



Spatial moment analysis of colloid facilitated radionuclide transport in a coupled fracture-matrix system

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Abstract

A numerical model is developed for studying the transport of colloid facilitated radionuclide transport in a coupled fracture-matrix system. The radionuclides and the colloids are assumed to decay, sorb on the fracture surface, as well as diffuse into the rock matrix. The sorption of the radionuclides onto the mobile and immobile colloids within the fracture is assumed to be linear. The governing equations describing the radionuclide and colloidal transport along the fracture axis and diffusive mass transport within the rock matrix, which is normal to the fracture axis are coupled together, while maintaining the continuity of fluxes at the fracture-matrix interface. Constant concentration is assumed at the inlet of the fracture for both colloids as well as radionuclides and varying grid is adopted at the fracture and rock-matrix interface to capture the flux transfer. Spatial moments are performed on the concentration profiles of the radionuclides in order to investigate the impact on the mobility and dispersion behavior of the radionuclides. Results suggest that a combination of fluid velocity and colloidal velocity, which influences the residence time of radionuclides within the fracture (against sorption onto the fracture walls, diffusion into the rock-matrix and sorption onto the colloids) ultimately decides the mobility of radionuclides. The presence of colloids retards the mobility of radionuclides in the aqueous phase. The presence of colloids have reduced the mixing of radionuclides within the fractures significantly as against the mixing experienced in a coupled fracture-matrix system in the absence of colloidal transport.

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Keywords: Fracture, Rock-matrix, Colloid, Spatial moments, Finite difference, Radionuclides.

1. Introduction

A geological repository for radioactive wastes consists of a multi – barrier system located hundreds of meters deep in the geological medium. The waste would be located in metal canisters surrounded by a barrier (e.g. compacted bentonite clay). The engineered barrier forms the near field of the repository. The nature of the components of the 'near-field' depends on the nature of the waste and the engineering aspects of the repository. The host geological formation made of hard crystalline rocks surrounding the barrier would form the 'far-field' of the repository [1]. The compacted barrier hinders or retards the migration of radionuclides in case of failure of the canister because of its high sorption capability and low permeability. However, the failure of the engineered barriers is a possible scenario [2] according to experiments in radioactive waste disposal sites [3,4]. Many studies have considered the presence of

colloids in transport of radionuclides in such hard rock fractures [5,6,7,8]. The migration of Plutonium over several kilometers from the nuclear test area in Nevada, U.S was attributed to colloid transport [9]. The performance assessment programs of Yucca Mountain project, USA [10] and H-12 of Japan [11] have considered the effects of the presence of colloids. Enhanced radionuclide transport at Chalk River Nuclear laboratories in Canada [12,13] Oak Ridge National laboratory in Tennessee [14,15] and Uranium deposit in Australia [16] were attributed with the movement of radionuclides with colloids. The rapid transport of Plutonium and Americium at experimental sites may be attributed to colloid facilitated transport [17].

Colloids are tiny particles in the size range of 1nm to 1 μm suspended in water, with high surface area and electrostatic charge [18]. The natural colloids in groundwater and the repository derived colloids influence the radionuclide migration significantly since they are smaller than the intergranular pores and fractures in rock and have the capacity to travel long distances in percolating waters [19].

Many researchers have developed models for colloid facilitated radionuclide transport in the subsurface media. Hwang et al. [20] developed mathematical model for colloid facilitated transport of radionuclides in fractured porous media for two types of colloid-radionuclide interaction, with linear adsorption processes. Grinrod [21] investigated the effect of pseudo colloids on the migration of radionuclides assuming a rapid reversible sorption between a radionuclide and both mobile and filtered colloids. In this work, the adsorption behavior was represented by equilibrium distribution coefficients and sensitivity studies were performed on partition coefficients of mobile and immobile bound radionuclides. Ibraki and Sudicky [22] developed a two dimensional numerical model that incorporates colloid facilitated transport of radionuclides in discretely fractured porous media. The model accounts for aqueous phase contaminant transport in fracture and matrix, colloid transport in fracture, and sorption of solutes. Their study includes both equilibrium and kinetic sorption. Baik and Hahn [23] developed a one dimensional analytical transport model which incorporates advective-dispersive transport, equilibrium sorption onto stationary solid medium, pseudo colloid formation, polydispersivity of colloids and radioactive decay and constant colloids concentration in the fracture. They concluded that the transport of radionuclides was accelerated by the presence of colloids. Baek and Pitt [24] numerically studied the colloid facilitated radionuclide transport in fractured porous media, considering the effects of colloid particle size. They developed a fully equilibrium model to describe the transport processes within the fracture. They concluded that the contaminant migrates to greater distances under the group of colloidal particles with smaller diameters than under that with large diameters. Li et al. [19] proposed a methodology to assess the transport within the fractured rock as well as various phenomenological coefficients employed in the different mechanisms such as filtration, remobilization, and matrix diffusion of colloids. The transport equations of colloids and radionuclides that consider the combination of aforementioned mechanisms were numerically modeled and linear sorption isotherm was considered for their numerical model. It was observed that the concentration of mobile radionuclides increased at any given time step due to high sorption partition coefficient of radionuclides associated with colloids. Tien and Li [25] developed a mathematical non linear sorption model for colloid facilitated single and two member decay chain radionuclide transport by the finite difference method. They studied the effect of parameters related to the surface forces on colloid mobility and colloid-radionuclide interactions through sensitivity analysis. The colloid concentration was assumed to constant in their model. Severino et al. [26] have quantified the potential effects of colloidal particles on the spatial development of radionuclides in a heterogeneous aquifer based on the Legrangian framework using the method of spatial moment, without the effect of pore-scale dispersion. Thus, the objective of this paper is to apply moment analysis to study the concentration profiles of the radionuclides in the coupled fracture- matrix system considering mixing characteristics as well.

2. Physical system and Governing equations

The conceptual model corresponding to a coupled fracture-matrix system is illustrated in Figure 1.

In Figure 1, b represents the half fracture aperture and H represents the thickness of the half fracture spacing. The following mechanisms are considered for the radionuclide transport coupled with colloid transport [23,24,27,19].

1. The transport of radionuclides is governed by advection, hydrodynamic dispersion and decay along the fracture axis.
2. Radioactive decay of radionuclides is considered.

3. Radionuclides sorb on the fracture walls and the mobile colloids also attach to the fracture wall surface.
 4. Radionuclides and colloids diffuse into the rock matrix from the fracture surface.
 5. Radionuclides and colloids are adsorbed by the porous rock-matrix.
 6. Colloids may be immobilized within the fracture by the filtration process.
 7. Radionuclides are adsorbed on the mobile colloids within the fracture and the rock-matrix.
 8. Radionuclides are adsorbed onto the immobile colloids deposited on the fracture surface and the rock-matrix.
 9. Sorption of radionuclide and colloids on the fracture surfaces is assumed to follow linear isotherm.
- The assumptions regarding the geometry and hydrodynamic properties are as follows:
1. The fracture aperture $2b$ is much smaller than the fracture length.
 2. The fracture walls are smooth and parallel plate assumption is considered for modeling transport in the fracture.
 3. Advection is considered to be negligible in the rock-matrix.
 4. Transverse diffusion and dispersion within the fracture assures complete mixing across fracture widths at all times.
 5. The permeability of the rock-matrix is low, and molecular diffusion is assumed to be the main transport mechanism in them.
 6. Transport along the fracture is much faster than transport in the rock-matrix.
 7. The fracture as well as the rock-matrix is saturated.

$$\frac{\partial C_m}{\partial y} = 0$$

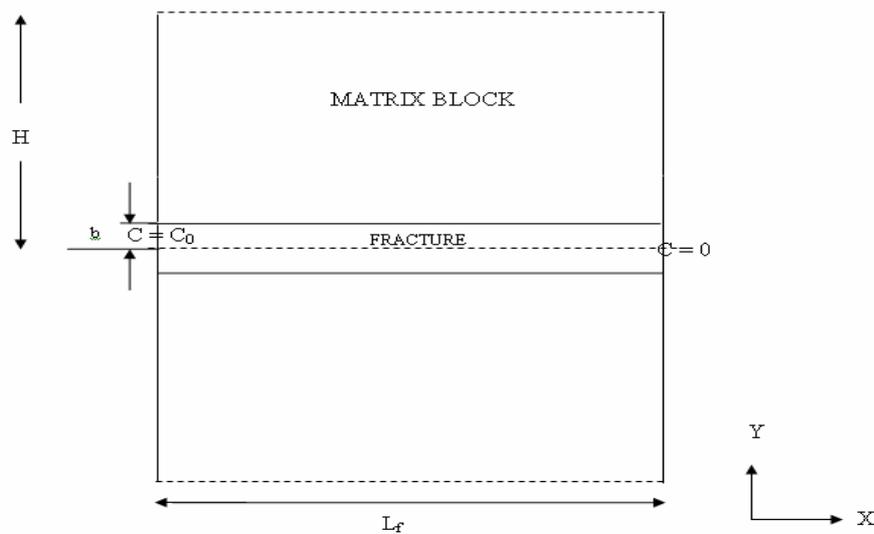


Figure 1. Schematic representation of a fracture-matrix coupled system

The governing equation for radionuclide transport in the fracture is expressed as follows [27]:

$$\frac{\partial}{\partial t} \left(N + \frac{\sigma_N}{b} + N_m + \frac{\sigma_{N_m}}{b} \right) = D \frac{\partial^2 N}{\partial x^2} + D_c \frac{\partial^2 N_m}{\partial x^2} - V \frac{\partial N}{\partial x} - V_c \frac{\partial N_m}{\partial x} + \frac{q}{b} - \lambda \left[N + \frac{\sigma_N}{b} + N_m + \frac{\sigma_{N_m}}{b} \right] \quad (1)$$

For $x \geq 0, b \geq z > 0, t \geq 0$

N = concentration of dissolved radionuclides in the fracture (kg/m^3), σ_N = concentration of radionuclides sorbed on the surface of the fracture (kg/m^3), N_m = concentration of radionuclides bounded on the mobile colloids in the fracture (kg/m^3), σ_{N_m} = concentration of radionuclides on the immobile colloids that are sorbed on the fracture surface (kg/m^3), D = hydrodynamic dispersion coefficient of the radionuclides dissolved in the fracture aqueous phase (m^2/yr), D_c = hydrodynamic dispersion coefficient of colloids suspended in the fracture (m^2/yr), V = average water velocity (m/yr), V_c = average velocity of colloids in the fracture (m/yr), b = half fracture aperture (m), q = diffusion flux from the fracture water

into the rock-matrix (kg/m² yr), x = distance along the fracture from the source (m), λ = radioactive decay for the radionuclide (l/yr).

$$q = -\theta D_p \left. \frac{\partial N_p}{\partial z} \right|_{z=b}$$

The total mobile concentration of radionuclides in the fracture can be given as

$$N_T = N + N_m \tag{2}$$

The concentration of the radionuclides in the aqueous phase and that are adsorbed can be expressed as

$$\sigma_N = K_{d_N} N \tag{3}$$

$$N_m = K_{d_m} CN \tag{4}$$

$$\sigma_{N_{im}} = K_{d_{im}} \sigma_C N \tag{5}$$

where K_{d_N} refers to the partition coefficient of the radionuclides on the fracture surface, K_{d_m} refers to the distribution coefficient of radionuclides on mobile colloids within the fracture and $K_{d_{im}}$ refers to the distribution coefficient of radionuclides on immobile colloids within the fracture.

Now, Eq. (1) can be rewritten as follows

$$\frac{\partial}{\partial t} \left[N + \frac{K_{d_N} N}{b} + K_{d_m} CN + \frac{K_{d_{im}} \sigma_C N}{b} \right] = D \frac{\partial^2 N}{\partial x^2} + K_{d_m} D_C \frac{\partial^2 CN}{\partial x^2} - V \frac{\partial N}{\partial x} - V_C \frac{\partial K_{d_m} CN}{\partial x} - \frac{q}{b} - \lambda \left[N + \frac{K_{d_N} N}{b} + K_{d_m} CN + \frac{K_{d_{im}} \sigma_C N}{b} \right] \tag{6}$$

The equations for radionuclide transport in the rock-matrix can be expressed as

$$\frac{\partial}{\partial t} (N_p + \sigma_{NP} + N_{P_m} + \sigma_{NP_{im}}) = D_p \frac{\partial^2 N_p}{\partial z^2} + D_{CP} \frac{\partial^2 N_{P_m}}{\partial z^2} - \lambda (N_p + \sigma_{NP} + N_{P_m} + \sigma_{NP_{im}}) \tag{7}$$

For $z \geq b, t \geq 0$

N_p = concentration of dissolved radionuclides in the porous rock-matrix (kg/m³), σ_{NP} = concentration of radionuclides sorbed on the rock-matrix (kg/m³), N_{P_m} = concentration of radionuclides sorbed on the mobile colloids in the porous rock-matrix (kg/m³), $\sigma_{NP_{im}}$ = concentration of radionuclides sorbed on the immobile colloids in rock-matrix (kg/m³), D_p = dispersion coefficient of the radionuclides in the rock-matrix, D_{CP} = dispersion coefficient of the colloids in the rock-matrix, λ = radioactive decay for the radionuclide (l/yr), z = cross fracture distance from the centre of the fracture axis (m), t = time (yr)

The adsorption of radionuclides in the rock-matrix is assumed to be a reversible reaction with constant distribution coefficients. Thus the transport equation for radionuclides can be rewritten based on the following equations

$$\sigma_{NP} = K_{d_{NP}} N_p \tag{8}$$

$$N_{P_m} = K_{d_{mP}} C_P N_p \tag{9}$$

$$\sigma_{NP_{im}} = K_{d_{imP}} \sigma_{CP} N_p \tag{10}$$

$$\sigma_{NP_{im}} = K_{d_{imP}} K_{d_{CP}} C_P N_p$$

$K_{d_{NP}}$ is the distribution coefficient for the radionuclides on the rock-matrix, $K_{d_{mP}}$ and $K_{d_{imP}}$ are the distribution coefficients for the radionuclides on the mobile and immobile colloids in the rock-matrix, and σ_{CP} is the concentration of the immobile colloids in the rock-matrix that can be modeled by the distribution coefficient $K_{d_{CP}}$. Thus the governing equation for the radionuclide transport in the presence of colloids in the rock-matrix can be expressed as

$$(1 + K_{d_{NP}}) \frac{\partial N_p}{\partial t} + (K_{d_{mP}} + K_{d_{CP}} K_{d_{imP}}) \frac{\partial C_P N_p}{\partial t} = D_p \frac{\partial^2 N_p}{\partial z^2} + D_{CP} K_{d_{mP}} \frac{\partial^2 C_P N_p}{\partial z^2} - \lambda (1 + K_{d_{NP}} + K_{d_{mP}} C_P + K_{d_{CP}} K_{d_{imP}} C_P) N_p \tag{11}$$

The equations (6) and (11) can be further simplified and written as:

In the fracture,

$$R^* \frac{\partial N}{\partial t} = D^* \frac{\partial^2 N}{\partial x^2} - V^* \frac{\partial N}{\partial x} - \lambda R^* N + \frac{\theta D_p^{**}}{b} \frac{\partial N_p}{\partial z} \Big|_{z=b} \quad (12)$$

$$R^* = 1 + \frac{K_{d_N} N}{b} + K_{d_m} C + \frac{K_{d_{im}} \sigma_c}{b} \quad (13)$$

$$D^* = D + K_{d_m} D_C C \quad (14)$$

$$D_p^{**} = D_p + \varepsilon D_{CP} K_{d_{mp}} C_p \quad (15)$$

$$V^* = V + K_{d_m} V_C C - 2D_C K_{d_m} \frac{\partial C}{\partial x} \quad (16)$$

In the porous rock-matrix,

$$R_p^* \frac{\partial N_p}{\partial t} = D_p^* \frac{\partial^2 N_p}{\partial z^2} - V_p^* \frac{\partial N_p}{\partial z} - \lambda R_p^* N_p \quad (17)$$

where,

$$R_p^* = 1 + K_{d_{NP}} + K_{d_{mp}} C_p + K_{d_{CP}} K_{d_{mp}} C_p \quad (18)$$

$$D_p^* = D_p + D_{CP} K_{d_{mp}} C_p \quad (19)$$

$$V_p^* = -2D_{CP} K_{d_{mp}} \frac{\partial C_p}{\partial z} \quad (20)$$

Initial and boundary conditions for the governing equations (12) coupled with (17) are given below.

$$N(x=0,t)=N_0 \quad (21)$$

$$N(x=L,t)=0 \quad (22)$$

$$N(x,t=0)=0 \quad (23)$$

$$N_p(x,z=b,t)=N(x,t) \quad (24)$$

$$N_p(x,z=H,t)=0 \quad (25)$$

$$N_p(x,z \geq b,t=0)=0 \quad (26)$$

where L is the length of the fracture and N_0 is the initial concentration of radionuclide in the inlet of the fracture.

It can be observed in equations (6) and (11) that the colloid concentration in the fracture (C) and the rock-matrix (C_p) is a function of space and time. The governing equation for the colloid transport in the fracture is expressed as:

$$\frac{\partial}{\partial t} \left(C + \frac{\sigma_c}{b} \right) + V_C \frac{\partial C}{\partial x} - D_C \frac{\partial^2 C}{\partial x^2} + \frac{Q_C}{b} = 0 \quad (27)$$

$$\frac{\partial \sigma_c}{\partial t} = \lambda_f V_C C b \quad (28)$$

Q_C is the diffusion flux of the colloids from the fracture into the rock-matrix and

$$Q_C = -\varepsilon \theta \frac{\partial C_p}{\partial z} \Big|_{z=b} \quad (29)$$

ε is the percentage of matrix flux diffusion into the rock-matrix since the diffusion of colloids may be hindered by the colloids filtered on the fracture surface and some colloids with diameters larger than the pores in the rock-matrix cannot diffuse into the rock-matrix.

The governing equation for the colloid transport in the rock-matrix is expressed as:

$$(1 + K_{d_{CP}}) \frac{\partial C_p}{\partial t} - D_{CP} \frac{\partial^2 C_p}{\partial z^2} = 0 \quad (30)$$

$$x > 0, z \geq b, t \geq 0$$

where C_p is the concentration of the colloids in the porous fracture-skin, D_{CP} is the dispersion coefficient of the colloids and $K_{d_{CP}}$ is the sorption partition coefficient for the colloids within the rock-matrix.

The initial and boundary conditions for colloid transport are given as:

$$C(x,t=0)=0 \quad (31)$$

$$C(x=0,t)=C_0 \quad (32)$$

$$C(x=L,t)=0 \tag{33}$$

$$C_p(x, z \geq b, t = 0) = 0 \tag{34}$$

$$C_p(x, z = b, t) = C(x, t) \tag{35}$$

$$C_p(x, z = H, t) = 0 \tag{36}$$

where C_0 is the concentration of the colloids at fracture inlet

3. Numerical solution

In this study, the system is described by a set of two partial differential equations for colloids and radionuclides; one for the fracture, and another for the rock-matrix, formulated for a one-dimensional framework. The continuity at the fracture-matrix interface is attained by iterating the solution at each time step. The coupled system is solved numerically using an implicit finite difference scheme. A varying cell width is adopted in the rock-matrix. A smaller grid size is adopted in the fracture-matrix interface to accurately capture the flux transfer at the fracture-matrix interface. The last term on the right hand side of equation (27) is discretized as

$$\frac{\partial C_p}{\partial z} = \frac{C_p^{n+1}{}_2 - C_p^{n+1}{}_1}{\Delta z(1)} \tag{37}$$

where $\Delta z(1)$ represents the cell width across the fracture-matrix interface. Here the colloid concentration in the first node in the rock-matrix, i.e., $C_p^{n+1}{}_1$ will be equal to the corresponding fracture colloid concentration $C^{n+1}{}_1$ perpendicular to the rock-matrix satisfying the boundary condition.

The colloid concentration at the second node of the rock-matrix $C_p^{n+1}{}_2$ is the fourth unknown in the $(n+1)^{th}$ time level in equation (27). The value of this unknown is assumed and iterated until convergence. Thus using tridiagonal matrix algorithm (TDMA), the three unknowns are solved for the fracture at i^{th} node, $(i+1)^{th}$ node and $(i-1)^{th}$ node at $(n+1)^{th}$ time level. Thus, the fourth unknown, the colloid concentration at the second node of the skin $C_p^{n+1}{}_2$ at $(n+1)^{th}$ time level is not solved by the TDMA as its value is assumed at $(n+1)^{th}$ time level and iterated until convergence. A similar approach is adopted in

solving the last term $\frac{\theta D_p^{**}}{b} \frac{\partial N_p}{\partial z}$ in equation (12) for radionuclide transport.

Having obtained the concentration distribution of radionuclides along the fracture for a coupled fracture-matrix system, at each time level, from the above numerical method, the method of spatial moments as a function of traveling time is calculated. Such spatial moments of point concentration data provide an integrated measure of the concentration field over the entire extent of the domain. In the present model, the radionuclide transport parameters are considered by characterizing the three spatial moments of the concentration distribution along the fracture.

The lower order spatial moments have been obtained using a similar approach to Guven et al. [28]. The zeroth moment (M_0) is proportional to the total mass of the fluid in the high permeability fracture. The first spatial moment (M_1) describes the displacement of the centre of the mass and the second spatial moment (M_2) describes the spread of the deviation from the centre of mass. The expressions for evaluation of zeroth moment, first moment and second moment is given below.

$$M_n = \int_0^L x^n c(x,t) dx \tag{38}$$

$$X_1(t) = \frac{M_1}{M_0}, \tag{39}$$

$$X_{11}(t) = \frac{M_2}{M_0} - \left[\frac{M_1}{M_0} \right]^2, \tag{40}$$

From these moments, the mobility and spreading of the concentration profiles can be obtained using the following expressions:

$$V(t) = \frac{d\{X_1(t)\}}{dt}, D(t) = \frac{1}{2} \frac{d\{X_{11}(t)\}}{dt}, \quad (41)$$

However, the above expressions are valid for pulse sources [29] only. Since the boundary condition in the inlet is assumed to be a constant continuous source, a first derivative of the concentration in the fracture is used to obtain the equivalent pulse in order to use the above expressions (38) to (41).

4. Results and discussion

For the purpose of verification of the numerical model, the concentration profiles of the colloids and radionuclides have been computed using a Dirichlet type boundary condition at the inlet as well as the outlet of the fracture. The results of the numerical model for the radionuclides in a fracture-matrix coupled system are compared with the analytical solutions of Sudicky and Frind [30] while the numerical model for colloids in a single fracture is compared with the analytical solution of Van Genuchten [31]. The parameters used for validation of the numerical results with the analytical solution for radionuclides and colloids are presented in Table 1 and 2. The base case data pertaining to radionuclides is adopted from Li et al. [19] and colloids from Abdel-Salam and Chrysikopolous [32].

Table 1. Parameters used for validation of the numerical model for radionuclides

Parameter	Symbol	Value
Initial fracture aperture (μm)	2b	10000
Fluid velocity (m/d)	V	1
Porosity of the rock-matrix	θ_m	0.01
Longitudinal fracture dispersivity (m)	α_L	0.5
Free molecular diffusion coefficient (m^2/d)	D^*	1e-05
Matrix diffusion coefficient (m^2/d)	D_m	1e-05
Length of the fracture (m)	L_f	50
Total simulation time (day)	T	25
Concentration of radionuclides at the inlet of the fracture (kg/m^3)	N_0	1

Table 2. Parameters used for the validation of the numerical model for colloids

Parameter	Symbol	Value
Initial half-fracture aperture (m)	b	1.25e-04
Fluid velocity (m/year)	V	1
Hydrodynamic dispersion coefficient in the fracture (m^2/year)	D	0.25
Length of the fracture (m)	L_f	150
Total simulation time (day)	T	5
Colloid dispersion coefficient (m)	κ	1e-10
Concentration of colloids at the inlet of the fracture (kg/m^3)	C_o	1

The results for the verification of the models have been shown in Figures 2 and 3. The analytical solution is represented by solid lines while the numerical solution is represented by data points.

It is observed from Figures 2 and 3 that the numerical results are in close agreement with the analytical solution for the data provided in Table 1 and 2, which illustrates the robustness of the numerical model. The parameters used for numerical simulation of colloid facilitated radionuclide transport are presented in Table 3 below.

Table 3. Parameters used for the colloid facilitated radionuclide transport

Parameter	Symbol	Value
Initial half-fracture aperture (m)	b	500e-06
Fracture-skin thickness (m)	d-b	0.02
Fracture spacing (m)	2H	15
Average fluid velocity (m/year)	V	1
Radioactive decay constant for ^{239}Pu (yr^{-1})	λ	2.881e-05
Diffusion coefficient of radionuclides within the fracture-skin (m^2/year)	D_p	0.01
Diffusion coefficient of radionuclides within the rock-matrix (m^2/year)	D_{mat}	0.01
Porosity of the fracture-skin	θ	0.09
Distribution coefficient for radionuclides on the fracture surface (m)	K_{d_N}	0
Distribution coefficient for radionuclides in fracture-skin (m)	$K_{d_{NP}}$	0
Distribution coefficient for radionuclides in rock-matrix (m)	$K_{d_{Nmat}}$	0
Hydrodynamic dispersion coefficient of radionuclides dissolved in the fracture aqueous phase (m^2/year)	D	10
Distribution coefficient for radionuclides with mobile colloids within the fracture (for colloid diameter of 300nm) (m^3/kg)	K_{d_m}	40
Distribution coefficient for radionuclides with immobile colloids within the fracture (m^3/kg)	$K_{d_{im}}$	40
Distribution coefficient for radionuclides with mobile colloids within the fracture-skin (m^3/kg)	$K_{d_{mP}}$	40
Distribution coefficient for radionuclides with immobile colloids within the fracture-skin (m^3/kg)	$K_{d_{imP}}$	40
Distribution coefficient for radionuclides with mobile colloids within the rock-matrix (m^3/kg)	$K_{d_{mmat}}$	40
Distribution coefficient for radionuclides with immobile colloids within the rock-matrix (m^3/kg)	$K_{d_{immat}}$	40
Average velocity of colloids in the fracture (m/year)	V_C	1
Colloid concentration at the inlet of the fracture (kg/m^3)	C_o	1
Hydrodynamic dispersion coefficient of colloids suspended in the rock fracture (m^2/year)	D_C	1
Filtration coefficient for colloids (m^{-1})	λ_f	0.5
Percentage of diffusion for colloids	ε	0.5
Diffusion coefficient of colloids within the fracture-skin (m^2/year)	D_{CP}	2.2e-08
Diffusion coefficient of colloids within the rock-matrix (m^2/year)	D_{Cmat}	1.5e-09
Distribution coefficient for colloids within the fracture-skin	$K_{d_{CP}}$	0.1
Distribution coefficient for colloids within the rock-matrix	$K_{d_{Cmat}}$	0.01
Remobilisation coefficient for colloids in the fracture (year^{-1})	Rmb	0.5
Length of the fracture (m)	L	20
Total simulation time (year)	T	5
Concentration of radionuclides at the inlet of the fracture (kg/m^3)	N_o	1

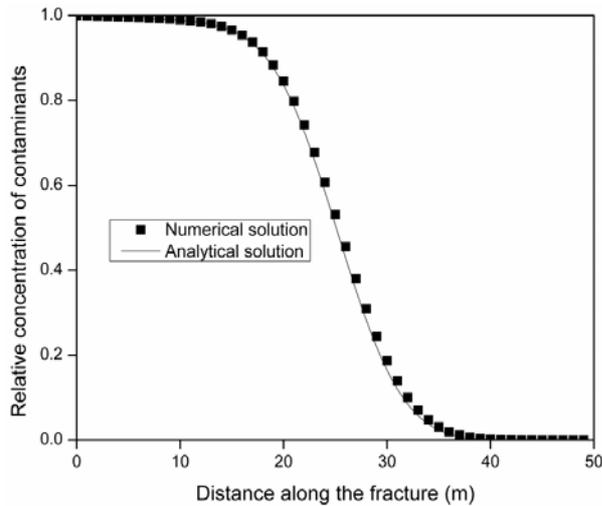


Figure 2. Validation of numerical results with analytical solution for radionuclides (in the absence of colloids) in a fracture-matrix coupled system (Refer to Table 1 for data)

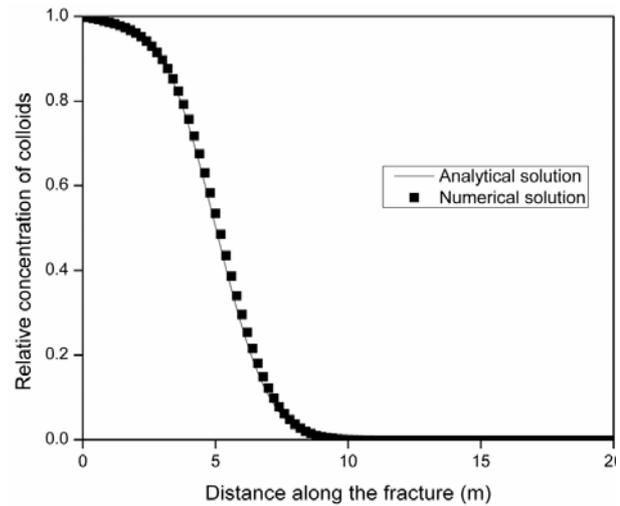


Figure 3. Validation of numerical results with analytical solution for colloid transport (in the absence of radionuclides) in a single rock fracture (Refer to Table 2 for data)

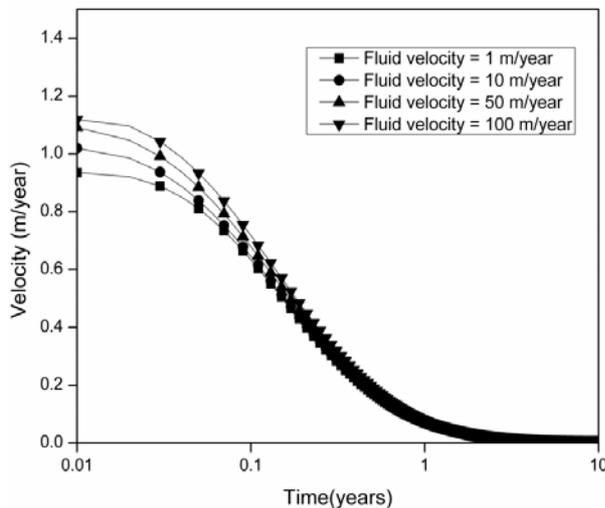


Figure 4. Temporal distribution of velocity of radionuclides facilitated by movement of colloids in a coupled fracture-matrix system with different fluid velocities. (Colloid velocity = 10 m/yr, Remobilization coefficient = 0.5 yr^{-1} , Filtration coefficient = 0.5 m^{-1}). Refer Table 3 for other parameters

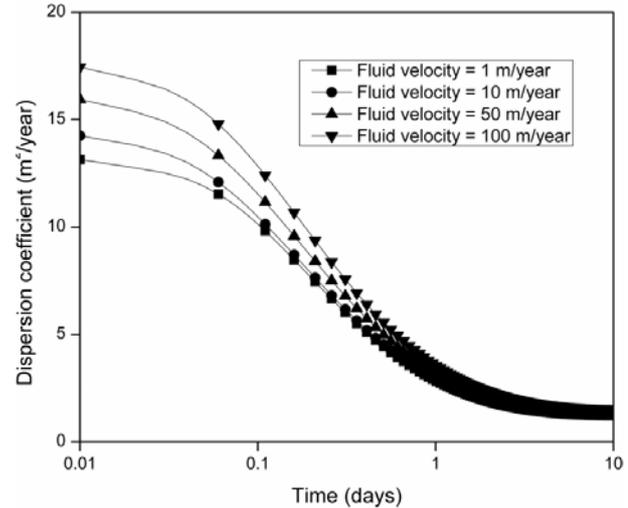


Figure 5. Temporal distribution of dispersion of radionuclides facilitated by movement of colloids in a coupled fracture-matrix system with different fluid velocities. (Colloid velocity = 10 m/yr, Remobilization coefficient = 0.5 yr^{-1} , Filtration coefficient = 0.5 m^{-1}). Refer Table 3 for other parameters

Figure 4 provides the temporal distribution of velocity of radionuclides facilitated by colloid transport for different fluid velocities in a fracture-matrix coupled system. It can be noted that slope of first spatial moment with time provides the velocity of the radionuclide. It is observed from Figure 4 that the velocity of the radionuclide has significantly reduced nearer to the source. The velocity further reduces as time progresses and becomes zero just after a year. The velocity of radionuclides reduces due to sorption onto colloids and fracture wall surface. It is observed from the figure that at the early stages the velocity of radionuclides is relatively higher when the fluid velocity is high ($V = 100 \text{ m/year}$) but as time progresses the velocity profiles merge with each other as the concentration of radionuclides gradually reduces along the length of the fracture due to matrix diffusion, sorption of the fracture wall surface and adsorption onto the colloids. The mobility of radionuclides follows the same trend irrespective of the fluid velocity. Figure 5 provides the temporal distribution of dispersion of radionuclides facilitated by colloid transport for different fluid velocities in a fracture-matrix coupled system. It can be noted that half the slope of

second spatial moment with time provides the effective dispersion coefficient. It is observed from Figure 5 that more mixing is experienced near the inlet of the fracture and dispersion coefficient reaches a constant value after a few years. In a conventional fracture-matrix coupled system without colloids, dispersion increases with decrease in velocity of radionuclides but it is observed from Figure 5 that dispersion reduces as velocity of radionuclides decrease with time. The deposition of colloids on the fracture surface and sorption of radionuclides on immobile colloids deposited on the fracture walls has resulted in such unique behavior in the fracture-matrix coupled system. It is observed from the figure that at the early stages the dispersion of radionuclides is relatively higher when the fluid velocity is high ($V = 100$ m/year) but as time progresses the profiles merge with each other as more colloids get sorbed on to the fracture walls.

Figure 6 provides the temporal distribution of velocity of radionuclides facilitated by colloid transport for different colloid remobilization coefficients in a fracture-matrix coupled system. It is observed from Figure 6 that the velocity of the radionuclides is approximately constant for all the remobilization coefficients and becomes zero after a few years. The velocity of radionuclides is relatively higher when there is no remobilization of colloids in the system as the absence of colloids increases the mobility of radionuclides in the aqueous phase. Remobilization is accompanied by filtration of colloids from the aqueous phase and thus the velocity profiles are similar for the range of fluid velocities.

Figure 7 provides the temporal distribution of dispersion of radionuclides facilitated by colloid transport for different colloid remobilization coefficients in a fracture-matrix coupled system. It is observed from Figure 7 that there is very high dispersion at the early stages as matrix diffusion is very low and dispersion decreases as time progresses and attains a constant value. When all the colloids are remobilized into the aqueous phase, i.e. $R_{mb} = 1$ yr⁻¹, dispersion is very significant. The remobilization process hinders the diffusion of radionuclides into the rock-matrix although their velocity is very low. Even though the residence time of the radionuclides is very low, the presence of colloids affects the dispersive behaviour of radionuclides in the fracture-matrix coupled system.

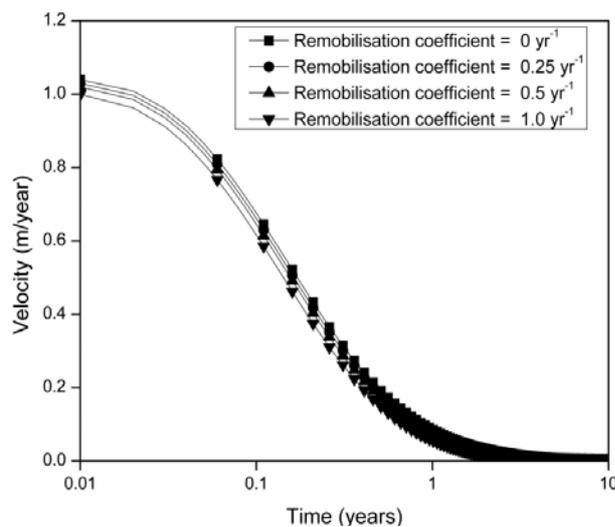


Figure 6. Temporal distribution of velocity of radionuclides facilitated by movement of colloids in a coupled fracture-matrix system with different colloid remobilization coefficients. (Colloid velocity = 10 m/yr, Fluid velocity = 10 m/yr, Filtration coefficient = 0.5 m⁻¹). Refer Table 3 for other parameters

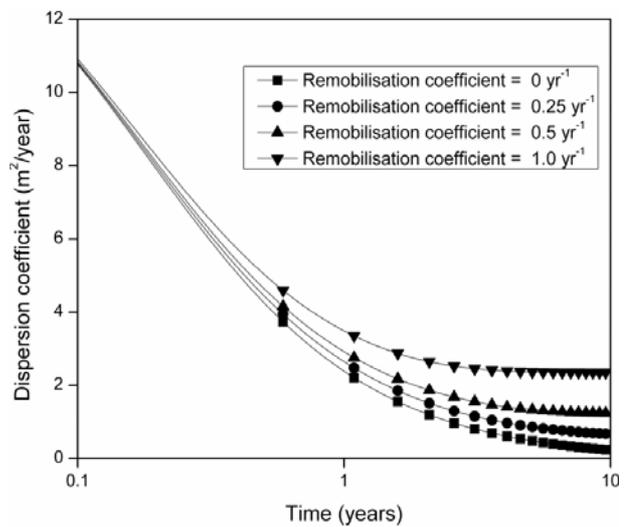


Figure 7. Temporal distribution of dispersion of radionuclides facilitated by movement of colloids in a coupled fracture-matrix system with different colloid remobilization coefficients. (Colloid velocity = 10 m/yr, Fluid velocity = 10 m/yr, Filtration coefficient = 0.5 m⁻¹). Refer Table 3 for other parameters

Figure 8 provides the temporal distribution of velocity of radionuclides facilitated by colloid transport for different colloid filtration coefficients in a fracture-matrix coupled system. It is observed from Figure 8 that when there is no filtration of colloids i.e. $\lambda_f = 0$, the velocity of radionuclides is zero throughout the simulation period. As mentioned above, it is confirmed that the presence of colloids retard the mobility of radionuclides in the aqueous phase. On the other hand, when there is maximum filtration of colloids

from the aqueous phase, it is observed from Figure 8 that there is very high velocity at the early stage. The velocity reduces as time progresses and finally reaches zero as significant quantity of colloids and radionuclides has entered into the rock-matrix.

Figure 9 provides the temporal distribution of dispersion of radionuclides facilitated by colloid transport for different colloid filtration coefficients in a fracture-matrix coupled system. It is observed from Figure 9 that dispersion is high at the initial time period and decreases as time progresses and becomes constant after few years except when the filtration coefficient is zero. When the colloids are not filtered there is complete mixing of radionuclides within the aqueous phase as the sorption of colloids on the fracture wall surface is negligible and this result in significant matrix diffusion. Consequently, the dispersion coefficient attains an asymptotic value. As filtration of colloids is progressively increased, dispersion reduces and attains a constant value. It can be concluded that significant mixing prevails along the fracture for all time levels.

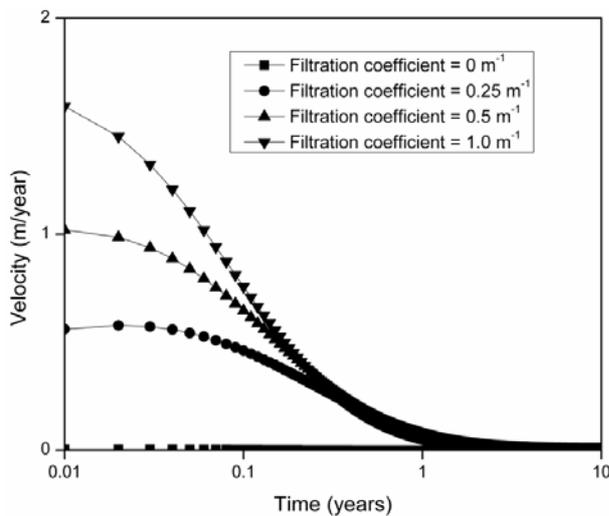


Figure 8. Temporal distribution of velocity of radionuclides facilitated by movement of colloids in a coupled fracture-matrix system with different colloid filtration coefficients. (Colloid velocity = 10 m/yr, Fluid velocity = 10 m/yr, Remobilization coefficient = 0.5 yr⁻¹). Refer Table 3 for other parameters

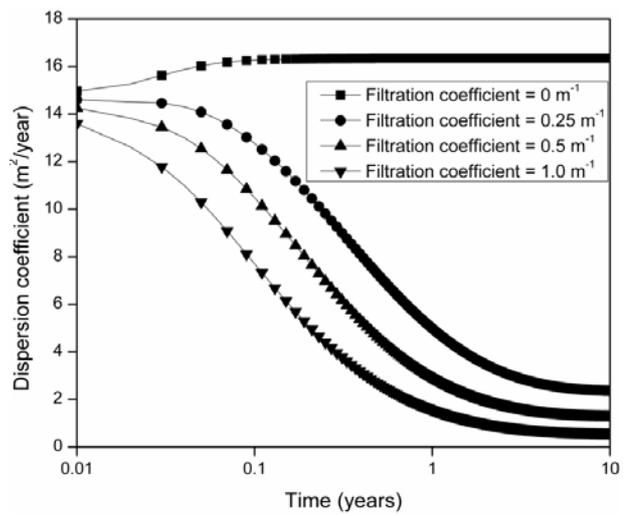


Figure 9. Temporal distribution of dispersion of radionuclides facilitated by movement of colloids in a coupled fracture-matrix system with different colloid filtration coefficients. (Colloid velocity = 10 m/yr, Fluid velocity = 10 m/yr, Remobilization coefficient = 0.5 yr⁻¹). Refer Table 3 for other parameters

5. Conclusion

In this paper, spatial moment of analysis has been performed on colloid facilitated radionuclide concentration obtained using the numerical model for various parameters. A constant continuous source has been assumed for the radionuclides and colloids. The fractures are assumed to be saturated. The conclusions from the study can be summarized as follows:

- 1- As time progresses the velocity profiles merge with each other as the concentration of radionuclide gradually reduces along the length of the fracture due to matrix diffusion, sorption of the fracture wall surface and adsorption onto the colloids. The mobility of radionuclides follows the same trend irrespective of the fluid velocity.
- 2- More mixing is experienced near the inlet of the fracture. Dispersion reduces as velocity of radionuclides decrease with time due to the deposition of colloids on the fracture surface and sorption of radionuclides on immobile colloids deposited on the fracture walls.
- 3- The velocity of radionuclides is relatively higher when there is no remobilization of colloids in the system as the absence of colloids increases the mobility of radionuclides in the aqueous phase.
- 4- Very high dispersion is observed at the early stages as matrix diffusion is very low and dispersion decreases as time progresses and attains a constant value. The remobilization process hinders the diffusion of radionuclides into the rock-matrix although their velocity is very low.

- 5- When there is no filtration of colloids i.e. $\lambda_f = 0$, the velocity of radionuclides is zero throughout the simulation period which confirms that the presence of colloids retard the mobility of radionuclides in the aqueous phase.
- 6- When the colloids are not filtered there is complete mixing of radionuclides within the aqueous phase as the sorption of colloids on the fracture wall surface is negligible and this result in significant matrix diffusion.
- 7- The presence of colloids have reduced the mixing of radionuclides within the fractures significantly as against the mixing experienced in a coupled fracture- matrix system in the absence of colloidal transport.
- 8- Significant mixing prevailed along the fracture for all time levels, when the filtration coefficient is zero.

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