Development of a model for the mass loss rate of chemical erosion in fracture intersection with the deposition hole

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Abstract. The erosion of bentonite buffer in a fracture intersecting a deposition hole will affect the safety function. The purpose of this study was to build up a model which was to predict the rate of erosion of buffer/backfill, and this research focused on the KTH model which was developed by SKB. The KTH model consists of fluid pressure, sodium concentration, and smectite volume fraction, which is governed by Darcy's Law and advection-diffusion equations respectively, and the mutual dependencies of these variables were considered. This model was implemented using the COMSOL Multiphysics, and the results show the different behavior of sodium concentration and smectite in various seeping water velocity, the erosion rate increases with the seeping water velocity, but the distance to the smectite rim border decreases with the water velocity. The KTH considers the relationship between sodium and smectite, and with scalability it lays a foundation for the evaluation buffer/backfill mass loss by erosion.

1. Introduction
The buffer/backfill erosion problem was first considered by The SR-Can report [1], and seeping water did not affect erosion rate with advection, which was dominated by diffusion. It means that bentonite intrudes into the fracture by diffusion effect, and the critical coagulation concentration (CCC) was used to evaluate stability and formation of the smectite. Colloids were assumed not to form when the ion strength in seeping water was greater than CCC. The CCC was assumed to be 1 mM. SKB assumed the repository will overlain by ice sheet in the future, and the ion strength of seeping water below the CCC, which could flow into the repository there is erosion at gel/sol/water interface and cause mass loss of bentonite. Liu and Neretnieks used the conceptual model discussed above to estimate bentonite erosion rates. and give a method to estimate the rate of loss of bentonite due to erosion, which depends on the rate of loss of calcium and the concentration of calcium in the bentonite [2]. These findings provide methods for estimating the rate of erosion of bentonite, but there is a lack of understanding of the mechanism and process of colloidal release from deposition hole and erosion by seeping water. Therefore, it's necessary to develop a model that describes the bentonite expands and intrudes into the fracture and the colloidal release process in the correct way [3]. Therefore, many experiments, numerical modeling, and in-situ tests have been conducted in Sweden and internationally to address the chemical erosion of buffer/backfill.

To describe the phenomenon of the bentonite expansion and extrusion into the fracture, Liu et al. developed a dynamic force balance model based on the force balance of Brownian force, drag force,
van der Waals force, diffuse double layer force, buoyant force, and gravitational force. In addition, the numerical model was verified by the free expansion experiment of the bentonite [4-6]. However, in addition to extrusion and expansion, erosion of the bentonite also involves the formation of colloids and the erosion process carried away by the seeping water. Therefore, Moreno describe the flow characteristics of colloids, taking into account the viscosity coefficient equation related to the smectite volume fraction and the sodium concentration in the seeping flow which were based on the dynamic force balance model [7]. This model contains three physical quantities, which are the pressure, the volume fraction of smectite, and the concentration of sodium. with Darcy’s law for the pressure, and advection-diffusion equation for smectite volume fraction and sodium concentration respectively which is called the KTH model [8-9]. Based on the results of the KTH model, the erosion rate of the bentonite is directly proportional to the velocity of the seeping water but inversely proportional to the depth of penetration of the smectite. In addition to this, Moreno also conducted extensive parameter research and proposed an equation to evaluate the erosion rate of the bentonite, where the erosion rate is proportional to the seeping water velocity to the power of 0.41 and the fracture aperture, and this equation was also used to estimate the time to reach advective conditions in the buffer [10]. However, on the subject of buffer/backfill erosion, SKB concluded in the SR-Site report that demonstrates the phenomenon can not be ruled out in the assessment of long-term safety. Furthermore, there is still uncertainty about the modeling of colloid formation and erosion of bentonite, and using larger bentonite mass may offer a slightly longer time before advective conitions are created, but it's not a solution to the erosion issue, hence continued study on buffer erosion mechanisms is needed [11].

2. The KTH model

The KTH model was developed by Moreno for mass loss rate evaluation of buffer and backfill under fracture intersecting the deposition hole which describes the bentonite expansion and the diffusion of sodium under the seeping water. The fluid flow described by the Darcy equation with viscosity function which depending on sodium concentration and smectite volume fraction, the sodium ions transport represented by advection-diffusion equation, and the bentonite expansion and intrusion into fracture, described by the dynamic force balance model.

2.1. Fluid flow

The seeping water flow in the fracture is modeling by the Darcy equation, the KTH model modified the hydraulic conductivity to account for the viscosity of colloid which strongly dependent on the concentration of bentonite [9].

\[ \rho S \frac{\partial p}{\partial t} + \nabla \cdot (\rho u) = Q_m \]  \hspace{1cm} (1)

In this equation, \( \rho \) is the fluid density (kg/m\(^3\)), \( S \) is storage coefficient (1/Pa), \( p \) is the fluid pressure (Pa), \( Q_m \) is a mass source term (kg/m\(^3\)-s) and \( u \) is the Darcy velocity (m/s), which can be expressed as

\[ u = -\frac{K}{\rho g} (\nabla p + \rho g \nabla D) \]  \hspace{1cm} (2)

Where \( K \) is the hydraulic conductivity (m/s), \( g \) is the gravitational acceleration (m/s\(^2\)) and \( \nabla D \) is a unit vector in the gravity direction, which could set to zero in 2D simulation. The hydraulic conductivity is assumed to depend on the viscosity of the fluid flow

\[ K = K_v \frac{\eta_w}{\eta} \]  \hspace{1cm} (3)

Therefore, the seeping water velocity can be calculated from the volumetric flow rate, \( Q \) (m\(^3\)/s) and its cross area, \( A \) (m\(^2\)).
Here $\bar{t}$ is hydraulic gradient (m/m), $\delta$ and $b$ represent the fracture aperture (m) and aquifer thickness (m) respectively, in this study, the fracture aperture is equal to the aquifer thickness.

In Eq. (3), $\eta$ is the viscosity of fluid (Pa-s) and the subscript $w$ indicates the fluid is water. In addition, the relative viscosity of the fluid is defined as

$$\frac{\eta}{\eta_w} = 1 + 1.022\phi_{\text{cov}} + 1.358(\phi_{\text{cov}})^3$$

(5)

Where $\phi_{\text{cov}}$ is the co-volume fraction of the fluid. The co-volume fraction is defined as follow when the smectite particle was assumed a coin-like particle

$$\phi_{\text{cov}} = \frac{2}{3} \left( \frac{D_p + 2mk^{-1}}{D_p^2 \delta_p} \right)^3 \phi$$

(6)

Here $\delta_p$ is the particle thickness of smectite (m), $D_p$ is the diameter of smectite particles (m), $m$ is a fitting parameter which set to be 1, besides, the Debye length (m) represented by sodium concentration $c$ (mol/m$^3$) could be written as

$$\kappa^{-1} = \frac{\varepsilon_0 \varepsilon_r RT}{2cz^2F^2}$$

(7)

Where $\varepsilon_0$ and $\varepsilon_r$ is the vacuum permittivity and the relative permittivity respectively, $R$ is the gas constant (J/K/mol) and $T$ is the absolute temperature (°C), $F$ is the Faraday constant (C/mol) and $z$ is the valence of the sodium ions which is 1. The relative viscosity as a function of the smectite volume fraction in different ion concentrations is plotted in Figure 1. As shown in the figure, as smectite volume fraction or ion concentration increases, the fluid becomes less mobile, therefore, the fluid velocity is affected by smectite volume fraction and ion concentration in the rim border of expansion bentonite. Which becomes slower when the smectite volume fraction or ion concentration decreases.

Figure 1. The relative viscosity as a function of the smectite volume fraction in different sodium concentrations

2.2. Smectite expansion

The advection-diffusion equation is used to simulation the smectite expansion in a fracture aperture, in which the diffusivity is modeled by the dynamic force balance model [4]
\[ \frac{\partial \phi}{\partial t} = -u \nabla \phi + \nabla \cdot \left( \frac{\chi}{f} \nabla \phi \right) \]  

(8)

Where \( \phi \) is the volume fraction of smectite and the ratio of \( \chi \) to \( f \) can be considered as diffusivity, in which \( \chi \) is the sum of the energy of the particles (J), where \( f = f_0/(1-\phi) \) is the effective friction coefficient which consider the smectite moving into the fracture and water is moving in the opposite direction and the friction coefficient \( f_0 \) is defined as

\[ f_0 = 6 \pi \eta_w \left[ \frac{\delta_p}{2} \left( \frac{4 S_p}{\pi \delta_p^2} - 1 \right) \tan^{-1} \left( \frac{4 S_p}{\pi \delta_p^2} - 1 \right) \right] + V_p k_0 T^2 a_p^2 \eta_w \left( \frac{\phi}{1 - \phi} \right)^2 \]  

(9)

Here \( \eta_w \) is the viscosity of water (Pa-s), \( S_p \) is the particle surface area of smectite (m\(^2\)), \( V_p \) is the volume of coin-like particles (m\(^3\)), \( a_p \) is the specific surface area per unit volume of particles (m\(^2\)/m\(^3\)) and \( k_0 T^2 \) is Kozeny’s constant. In addition, the function \( \chi \) is defined as

\[ \chi = k_B T + \left( h + \delta_p \right)^2 \left( \frac{\partial F_A}{\partial h} - \frac{\partial F_B}{\partial h} \right) \]  

(10)

Where \( k_B \) and \( T \) is the Boltzmann constant (J/K) and absolute temperature (K), respectively, \( h \) is the separation between particles is given by

\[ h(\phi) = \delta_p \left( \frac{\phi_{\text{max}}}{\phi} - 1 \right) \]  

(11)

Explicitly, the derivative terms in Eq. (10) are given by

\[ \frac{\partial F_A}{\partial h} = -A_H S_p \left[ \frac{1}{h^4} - \frac{2}{(h + \delta_p)^2} + \frac{1}{(h + 2 \delta_p)^2} \right] \]  

(12)

\[ \frac{\partial F_B}{\partial h} = -4 \kappa \rho R T S_p \tanh \frac{y_m}{y_w} \sinh \frac{y_m}{2} \]  

\[ + \frac{1}{kh} \sinh \frac{y_m}{2} + \frac{2}{(kh)^2} \sinh \left( \frac{y_m}{2} \right) \]  

(13)

Where \( A_H \) is the Hamaker constant (J), and

\[ y_m = \sinh^{-1} \left[ 2 \sinh \frac{y_m}{2} + \frac{4}{\kappa h} \sinh \left( \frac{y_m}{2} \right) \right] \]  

(14)

\[ y_w = 4 \tanh^{-1} \left[ \tan \left( \frac{y_m^0}{2} \right) \exp \left( - \frac{\kappa h}{2} \right) \right] \]  

(15)
$$y_{\infty}^h = 4 \tanh^{-1} \left[ \tanh \left( \frac{y_{\infty}^0}{2} \right) \exp (-\kappa h) \right]$$

(16)

and

$$y_{\infty}^0 = 2 \sinh^{-1} \left( \frac{z F \sigma^0}{2 \sigma_0 \varepsilon} \kappa RT \right)$$

(17)

Where $\sigma_0$ is the specific charge on the particle surface (C/m$^2$). The effective diffusivity $D_F$ could be calculated depends on $\chi$ and $f$, and the diffusivity as a function of smectite volume fraction at different sodium concentrations is plotted in Figure 2. As shown in the figure, the diffusivity is constant at different sodium concentrations when the volume fraction less than $10^{-4}$ and above $10^{-1}$ but quite non-linearly between the range.

![Figure 2. Diffusivity as a function of smectite volume fraction at different sodium concentrations](image)

2.3. Sodium concentration

The sodium ions transport in the fluid is described by the advection-diffusion equation which is given by [9]

$$\frac{\partial c}{\partial t} = -u \nabla c + \nabla \cdot (D \nabla c)$$

(18)

In this equation, $u$ is the Darcy velocity (m/s), $t$ is time (s), $c$ is the sodium concentration (mol/m$^3$) and $D$ is the diffusion coefficient for sodium in seeping water (m$^2$/s), which varies as a function of smectite volume fraction with $D_0$ equals to $2 \times 10^{-9}$ (m$^2$/s)

$$D = D_0 \left(1 - \phi \right)^{1.6}$$

(19)

3. Case study-based model validation

The solver validation was performed by using a case study of bentonite expansion and sodium ions migration with fracture intersecting deposition hole. The purpose of this study is to establish the KTH model and evaluate the mass loss rate of bentonite under the seeping water effect, in addition, investigate the distribution of sodium ion concentration and smectite volume fraction under different seeping water conditions.

3.1. Computational details
A fracture intersects the deposition hole with 10m×5m geometry was modeled and implemented using the COMSOL Multiphysics. The cylindrical deposition hole has a radius of 0.875m. The fracture aperture and the hydraulic gradient is modeled with constant values which are $10^{-3}$ m and 0.1 respectively. The hydraulic conductivities are $1\times10^{-6}, 3\times10^{-6}$, and $1\times10^{-5}$ (m/s), therefore the seeping water velocities are 3.15, 9.45, and 31.5 (m/year) respectively according to Eq. (4). The hydraulic head is assigned to be 1 m at the left-hand side boundary and inflow seeping water with sodium concentration 0.1 mM but no flux of smectite. The outflow boundary condition and constant hydraulic head 0 m at the right-hand side are imposed. In addition, assigned no flux boundary conditions at the fracture mouth but the volume fraction of smectite is 0.4, and sodium concentration is 10 mM. The initial condition of sodium concentration is set to be 0.1 mM, and the smectite volume fraction at fracture mouth is set to zero.

The fracture water flow is calculated using Darcy's law in COMSOL, which describes the different seeping water velocities by varying the hydraulic conductivities. Also, the dynamic force balance model used to describe the expansion of smectite was developed using the transport of diluted species interface, in which the diffusivity represent by the ratio of $\chi$ to f. Besides, the same interface solved the sodium concentration, but the diffusivity is set as a function of smectite volume fraction directly.

### 3.2. Simulation results

The results of the seeping water velocity of 3.15, 9.45, and 31.5 (m/year) are shown in Figures 3 to 5, respectively. The contours represent the sodium concentration (mM), the color bar represents the distribution of smectite volume fraction (-), and the Darcy flow field is represented by the vector arrow (m/s). In addition to the release of sodium ions from the edge of the deposition hole into the fracture, the upstream also inflow the seeping water with 0.1 mM sodium ions concentration.

![Figure 3](image_url)

**Figure 3.** Results of KTH model for the case of 3.15 m/yr of seeping water (c: contour plot, U: arrows, phi: color bar)

The results of the ions concentration analysis are affected by both the seeping water and the deposition hole geometry. Under the effect of diffusion and advection, the ions concentration spreads outward uniformly from the edge of the deposition hole, and the distribution of contours has a higher density at the front of the disposal hole and a smaller outward spread. Also, the ions concentration near the low-pressure zone at the right-hand side of the deposition hole expands significantly outward and forms a wake flow of smectite that extends to the outlet of the computational domain. Besides, the diffusion effect causes the smectite to expand outward along the edge of the deposition hole. The expansion of smectite at the left-hand side of the deposition hole is suppressed under the influence of geometric and advection effects, while the smectite wake flow with a certain thickness is formed at the right-hand side of the deposition hole, and the smectite volume fraction decreases as the distance from the edge of the deposition hole increases.
The smectite spreads outward mainly by diffusion, there is lower mobility around the edge of the deposition hole. As the volume fraction decreases, the effect of advection becomes more obvious, and its mobility is increased gradually. Therefore, the mobilities of the smectite volume fraction mainly control the shape variation, but without any change, its mobility properties still follow the description of Eq. (5).

**Figure 4.** Results of KTH model for the case of 9.45 m/yr of seeping water (c: contour plot, U: arrows, phi: color bar)

**Figure 5.** Results of KTH model for the case of 31.5 m/yr of seeping water (c: contour plot, U: arrows, phi: color bar)

To quantify the results of the analysis of the chemical erosion rate of the buffer material, the mass flow rate of the buffer material through the right-hand side boundary of the fracture can be calculated as the product of the Darcy velocity and the smectite volume fraction.

\[
N = \rho_s \delta \int_0^y u(y) \phi(y) dy
\]  

(20)

The product of the Darcy velocity and the smectite volume fraction in the above equation can be represented as the mobility of smectite. The product of darcy velocity and smectite volume fraction with different seeping water velocities \(u=3.15, 9.45, \text{ and } 31.5 \text{ (m/year)}\) are shown in Figures 6 to 8, respectively. The figure shows that "flowing" smectite exists only in the edge region at the interface between the seeping water and the bentonite. There is no smectite volume fraction outside this area, while the Darcy velocity inside is close to zero, which means that the erosion rate of the buffer material is mainly contributed by the "flowing" smectite in the marginal area.
3.3. Mass loss rate evaluation

To evaluate the results of the mass loss rate, the mass flow rate of the smectite through the right-hand side boundary of the fracture can be calculated by the Eq. (20), in which, the fracture aperture $\delta_h$ is $10^{-3}$ m, and the specific density of smectite is 2750 kg/m$^3$. The corresponding mass loss rate is obtained by multiplying the integral of Darcy velocity and smectite volume fraction with the specific density of smectite and the fracture aperture in each case. The results are shown in Table 1 for the mass loss rate.
calculations at seeping water velocities $u=3.15, 9.45,$ and $31.5$ m/yr and compared with the reference [8]. As shown in the results, the model of this study is better validated in the case of $u=3.15$ and $9.45$ m/yr, with relative errors of $-6.42\%$ and $1.54\%$, respectively, but $46.58\%$ in the case of $u=31.5$ m/yr. This is because the edge region of the buffer will change with time, a high-resolution mesh should be used to solve the problem to provide a good description of the temporal and spatial behavior of the rim border of the smectite.

Finally, in Figures 3 to 5, the mass loss rate of smectite increases with the increase of seeping water velocity, but the thickness of the wake flow of smectite decreases with the increase of the seeping water velocity can be observed. The phenomenon of reduced thickness and increased mass loss means that the erosion rate of the bentonite is dominated by the mobile smectite, which, despite the reduced thickness of the wake flow of smectite, can transport more mass per unit of time at higher seeping water velocity.

| Seeping water velocity (m/yr) | Mass loss rate (kg/yr) | Relative Error (%) |
|------------------------------|------------------------|--------------------|
| 0.95                         | 0.026                  | -                  |
| 3.15                         | 0.043                  | -6.42              |
| 9.45                         | 0.071                  | 1.54               |
| 31.5                         | 0.117                  | 46.58              |
| 94.5                         | 0.18                   | -                  |
| 315                          | 0.292                  | -                  |

4. Conclusions
The KTH model is a numerical model based on different physical parameters, and then the dependencies between the parameters are used to achieve the coupling calculation. And the fluid flow is described by Darcy's equation, and the hydraulic conductivity is modified by using the relative viscosity to describe the flow characteristics of both the fluid and the smectite. The sodium concentration and smectite volume fraction are governed by advection-diffusion equations, in which, the diffusion effect is defined by the corresponding diffusion coefficients, while the advection effect is solved by the coupled Darcy velocity. The KTH model has good extensibility. The gravitational effect can be considered to analyze the effect of a non-horizontal fracture or the condition of different shapes or multiple fractures. Furthermore, it is possible to replace the Darcy equation with the Navier-Stokes equation to account for higher Reynolds number flow conditions or turbulence, but due to the non-linear relationship between the physical quantities, it can be expected that will take more time to compute than the original model. It can also be observed from the KTH model that the flux of smectite through the right-hand side boundary of the fracture increases with the increase of the seeping water velocity, but the thickness of the smectite wake flow decreases with the increase of the seeping water velocity. The phenomenon of reduced thickness and increased mass loss means that the erosion rate of the bentonite is dominated by the mobile smectite, which, despite the reduced thickness of the wake flow of smectite, can transport more mass per unit of time at higher seeping water velocity. Finally, to avoid the lack of mesh resolution, which makes it impossible to properly describe the process of smectite volume fraction change over time and space, future research work should include the study of mesh convergence analysis.

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