Investigation of Colloid-facilitated Effects on the Transport of N-member Radionuclide Chains in the Fractured Medium

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ABSTRACT

A previous analytical model for N-member radionuclide decay chains has been extended to include the effect of radionuclide sorption with groundwater colloids.   Site-specific distribution coefficients were employed in the nuclide decay chain to illustration the present model.   The colloid concentration was assumed constant in time and space owing to equilibrium between colloid generation and sedimentation by chemical and/or physical perturbations.   Furthermore, the diffusion of colloids into the rock matrix was ignored because the diameter of colloid is relatively large and colloids and fracture surfaces are like-charged.   The results indicated that colloids could facilitate the transport of radionuclides and the large adsorbability of nuclides with colloids enlarged the effect of acceleration by colloids.   The influence of colloids on the radionuclide transport was expected to be crucial to the actinides with large adsorbability; however, the present results revealed that the low-adsorbing nuclides whose parent nuclides have large capability of sorption could be also facilitated significantly by colloids indirectly. Therefore, the role of colloids played in the transport of the radionuclides decay chain should be assessed carefully in the radioactive waste disposal.   The analytical method presented herein is helpful to verify/validate further complex far-field models.

INTRODUCTION

Colloids are tiny particles in the size range 1 nm to 1 μm which can remain suspended in water. Groundwater often contains significance populations of natural colloids which can interact with pollutants and influence their transport.   Natural colloids as well as waste- and repository-derived colloids in radioactive waste disposal may influence nuclide transportation significantly.   Laboratory experiments[1] in terms of colloid-facilitated contaminant transport have confirmed that colloids can enhance the transport of cationic and anionic metals through fractured media.   Moreover, the unexpectedly rapid transport of plutonium and americium at experimental sites may be also attributed to colloid-facilitated transport[2].

In many countries, the processes of colloid formation and colloid transport are included in the features, events, and processes (FEPs) list of their Performance Assessment (PA) programs for high-level waste/spent nuclear fuel disposal.   Although some PAs have deferred the consideration of colloid-nuclide transport until detailed evaluations has been performed, other PAs, for instance, H-12 of Japan[3]   and the Yucca Mountain Project of USA[4], have

considered the effects of the presence of colloids.   The experimental results[3] indicated that the engineered barrier designed to have the practical density bentonite more than 800 kg/m^3 had enough property to eliminate the colloid transport through the engineered barrier.   However, failure of the engineered barriers and bentonite buffer to filter colloids should be considered as an alternative scenario[5], especially according to the colloid and radionuclide retardation experiments[6,7] recently carried out in situ at Grimsel test site.

In the past decades, the single radionuclide transport facilitated by colloids has attracted lots of researcher's attention[8-10].   When a radionuclide is adsorbed onto the colloidal surface, it can not diffuse into the rock matrix and be adsorbed in the matrix rock, since the sizes of colloids are conservatively assumed to be larger than those of the rock matrix pores.   Colloids may move along the groundwater, therefore, radionuclides attached to these colloidal surfaces will be transported further such that the concentration in the fracture will be higher than that in the absence of colloids.   Those studies did not address the significance of the colloids in the chain decay, however, the in-growth and decay of actinides are crucial for assessing their transport ability owing to the long half-life times and radiotoxicity of actinides.   Sun and Buscheck[11] reported an analytical model for N-member decay chains in a single fracture in the absence of colloids.   We previously developed a semi-analytical model to access the co-transport of a two-member decay chain radionuclides and groundwater colloids[12].   Currently, the role of colloids in facilitating the transport of radionuclides is not understood enough to effectively model radionuclide transport.   However, there is a growing body of evidence that suggested low-soluble and strong-absorbing radionuclides, such as actinides, could be accelerated by colloids[2,6]. This present study extends previous studies to investigate the effects of the presence of colloids on actinides decay chains transport.   Parent and daughter radionuclides could perform completely different retardation properties which are explicitly represented within this study.   Thus, the present model is useful to verify complex numerical models for multi-species transport with different retardation factors and to understand the significances of colloids on the retardation properties of radionuclide decay chains.

CONCEPTUAL MODEL

The "Far- Field" (geosphere) is a physical region representing the natural barrier in the concept of geological disposal of nuclear waste.   In the concept of deep geological disposal of nuclear waste, the natural barrier is the geological material hosting the repository.   In determining the medium type of the far field, any potential pathways must be identified.   Usually tight formations are considered ideal candidates for hosting repositories.   Due to various reasons such as stresses, defects, and heterogeneous, geological materials are seldom perfect.   The most common defects are fractures.   Most rock formations considered in geological disposal of nuclear waste contain fractures.   Because fractures have greater void spaces than rock matrix, they are considered, in PA modeling, as the fast pathway of the geological medium.   Radionuclide transport in fractured rock is characterized by advection and diffusion/dispersion attenuated by sorption and matrix diffusion.   Apparently, these transport process must be considered in the PA modeling.

      Approaches to modeling flow and transport in fracture systems are not unique.   However, it is known that knowledge of various processes taking place in a single fracture is fundamental to improve our understanding radionuclide transport in fractured subsurface formations[13].   Therefore, the case of a thin rigid fracture slab in a saturated porous rock is considered herein.   Several assumptions are made regarding the properties of the groundwater colloids and the

system[8-14]:
1. The fracture is open and hence has unit porosity.
2. In the fracture, groundwater flows in one direction. Radionuclides in the aqueous phase, both dissolved and sorbed on the mobile colloids, transport advectively and dispersed along the fracture.
3. Transverse dispersion is efficient to ensure mixing of radionuclide concentrations across the fracture slab in the direction perpendicular to flow.
4. In the matrix, groundwater is at rest and radionuclide diffusion only occurs in the direction perpendicular to the fracture.
5. Considering radioactive in-growth and decay.
6. Colloidal particulates are mobile in the fracture. However, they cannot diffuse into the rock matrix due to their large size.
7. The colloidal concentration is constant and uniform along the fracture.
8. Parameters (porosity, groundwater/colloid velocity, sorption coefficients, diffusion and dispersion coefficients, etc.) are assumed to be constant and uniform.

MATHEMATICAL MODEL

Accordingly, the equation describing the ith decay chain radionuclide transport with colloids in the fracture is[12,15,16]:

$$R^*_{f,i}\frac{\partial N_{f,i}}{\partial t} + v^*_f\frac{\partial N_{f,i}}{\partial x} - D^*_f\frac{\partial^2 N_{f,i}}{\partial x^2} - a_f\theta_m D_m\frac{\partial N_{m,i}}{\partial z}\bigg|_{z=b} = \lambda_{i-1}R^*_{f,i-1}N_{f,i-1} - \lambda_i R^*_{f,i}N_{f,i},$$

$x \geq 0, b \geq z \geq 0, t \geq 0$ (Eq. 1)
where,
$$R^*_f = 1 + Kd_m\rho_m\delta a_f + Kd_c C_0, \quad v^*_f = v_f + v_c Kd_c C_0, \quad D^*_f = D_f + D_c Kd_c C_0.$$
Similarly, the transport equation for the ith radionuclide in the matrix is:

$$R_{m,i}\frac{\partial N_{m,i}}{\partial t} - D_m\frac{\partial^2 N_{m,i}}{\partial z^2} = \lambda_{i-1}R_{m,i-1}N_{m,i-1} - \lambda_i R_{m,i}N_{m,i}$$

$x \geq 0, z \geq b, t \geq 0$ (Eq. 2)
Symbols appearing in the above equations are defined as follows:
N is the aqueous concentration (kg/m^3),
$v_{f/c}$ is the groundwater/colloid velocity in the fracture (m/yr),
$D_{f/c}$ is the longitudinal dispersion coefficient of radionuclide/colloid in the fracture (m^2/yr),
θ_m is the rock matrix porosity,
D_m is the diffusion coefficient of radionuclide in the rock matrix pores (m^2/yr),
λ is decay constant (1/yr),
$Kd_{m/c}$ is sorption (distribution) coefficient for radionuclide on rock/colloid (m^3/kg),
ρ_m is bulk density of the rock (kg/m^3),
R_m is the matrix retardation coefficient defined as: $R_m = 1 + \dfrac{Kd_m\rho_m}{\theta_m}$,

δ is sorption length for radionuclide on the fracture surface (m),
b is the half of the fracture aperture (m),

a_f is specific flow-wetted area of fracture per unit flow volume (1/m), for smooth fracture surface: $a_f = \dfrac{1}{b}$,

C_0 is the colloidal concentration in the fracture (kg/m^3).

Subscripts "i" and "i-1" refer to the ith and (i-1)th radionuclides, respectively, and subscripts "f" and "m" refer to the fracture and rock matrix, respectively.

As for the case of the single radionuclide transport in the absence of colloids, Eq. (1) could be reduced to the form similar to that used in the literature[14] for modeling advective/dispersive radionuclide transport in a 1-D fracture. Initial and boundary conditions for the transport equations, Eq. (1) coupled with Eq. (2) are given as:

$$N_{f,i}(x = 0, t) = N_{0,i}\delta(0),$$ (Eq. 3a)

$$N_{f,i}(x = \infty, t) = 0,$$ (Eq. 3b)

$$N_{f,i}(x, t = 0) = 0,$$ (Eq. 3c)

$$N_{m,i}(x, z = b, t) = N_{f,i}(x, t),$$ (Eq. 3d)

$$N_{m,i}(x, z = \infty, t) = 0,$$ (Eq. 3e)

$$N_{m,i}(x, z \geq b, t = 0) = 0,$$ (Eq. 3f)

where N_0 is the initial concentration at the inlet of the fracture, $\delta(0)$ is the delta function, hence, it represents the pulse injection case at $t = 0$. Any other injection model, e.g., time series outputs from near-field modeling, can be analyzed using the principle of superposition.

For the single-radionuclide modeling, the analytical solution of the transport equation, Eq. (1), subject to the conditions of Eqs. (3a-f) has been obtained and the total mobile concentration of radionuclide, N_T, in the fracture can be given as[14-16]:

$$N_T = \frac{N_{T0}}{\pi} \int_{\frac{\eta}{2\sqrt{t}}}^{\infty} \frac{Y}{T^{3/2}} \exp\left(\alpha x - \xi^2 - \frac{\alpha^2 x^2}{4\xi^2} - \lambda t - \frac{Y^2}{4T} \right) d\xi .$$ (Eq. 4)

where, $N_{T0} = N_0(1 + Kd_c C_0)$, $Y = \dfrac{R_f^* x^2}{4AD_f^* \xi^2}$, $A = \dfrac{bR_f^*}{\theta(D_m R_m)^{1/2}}$, $T = t - YA$, $\alpha = \dfrac{v_f^*}{2D_f^*}$, and ξ is the integration parameter.

Sun and Buscheck[11] employed a transformed function to decouple the "in-growth" and "decay" terms from Eqs. (1) and (2). The transport system of each radionuclide became independent with an identical mathematical format in the transformed domain. The transformed function is defined as:

$$N_{T,i}^* = N_{T,i} + \sum_{j=1}^{i-1} \left[\prod_{n=j}^{i-1} \frac{K_n \lambda_n}{\lambda_n - \lambda_i} \right] N_{T,j} .$$ (Eq. 5)

where, $N_{T,i}^*$ is the analytical solution, with the form of Eq. (4), of the "ith" radionuclide in the transformed domain and $K_n = \dfrac{R_{f,n-1}^*}{R_{f,n}^*}$. Therefore, the analytical solution of the "ith" total mobile concentration of radionuclide in the presence of colloids could be expressed as:

$$N_{T,i} = N_{T,i}^* - \sum_{j=1}^{i-1} \left[\prod_{n=j}^{i-1} \frac{K_n \lambda_n}{\lambda_n - \lambda_i} \right] N_{T,j}$$ (Eq. 6)

NUMERICAL ILLUSTRATIONS

The analytical solution of Eq. (4) can be integrated by Gauss-Legendre quadrature.   Numerical demonstrations will be conducted by employing the parameters listed in Table I.   Colloidal velocity is assumed equal to groundwater velocity and the value of colloidal dispersion coefficient is quoted from Grindrod et al.[17].   The colloidal concentration in groundwater could range from 10^{-4} (experimental data[18]) to 10^{-1} (assumed value[8]) kg/m^3.   Our previous study[12] illustrated that the effect of the presence of colloids becomes significant when the colloidal concentration is greater than 10^{-4} kg/m^3.   Thus, the colloidal concentration used in the present study is assumed as 10^{-1} kg/m^3, for a conservative point of view.   However, the colloids composition is not included in our present modeling.   Table II summarized the sorption coefficients of several isotopes used in different PA programs.   The sorption coefficients of H-12, Kristallin-I and SITE-94 are basically the same order of magnitude; hence, the coefficients of H-12 are adopted for numerical illustrations herein.   The sorption coefficients of H-12 and half-life of radionuclides are listed in Table III.

First of all, the results obtained by our analytical model was compared with those solve by CRYSTAL, which is a numerical code for far-field radionuclide transport used in SKI's SITE-94 program[24].   CRYSTAL solves one-dimensional radionuclide transport through fractured media in the absence of colloids by implementing the numerical inversion of Laplace transform. For the purpose of verification, the analytical breakthrough curves at 100 m for the four radionuclides are almost identical with those obtained by CRYSTAL.

Table I Transport parameters used in the present work

Parameter	Value
Transport length (m)	100
Sorption length, δ (m)	0.05
Fracture aperture, 2b (m)	5×10^{-3}
Bulk density of the rock, ρ_m (kg/m^3)	2700
Rock matrix porosity, θ_m	0.02
Groundwater velocity in the fracture, v_f (m/yr)	40
Longitudinal dispersion coefficient of nuclide, D_f (m^2/yr)	50
Diffusion coefficient of nuclide in the rock matrix pores, D_m (m^2/yr)	7.88×10^{-4}
Colloidal velocity in the fracture, v_c (m/yr)	40
Longitudinal dispersion coefficient of colloid, D_c (m^2/yr)	140
Colloidal concentration, C_0 (kg/m^3)	0.1
Initial concentration of the ith nuclide, $N_{0,i}$ (kg/m^3)	1

Table II Sorption coefficients of several isotopes for different PA programs.

Isotope	Kd_m (m^3/kg)				
	H-12[19]	Kristallin-I[20]	SITE-94[21]	TILA-99[22]	SR-97[23]
U	1	1	5	0.1	5
Th	1	1	1	0.2	5
Am	5	5	5	0.04	3
Np	1	1	1	0.2	5
Ra	0.5	0.5	0.5	0.2	0.02

Table III Sorption coefficients and half-life of radionuclides (H-12)

Nuclide	Kd_m (m^3/kg)	Half-life (yrs)
Am-241	5	4.33×10^2
Np-237	1	2.14×10^6
U-233	1	1.59×10^5
Th-229	1	7.34×10^3
U-238	1	4.47×10^9
U-234	1	2.45×10^5
Th-230	1	7.71×10^4
Ra-226	0.5	1.60×10^3

The results obtained by the analytical solution and the numerical results by CRYSTAL for Neptunium decay chain: Am-241; Np-237; U-233; Th-229.   The retardation factors for sorption on fracture surface are all set to unity for all radionuclides in this case[25].   The analytical solutions not only agree well with the numerical results, but also eliminate the phenomenon of oscillation, which is the inherent drawback while using numerical inversion of Laplace transform. The analytical method is helpful to verify/validate further complex far-field models.
Figure 1 depicts the breakthrough curves for the four nuclides in the Neptunium decay chain based on the distribution coefficients for H-12[19], when the concentration of colloids is 0.1 kg/m^3.   The distribution coefficients for radionuclides with colloids (Kd_c) might be quite different than those with the matrix rock (Kd_m) due to the discrepancy in sorption surface area and/or composition.   The nuclide-colloid sorption coefficients are then assumed to be an order of magnitude greater than the nuclide-rock sorption coefficients[1] to elucidate the effect of adsorbability of nuclides with colloids in Figure 1.   As shown in Figure 1, it indicates that radionuclides transported in the fracture are greatly enhanced and accelerated by the presence of colloids.   The peak concentrations of radionuclides when the colloids exist in the system are approximately one order of magnitude larger than those without colloids.   The results herein show clearly that the transport of radionuclides in the fracture is facilitated by the large sorption of nuclides with colloids.   The large adsorbability of nuclides with colloids, which provide an

alternative mobile carrier for nuclides, implies the reinforcement of mobility of radionuclides. In other words, increasing the sorption coefficients of radionuclides with colloids will increase the capability of colloids to adsorb radionuclides and enhance the effects of acceleration by colloids.   As illustrating in Table II, the sorption coefficients of TILA-99[22] are extraordinarily smaller than other data.   Figure 2 illustrates the breakthrough curves with and without colloids for the four nuclides in the Neptunium decay chain based on the distribution coefficients for TILA-99 for both $Kd_c=Kd_m$ and $Kd_c=10 \times Kd_m$.   This figure indicates that the influence of colloids on the transport of radionuclides in the fractured media is insignificant because of their low adsorbability, even when the nuclide-colloid sorption coefficients are assumed to be an order of magnitude greater than the nuclide-rock sorption coefficients; there is only slightly enhancement on the transport of radionuclides.
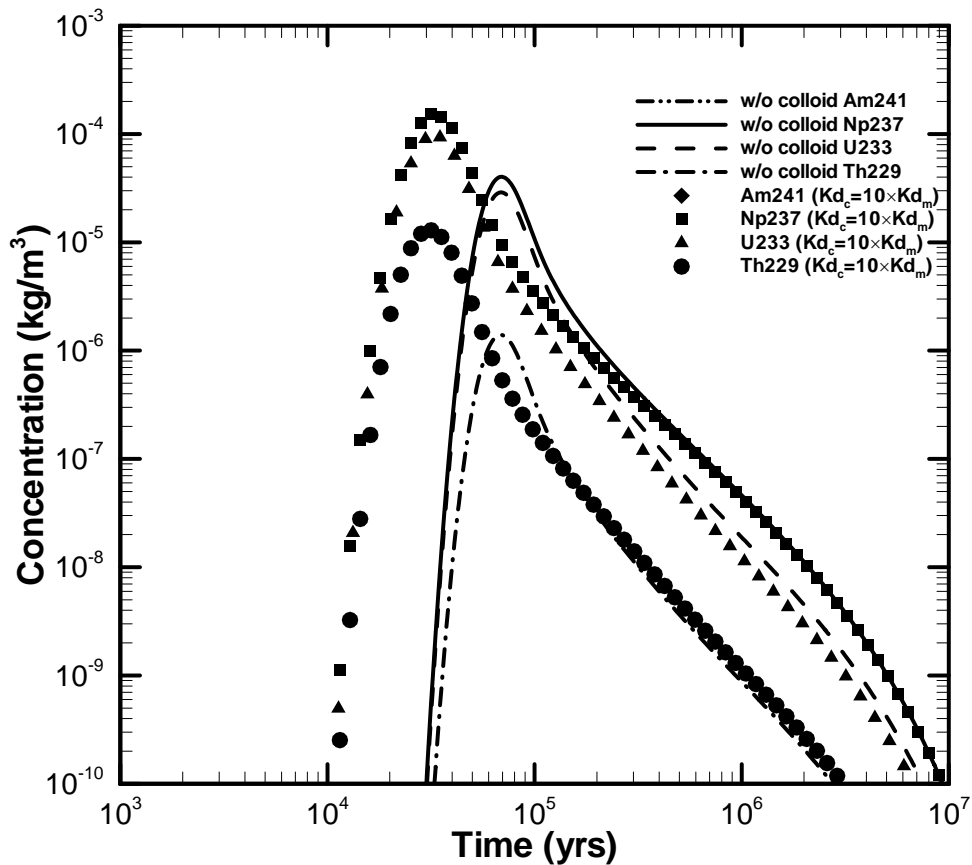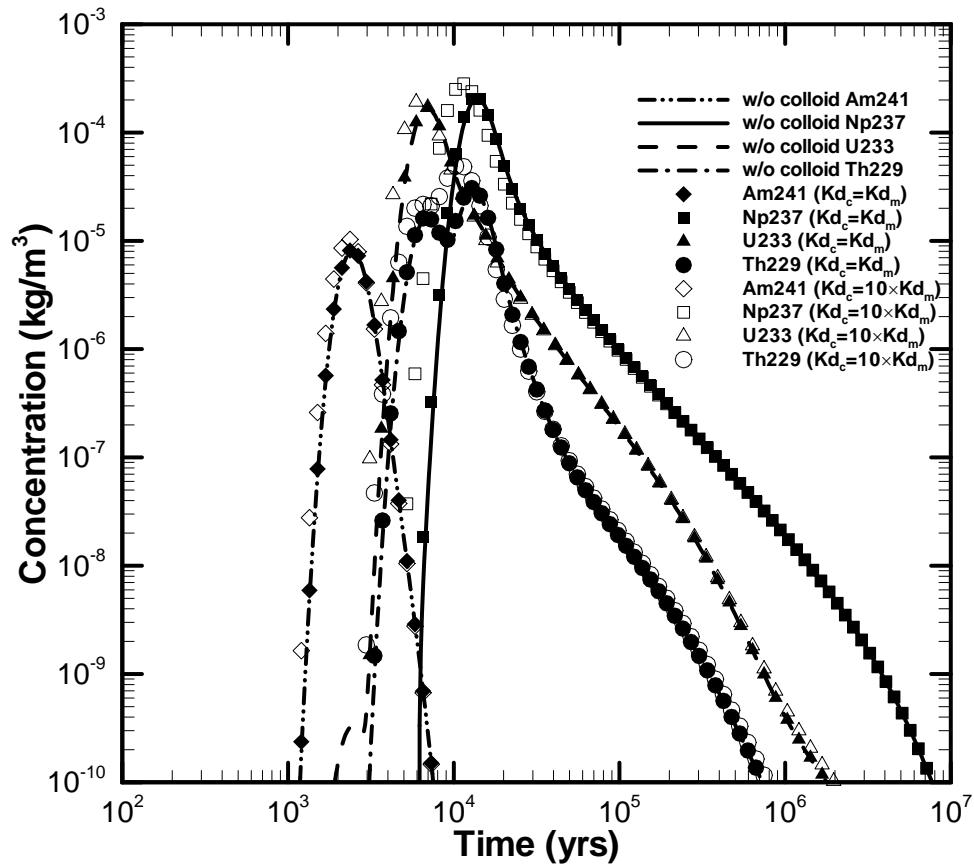

Figure 1. The breakthrough curves of four nuclides in the Neptunium decay chain at the distance of 100 m away from the radioactive source with/without colloids (the colloidal concentration is 0.1 kg/m^3 and $Kd_c=10 \times Kd_m$).

Figure 2. The breakthrough curves of four nuclides in the Neptunium decay chain at the distance of 100 m away from the radioactive source with/without colloids (the colloidal concentration is 0.1 kg/m^3) based on the parameters of TILA-99[22].

To further investigate the effect of colloids for the different decay chain, the breakthrough curves for the four nuclides in the Uranium decay chain based on the distribution coefficients for H-12[19] are plotted in Figure 3, for Kd_c is set to be equal to Kd_m and Kd_c is ten-fold of Kd_m. The transport of radionuclides in the Uranium decay chain is also slightly enhanced due to the existence of colloids when Kd_c is equal to Kd_m. The enhancement becomes significant while increasing the nuclide-colloid sorption. Moreover, Figure 4 shows the breakthrough curves for the four nuclides in the Uranium decay chain based on the distribution coefficients for SR-97[23] to investigate the effect of colloids on the transport of radionuclides in the Uranium decay chain. The breakthrough curves of U-238, U-234 and Th-230 in the right hand side of Figure 4 are enhanced and accelerated by mobile colloids because of the large sorption capability. There are two breakthrough curves of Ra-226 in the Uranium decay chain: one is contributed by Ra-226 itself; the other is imputed by the parent nuclide of Th-230. The breakthrough curve of Ra-226 in the left of the figure, which is contributed by Ra-226 itself, shows that the transport of Ra236 is not facilitated by the colloids because of its low adsorbability (0.02 m^3/kg in Table II); even Kd_c is ten times the value of Kd_m. However, another breakthrough curve of Ra-226 decayed from the parent nuclide of Th-230 in the right of the figure is significantly facilitated by colloids in the fractured media. The transport of Th-230 is enhanced by colloids; therefore, this enhancement facilitates the transport of Ra-226 indirectly. When Kd_c is ten times the value of

Kd_m, the effect of colloids on the transport becomes stronger.   The nuclide of Ra-226 with low adsorbability is accelerated unexpectedly because of the sorption of its parent nuclide of Th-230 on colloids.   The Th-230 nuclide is kidnapped by the colloids in the aqueous phase to keep its mobility and decays into Ra-226 afterward, thus, the concentration of mobile Ra-226 in the fracture increases.   The influence of colloids on the radionuclide transport was expected to be crucial to the actinides with large adsorbability; however, the present results reveal that the low-adsorbing nuclides whose parent nuclides have large capability of sorption could be also facilitated significantly by colloids indirectly.   This unexpected enhancement of the transport of such nuclides, for instance Ra-226, indicates that the existence of colloids should be seriously treated in the transport of the radionuclide decay chain in the geologic system.
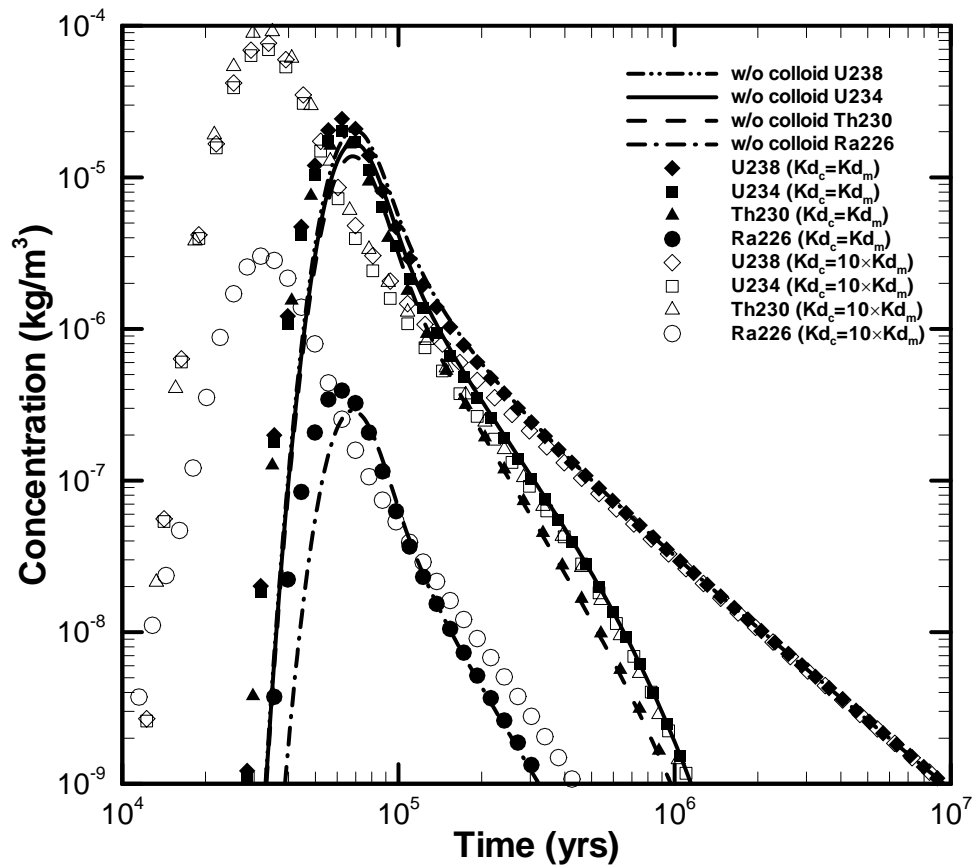

Figure 3. The breakthrough curves of four nuclides in the Uranium decay chain at the distance of 100 m away from the radioactive source with/without colloids (the colloidal concentration is 0.1 kg/m^3).
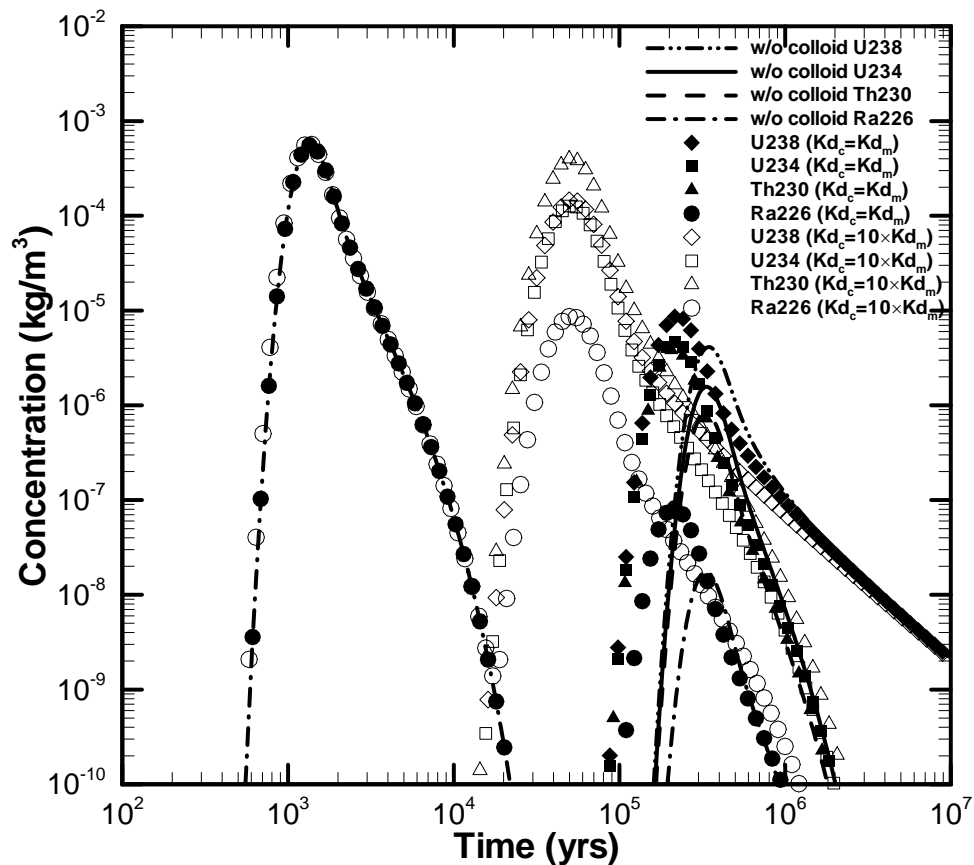
Figure 4. The breakthrough curves of four nuclides in the Uranium decay chain at the distance of 100 m away from the radioactive source with/without colloids (the colloidal concentration is 0.1 kg/m^3) based on the parameters of SR-97[23].

CONCLUSIONS

This present study has extended previous studies to study the effects of colloids on actinides decay chains transport in the fractured media. The site-specific parameters were employed herein to demonstrate our model. The results indicated that colloids could enhance and facilitate the transport of radionuclides. The large adsorbability of nuclides with colloids would enlarge the effect of acceleration by colloids. On the other hand, the facilitated transport by colloids is reduced while the decreasing adsorpability of nuclides with colloids. When the mobile colloids exit in a non-negligible concentration and have the capability of adsorb nuclides stably in the geologic system, colloids will be important for the transport of radionuclides[26]. Furthermore, an unexpected enhancement of the transport of the low-absorbing Ra-226 in the Uranium decay chain under the parameters of SR-97 occurred in the presence of colloids due to the large adsorbability of its parent nuclide of Th-230. The mobility of Th-230 is maintained by adsorbing on colloids and decays into Ra-226, therefore, the transport of Ra-226 contributed by the parent nuclide of Th-230 was enhanced indirectly. Consequently, the role of colloids played in the transport of the radionuclides decay chain should be assessed carefully in the radioactive waste disposal.

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