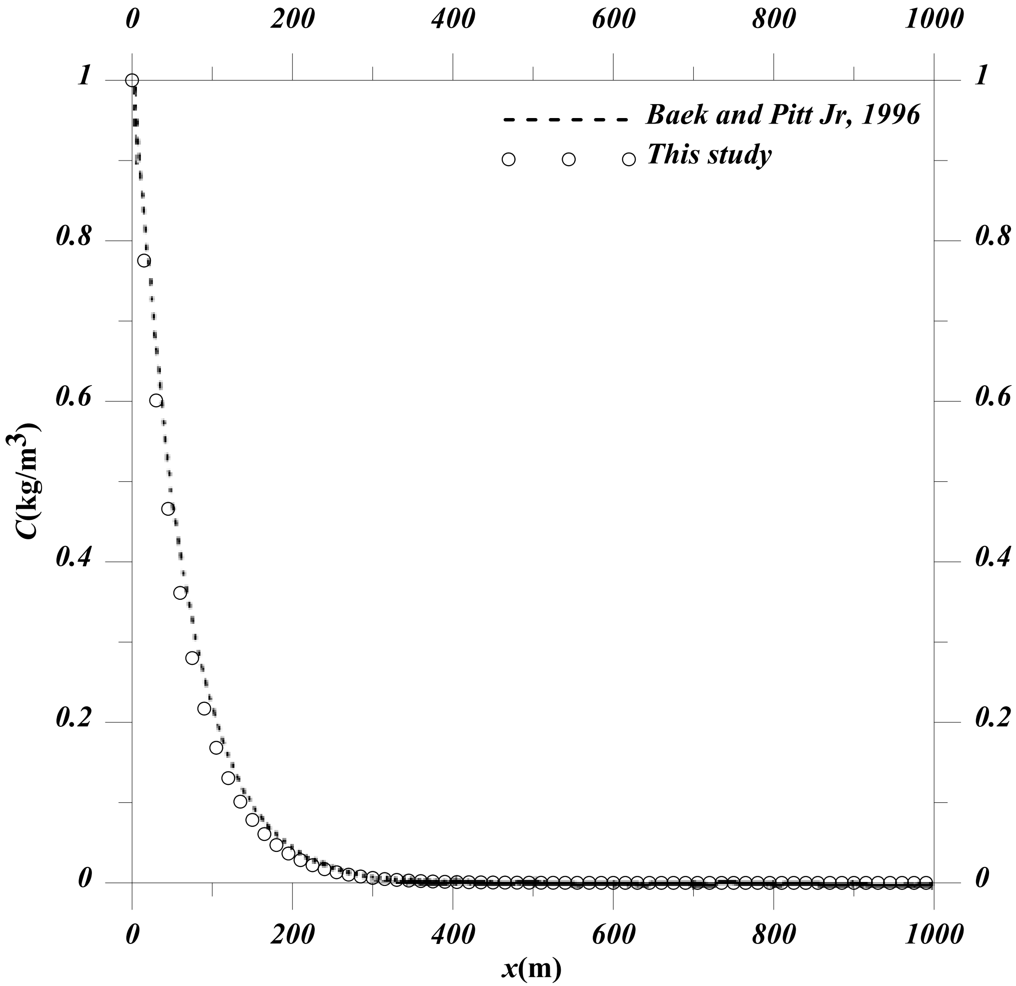
04

Sensitivity analysis for parameters

3

Results and discussion

Results and discussion / **Verification**



Parameter	Value
$v_f\ (myear^{-1})$	10
$D_f\ (m^2year^{-1})$	100
$D_m\ (m^2year^{-1})$	0.01
$R_f\ (-)$	1
$R_m\ (-)$	100
$b\ (m)$	0.005
$L\ (m)$	1000
$W\ (m)$	5
$\phi\ (-)$	0.01
$t\ (year)$	100
$\lambda\ (year^{-1})$	3.24E-7
$C_{f,0}\ (kgm^{-3})$	1

(Baek and Pitt Jr, 1996)

Results and discussion / Parameter for sensitivity analysis

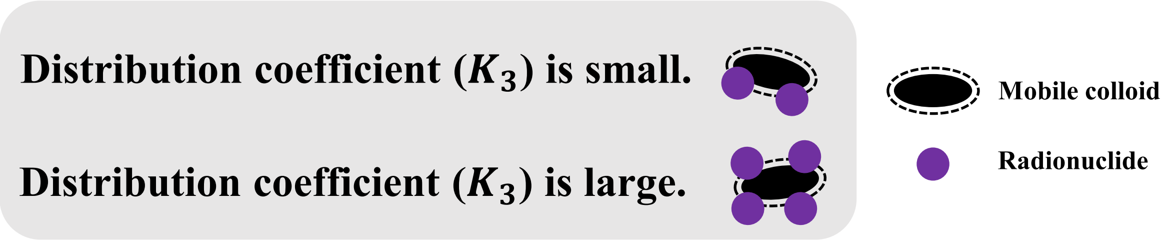
Parameter	Value	Parameter	Value	Parameter	Value
$v_f\ (m/year)$	10	$L\ (m)$	1000	$K_1\ (m)$	0
$D_f\ (m^2/year)$	100	$W\ (m)$	5	$K_2\ (m)$	0
$D_m\ (m^2/year)$	0.01	$\phi\ (-)$	0.01	$K_3\ (m^3/kg)$	100
$R_f\ (-)$	1	$t\ (year)$	100	$K_4\ (m^3/kg)$	100
$R_m\ (-)$	100	$\lambda\ (year^{-1})$	3.24E-07	$C_c\ (kg/m^3)$	0.1
$b\ (m)$	0.005	$C_{f,0}\ (kg/m^3)$	1	(Baek and Pitt Jr, 1996)	

K_1 is the distribution coefficient of radionuclides from water in fracture to fracture surface.

K_2 is the distribution coefficient of colloids with fracture surface.

K_3 is the distribution coefficient of radionuclides with mobile colloid.

K_4 is the distribution coefficient of radionuclides with immobile colloid.

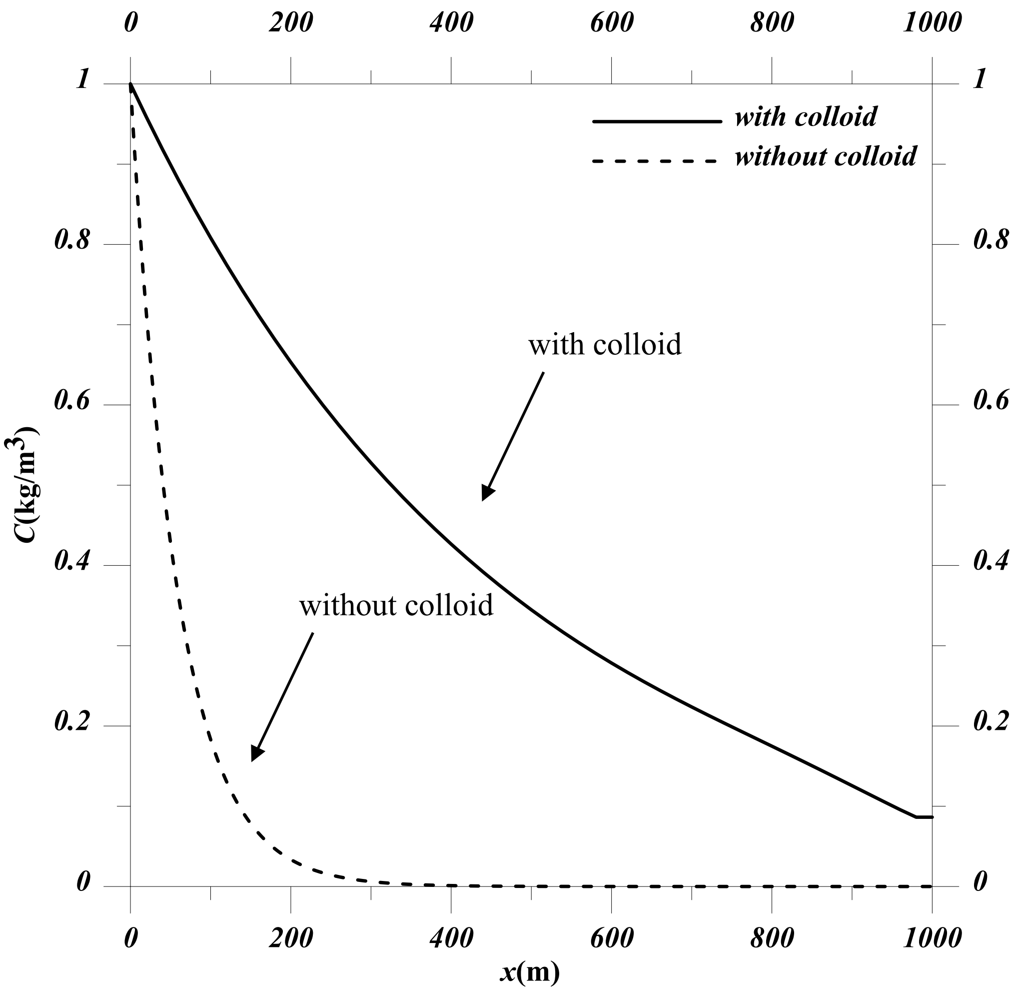


Distribution coefficient (K_3) is small.

Distribution coefficient (K_3) is large.

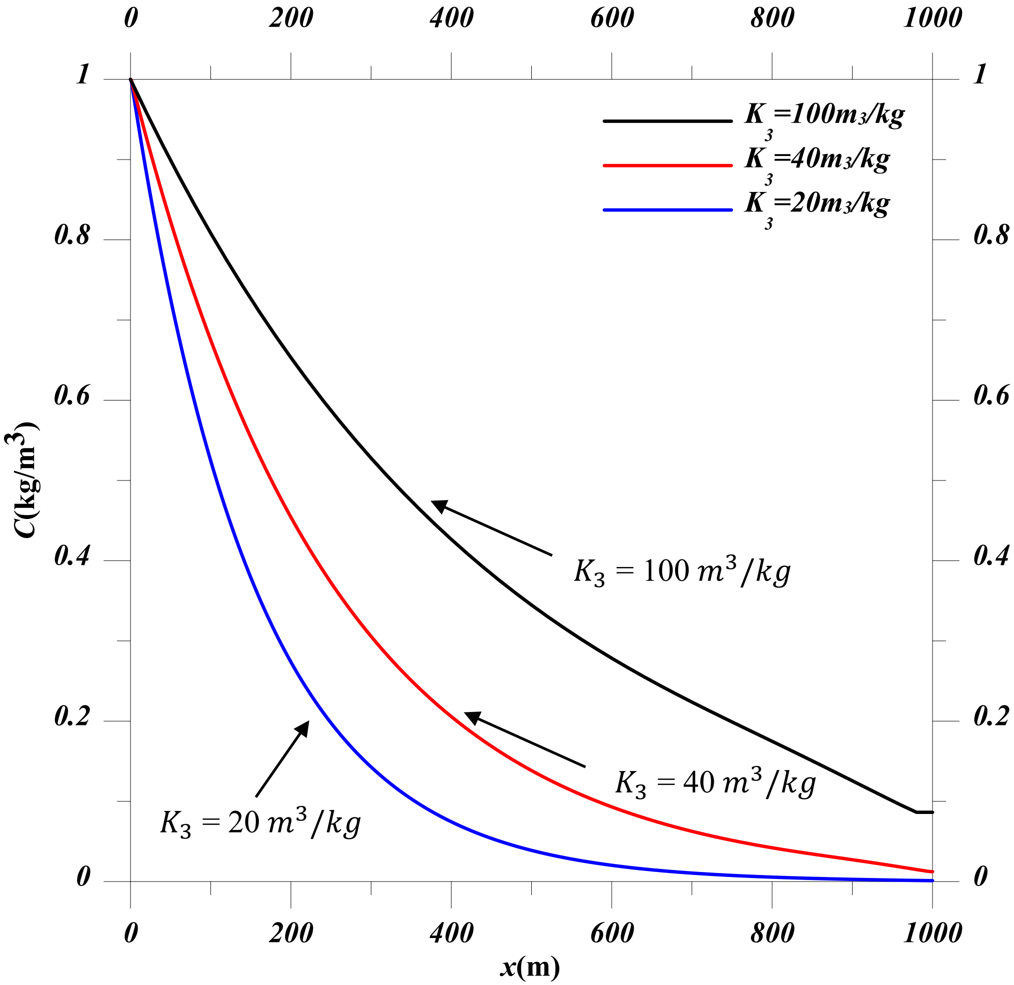
Results and discussion / **Colloid-facilitated the transport**

- ✓ **The radionuclides with colloids transport faster.**



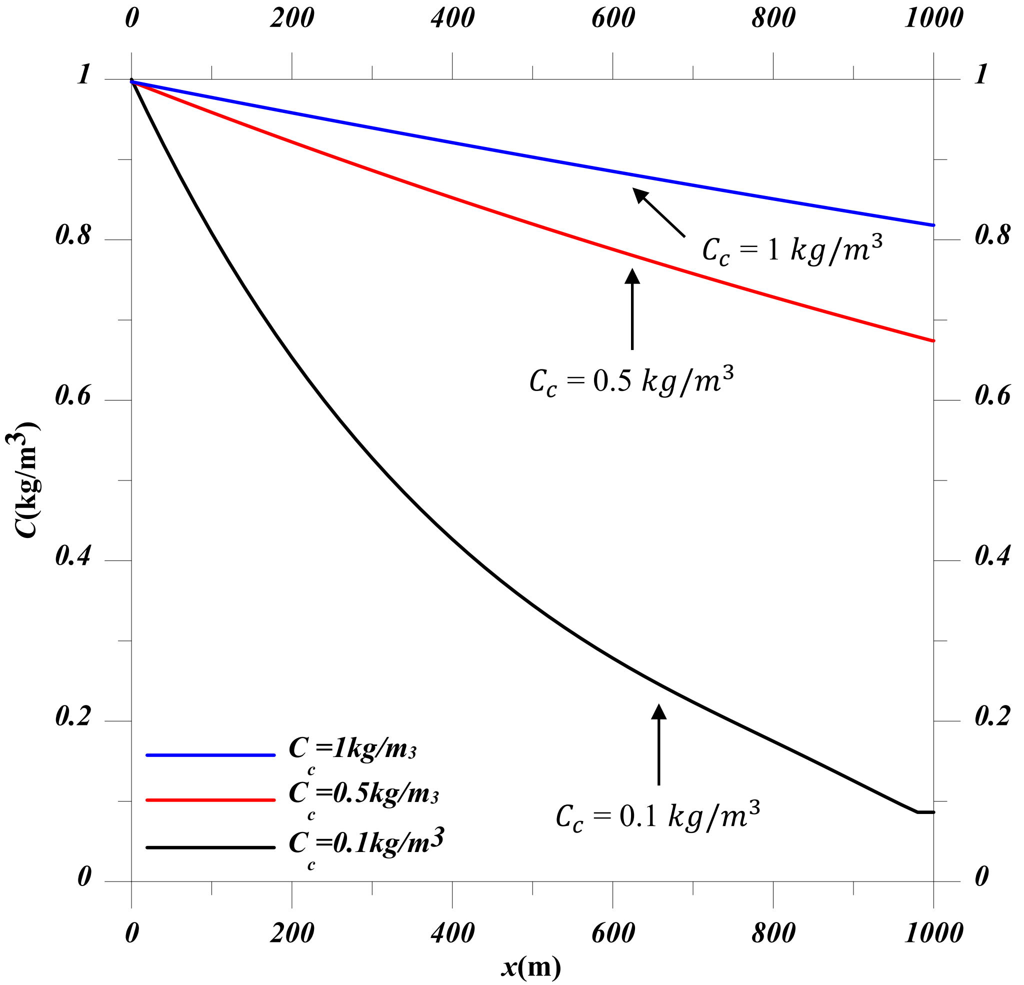
Results and discussion / Effect of distribution coefficient (K_3)

- ✓ The higher the distribution coefficient, the more radionuclides are sorbed on the colloid. So the transport of radionuclides is enhanced.



Results and discussion / Effect of colloid concentration (C_c)

- ✓ As the concentration of colloid increases, the transport of radionuclide is enhanced and its concentration within the fracture become more and more uniform.



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Conclusions

Conclusions

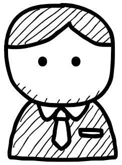
- **The presence of colloids facilitates the transport of radionuclides in the fracture.**
- **When more radionuclides are sorbed onto the colloids, the greater the transport ability.**
- **As the concentration of colloid increases, the radionuclides also transport over greater distances.**

5

Future work

Future work

- **Simulate the transport of radionuclides in the fracture under the variation of colloid concentration.**
- **Continue to simulate the transport of multiple members of a radionuclide decay chain.**



Thank you for your attention