

# Coupled chemical-hydraulic-mechanical modelling of long-term alteration of bentonite

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**ABSTRACT:** In order to evaluate the long-term behaviour of the engineered barriers in geological disposal sites for transuranic element-bearing (TRU) waste, an evaluation by numerical analysis is required. Although chemical and hydraulic/mechanical analyses have been conducted independently until now, essentially both type of phenomena occur simultaneously and produce synergistic effects. Therefore, we focused attention on the buffer (bentonite) engineered barrier and conducted a study of which involved incorporating hydraulic/mechanical phenomena into the chemical analysis of bentonite alteration. The simulations employed weakly-coupled chemical and hydraulic/mechanical effects to study the behaviour in one dimension.

The results showed that the dissolution of the montmorillonite is suppressed in the buffer section nearest the cement material. Moreover, in order to achieve a fully coupled analysis in future, the present study also identifies issues that need to be resolved.

**KEYWORDS:** engineered barriers, storage/disposal radioactive waste, bentonite buffer, alteration, batch, geomechanics.

In order to carry out long-term safety evaluation of an artificial barrier for a transuranic element-bearing (TRU) waste repository, we have examined analysis methods to reduce uncertainties in safety evaluations. We analysed the chemical alteration of the bentonite engineered barrier material in a previous study, but did not consider the hydraulic/mechanical modeling of the materials. The bentonite in a geological repository must maintain very low hydraulic conductivity by swelling, and the

mechanical and hydraulic properties of bentonite swelling may affect the evaluation of chemical alteration. In current research, the coupled analysis of hydraulic, mechanical and chemical (HMC) properties or thermal, hydraulic, mechanical and chemical (THMC) properties intended for the compacted bentonite have been examined (Xie *et al.*, 2006; Zheng *et al.*, 2010). However, there are few published papers concerning chemical analysis that have considered the mechanical effects on the alteration of the bentonite by the influence of hyperalkaline cement over a long term.

Therefore, in this paper, we add the effects of mechanical and hydraulics properties to the safety evaluation of conventional chemical alteration. In

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particular, we consider the effect of weak coupling by use of a coupled geochemical reaction/mass-transport code and a hydraulic/mechanical code.

The performance properties of bentonite relate to each other and there are many kinds of links. We consider part of these in this study and Fig. 1 shows the scope of our study. We considered four factors that are common parameters in the mechanical and chemical analysis among these links (upper part of figure). The lower part of the figure shows properties not considered in this study and show scope for the future studies. The model which we are using is straightforward and preliminary. However, we believe that we can identify potential problems by using this model.

COUPLED ANALYSIS METHOD

Normally, changes in the mechanical properties and the chemical alteration of bentonite take place

simultaneously. However, a fully coupled analysis method is very difficult to solve accurately and quickly. In a coupled analysis, the governing equations are non-linear and a solution cannot be guaranteed. Moreover, even if equations can be solved, the calculation convergence time may be very long.

Therefore, for this study, we used a weakly-coupled analysis in order to allow the following: (1) shortened simulation time, despite the lower accuracy of the solution, and (2) development of analytical tools identifying separate chemical and hydraulic/mechanical model behaviours. The geochemical code used here was PHREEQC-TRANS (2005) and the hydraulic/mechanical code was DACSAR-MP (Kanazawa, 2010). The coupled analysis method for the hydraulics, mechanics, and chemistry is shown in Fig. 2.

It is necessary to select an appropriate technique because there are various methods for coupled

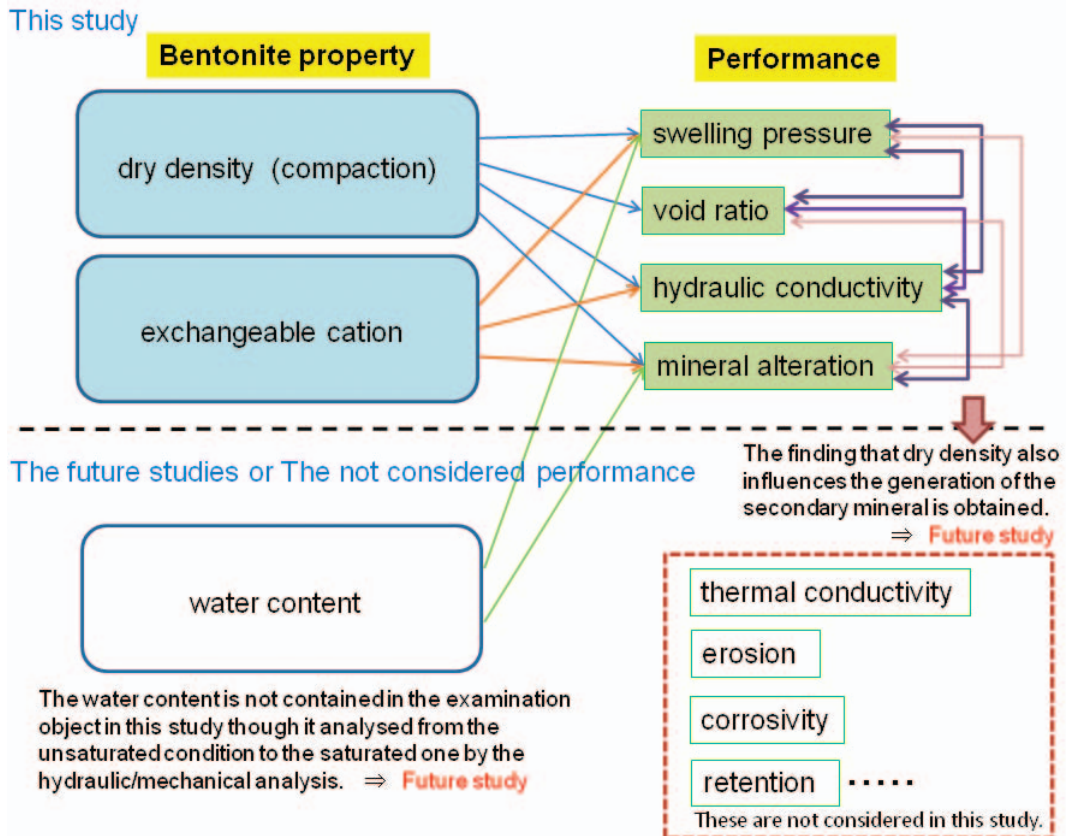


FIG. 1. Linkage of the property and the performance of bentonite.

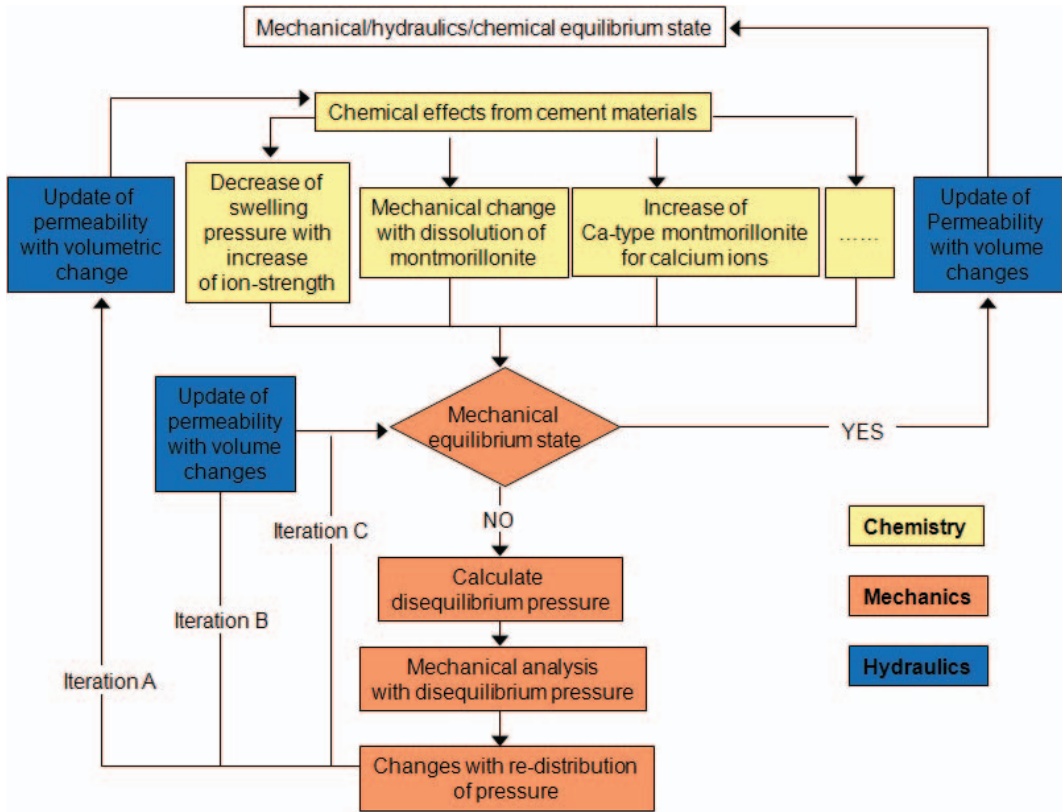


Fig. 2. Schematic diagram of the coupled hydraulic/mechanical/chemical analysis approach employed here.

analysis which range from weak coupling to strong coupling. A weakly-coupled method shortens the computation period but with less accuracy in the solution. Also, both tools for the chemical and hydraulic/mechanical analyses can be developed individually. On the other hand, a strongly coupled analysis provides no guarantee of obtaining a solution because non-linearity is expected to appear at the rule equation level. Even if a solution can be obtained, the strong non-linearity means that the time for solution convergence will be very long. One merit of this approach is that the solution is rigorous. However, if the convergent calculations are not completed, increasing the frequency at which data is exchanged in a weakly coupled analysis may attain a higher accuracy than that using a fully-coupled analysis. We therefore devised three methods that are shown in a schematic diagram of the coupled analysis used in the present study (Fig. 2). Iteration A addresses the re-distribution of stress that occurs during hydraulic

disequilibrium caused by chemical alteration. Iteration B computes the re-distribution of stress, and then the chemical analysis is conducted when the calculation attains a dynamic equilibrium. Finally, Iteration C is an analysis in which the hydraulic and chemical analyses are not conducted during the convergent calculation for computing the re-distribution of stress, but are conducted after dynamic equilibrium is attained. All methods, Iterations A, B and C, are classified as weakly coupled analyses. Iteration A is the strongest coupled analysis among these three methods and appears to take a longer computation period. It is necessary, therefore, to examine the differences in the numerical solutions by the respective differences in the coupled analytical approaches and to attempt to increase the accuracy and optimize the analytical period.

In this study, we evaluate the long-term behavior of the alteration of the engineered barriers predicted by use of Iteration C.

## ANALYSIS PARAMETERS AND A MODEL

In this study we selected four common parameters: (1) content of montmorillonite, (2) Ca exchange rate, (3) void ratio, and (4) dry density as shown in Table 1. The considered each parameter is described in the analysis as follows.

### Content of montmorillonite

The time change of the content of the montmorillonite in each mesh was calculated by a chemical analysis including the mass-transport, in consideration of the solution velocity. Because the relation between the swelling pressure and the density changes depends on the content of the montmorillonite (Kobayashi *et al.*, 2011), the results of the chemical analysis are reflected in the hydraulic/mechanical analysis.

### Ca exchange rate

The interlayer cation of Na-montmorillonite is exchanged from Na to Ca by  $\text{Ca}^{2+}$  eluted from the cement. The changes in relation between consolidation stress and void ratio which were used in the hydraulic/mechanical analysis was calculated based on the ratio of Ca-montmorillonite. The change in the hydraulic/mechanical characteristic according to the ratio of Ca-montmorillonite was estimated following Kobayashi *et al.* (2011).

### Void ratio

In the hydraulic/mechanical analysis, the density was redistributed with the change in the mechanical characteristic, taking into account the content of montmorillonite and the Ca exchange ratio. Both had been obtained from the result of the chemical analysis. This hydraulic/mechanical analysis suggested that the density (i.e. void ratio) distribution in this result was assumed to result from the chemical analysis. We further restarted the chemical analysis. The void ratio in response to the dry density was calculated by the following equation:

$$\text{Void ratio: } e = \rho_s / \rho_d - 1 \quad (1)$$

where  $\rho_s$ : the density of soil particle,  $\rho_d$ : the dry density.

$$e = \varepsilon / (1 - \varepsilon) \quad (2)$$

where  $\varepsilon$ : porosity (-).

### Dry density

The density (i.e. void ratio) distribution that had been obtained by this hydraulic/mechanical analysis result was reflected in that of a chemical analysis in the following way.

In the chemical analysis of bentonite alteration, the dry density changed with dissolution and precipitation of minerals in the bentonite. However, the change in the amount of the solid phases cannot be observed in the hydraulic/

TABLE 1. Parameters considered by both chemical and mechanical approaches (the arrows indicate receiving and passing).

Mechanical analysis	Chemical analysis
Montmorillonite content	Montmorillonite content
Ca type montmorillonite ratio	Ca type montmorillonite ratio
Void ratio	Porosity
Dry density	Dry density
Water content of double layer swelling	Component of pore water
Specific surface	Mineral density
:	:

TABLE 2. Dry density by hydraulic/mechanical analysis result after 1000 years (bottom buffer material).

Mesh no.	1 Concrete side	2	3	4	5	6	7	8 Waste side
Width of mesh $L_{bx}$ (m)	0.1150	0.1315	0.1460	0.1575	0.1705	0.1850	0.1715	0.1230
Dry density $\rho_{Mbx}$ (Mg/m <sup>3</sup> )	1.624	1.612	1.605	1.599	1.594	1.590	1.600	1.640
Average dry density $\bar{\rho}_{Mb}$ <sup>1)</sup> (Mg/m <sup>3</sup> )	1.6059							

mechanical analysis. Therefore we reset the dry density in the chemical analysis. We fixed the whole quantity of the solid-phase to reproduce the distribution of the dry density. There are two parts of the buffer material, the upper and the lower, in our conceptual model of the analytical system. The upper part is decompressed and the lower one is compressed. In order to reflect this phenomenon, each part should be allotted appropriately. The specific amount of the lower buffer material is shown in Tables 2 and 3. Table 2 shows the result of the hydraulic/mechanical analysis of the lower buffer material over 1,000 years. The value used for a chemical analysis was calculated based on this dry density distribution by the following

equations:

$$\bar{\rho}_{Mb} = \frac{A_{Mb}}{\sum_{x=1}^m L_{bx}} \quad (3) \text{ [in Table 2.1]}$$

$$\bar{\rho}_{Cb} = \frac{A_{Cb}}{\sum_{x=1}^m L_{bx}} \quad (4) \text{ [in Table 3.2]}$$

$$\bar{\rho}'_{Cb} = \bar{\rho}_{Cb} \times \frac{A'_{Cb}}{A_{Cb}} \quad (5) \text{ [in Table 3.3]}$$

$$\rho'_{Cbx} = R_{pMbx} \times \bar{\rho}'_{Cb} \quad (6) \text{ [in Table 3.4]}$$

TABLE 3. Dry density by chemical analysis result that reflected hydraulic/mechanical analysis after 1000 years (bottom buffer material).

Mesh no.	1 Concrete side	2	3	4	5	6	7	8 Waste side
Width of mesh $L_{bx}$ (m)	0.1150	0.1315	0.1460	0.1575	0.1705	0.1850	0.1715	0.1230
Dry density by chemical analysis $\rho_{Cbx}$ (Mg/m <sup>3</sup> )	1.606	1.604	1.597	1.593	1.588	1.584	1.588	1.619
Average dry density (Mg/m <sup>3</sup> )	Average dry density that considers hydraulic/mechanical analysis result $\bar{\rho}_{Cb}$ <sup>2)</sup> = 1.5958 $\bar{\rho}'_{Cb}$ <sup>3)</sup> = 1.5968							
Dry density set again $\rho'_{Cbx}$ <sup>4)</sup> (Mg/m <sup>3</sup> )	1.615	1.603	1.596	1.590	1.585	1.581	1.591	1.631

where

$A_{Mb} = \sum_{x=1}^m \rho_{Mbx} \times L_{bx}$ ; weight of lower part in hydraulic/mechanical analysis

$A_{Mu} = \sum_{y=1}^n \rho_{Muy} \times L_{uy}$ ; weight of upper part in hydraulic/mechanical analysis

$A_{Cb} = \sum_{x=1}^m \rho_{Cbx} \times L_{bx}$ ; weight of lower part in chemical analysis

$A_{Cu} = \sum_{y=1}^n \rho_{Cuy} \times L_{uy}$ ; weight of upper part in chemical analysis

$A'_{Cb} = (A_{Cu} + A_{Cb}) \times \frac{A_{Mb}}{A_{Mu} + A_{Mb}}$ ; weight that considers hydraulic/mechanical analysis

$R_{\rho_{Mbx}} = \frac{\rho_{Mbx}}{\rho_{Mb}}$ ; ratio of dry densities of individual mesh.

*Conceptual model of the analytical system*

We considered a one-dimensional horizontal cross-section of the repository for the conceptual model of the analysis system as shown in Fig. 3.

Figure 4 shows the process by which the bentonite is changed through chemical alteration and undergoes a dynamic change in its properties. First, the alkaline components such as  $Ca^{2+}$  and  $OH^-$  ions infiltrate from the cementitious material into the bentonite. Then, montmorillonite dissolution, precipitation of secondary minerals, and the production of Ca-type montmorillonite occur in the bentonite (C).

Re-equilibrium of pressure occurs in the bentonite because the production of Ca-type montmorillonite causes a change in the degree of swelling (H/M). Because the ions are moved by mass transfer,  $Ca^{2+}$  and  $OH^-$  ions infiltrate further into the bentonite. These steps are then repeated by iteration.

RESULTS AND DISCUSSION

We first performed a hydraulic/mechanical analysis from unsaturated moisture conditions to saturation. The hydraulic/mechanical analysis that was conducted until re-saturation showed that dry density had a heterogeneous distribution at the saturation point. We set this distribution as the

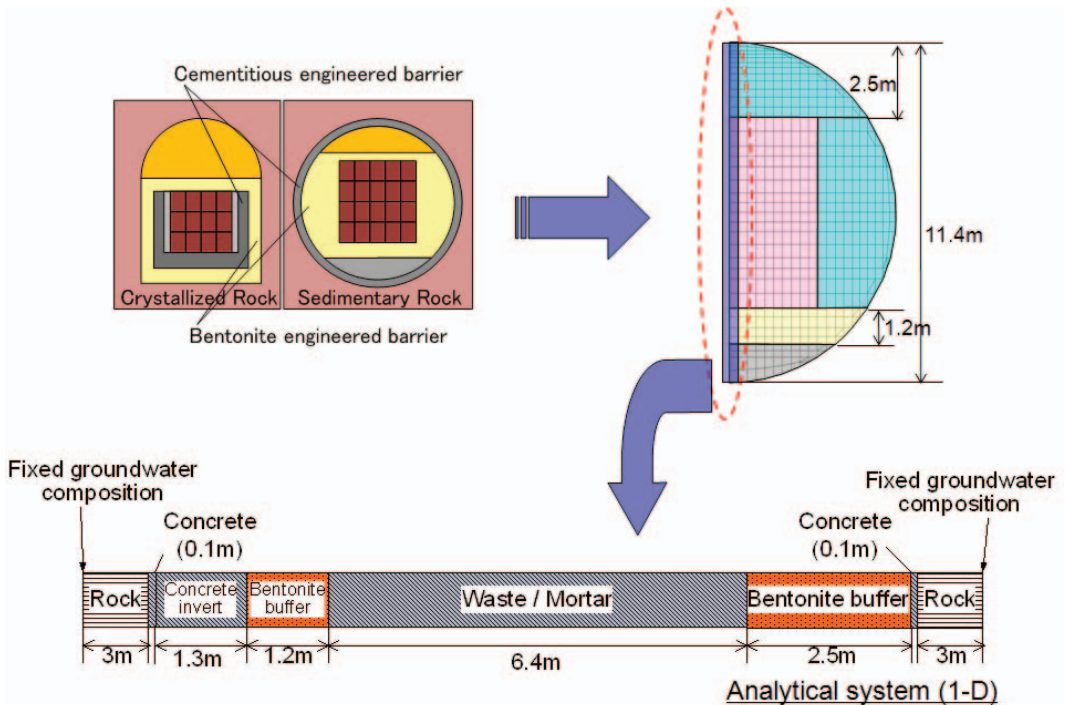


FIG. 3. Conceptual model of the analytical system, showing the distribution of engineered barriers (top left), a cross-section of the 2D analytical system (top right), and how this is transposed to a 1-D system (bottom).



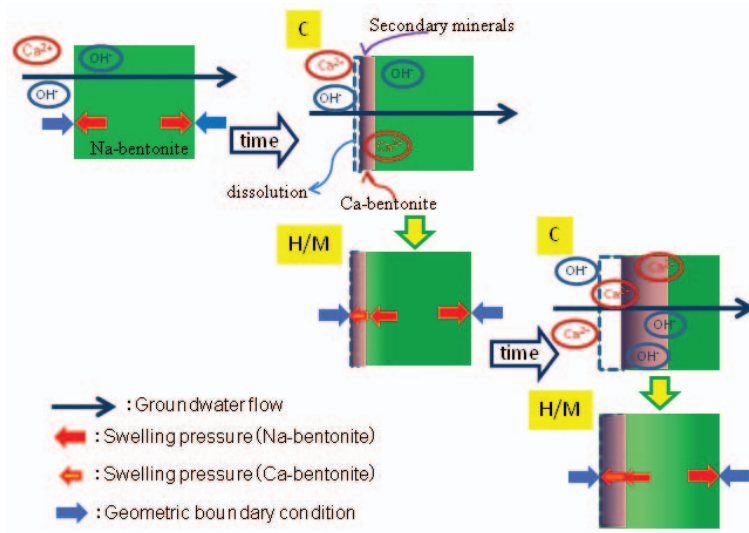


FIG. 4. Process of changes in the bentonite. See text for details.

initial condition for the chemical analysis. As a reference case, we also conducted another analysis that set a homogeneous distribution for the initial conditions.

The solid phase composition distribution is shown in Fig. 5 (initial) as the changes in analysis results after 2000 (Fig. 6) and 5000 (Fig. 7) years.

In each figure the results for the case of only chemical analysis (the reference case) in the left-hand column are presented; the results for the case of weakly coupled analysis are in the right-hand column.. The simulation period for these two analyses was set to 10,000 years, and data were applied to the chemical analysis and hydraulic/

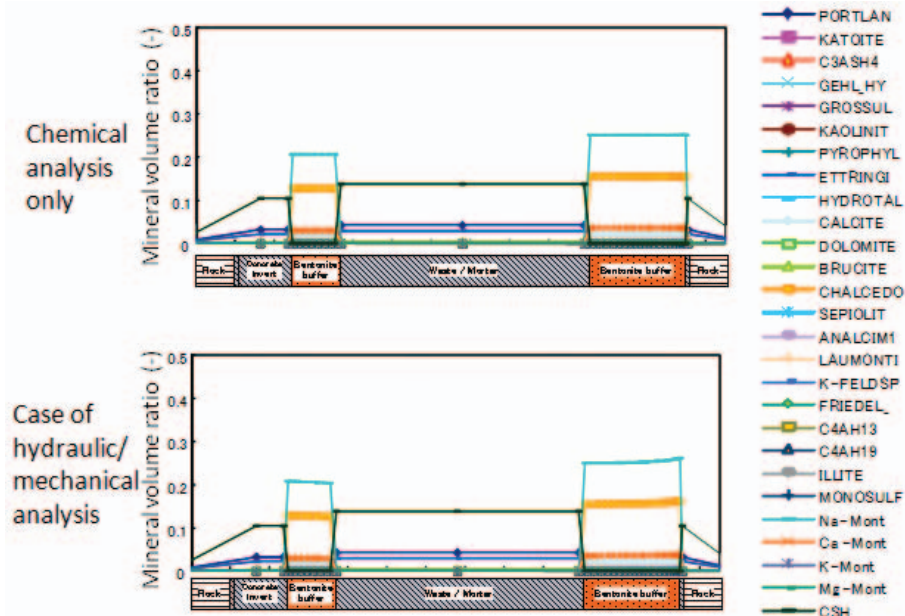


FIG. 5. Solid phase composition distribution at the initial setting.

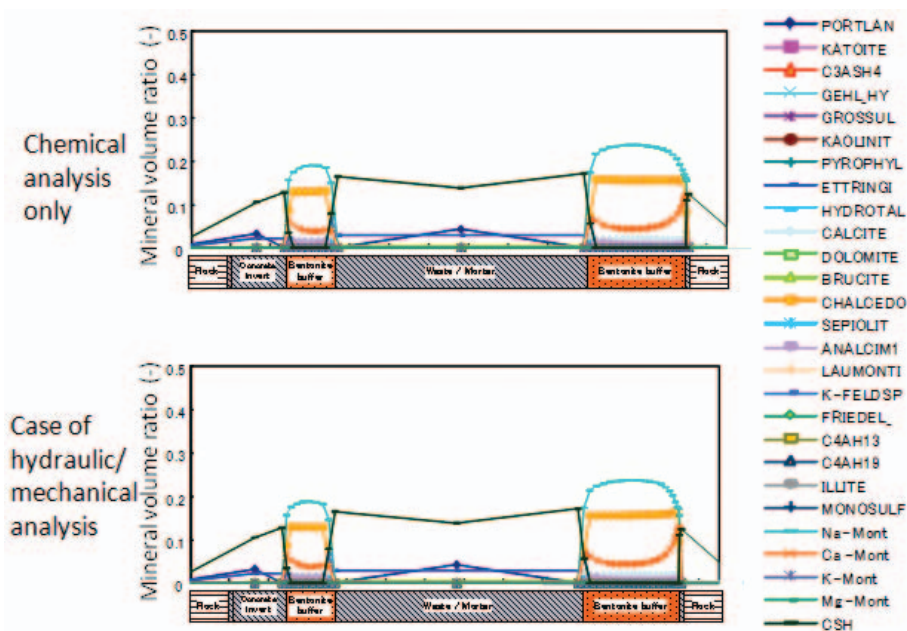


FIG. 6. Solid phase composition distribution after 2000 years.

mechanical analysis at 1000, 2000, 3000 and 5000 years. The amount of Na-type montmorillonite in the buffer decreases at the surface in contact with

the cementitious material, whereas the amount of Ca-type montmorillonite increases. A comparison of both cases shows few differences.

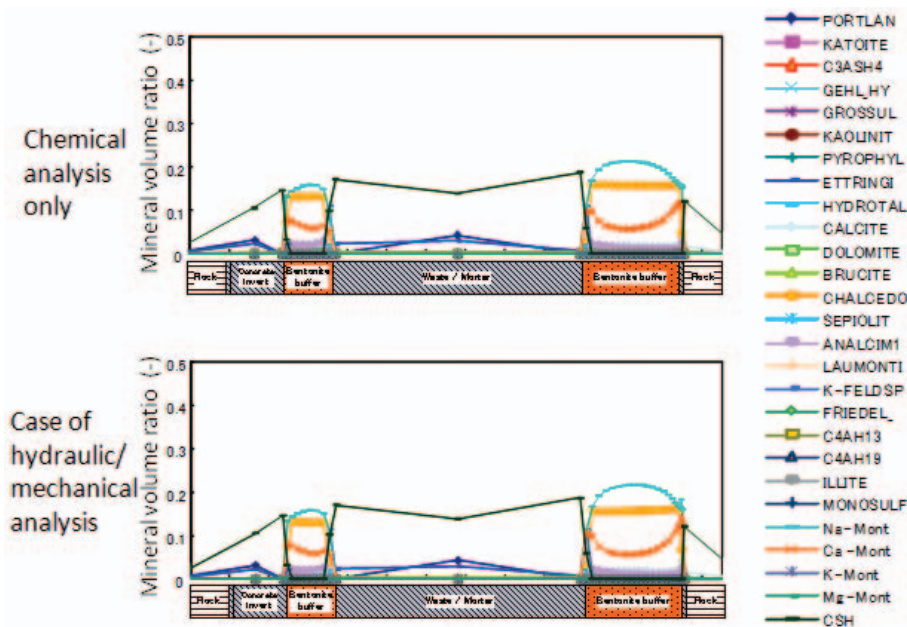


FIG. 7. Solid phase composition distribution after 5000 years.



The hydraulic conductivity of the buffer material was calculated by using the amount of montmorillonite, the ratio of Na-type montmorillonite, and the composition of the interstitial water included in the bentonite. The calculation is expressed as follows:

Hydraulic conductivity of bentonite

$$K = 10^{1.30C_i} \text{esme}^{3.48C_i} K_0$$

However,  $C_i \lesssim 10^{1.49\text{ESP}-1.0}$   
 $K \lesssim 10^{1.63\text{esme}-0.24} K_0$  and  $K \lesssim 10^{-5}$   
 $K_0 = (0.91-1.57\text{ESP}+2.00\text{ESP}^2) \times 10^{-13} \times \text{esme}^{7.44-5.69\text{ESP}}$ ;  $\text{esme} \lesssim 7.0$   
 $K_0 = (0.91-1.57\text{ESP}+2.00\text{ESP}^2) \times 10^{-13} \times 7.0^{7.44-5.69\text{ESP}} (\text{esme}/7.0)^{11.4}$ ;  $7.0 < \text{esme}$   
 However,  $K_0|_{\text{ESP} = 1} \lesssim K_0 \lesssim 10^{-5}$

$C_i$ : Equivalent ion concentration of pore water  
 ESP: Exchangeable Na proportion in bentonite  
 esme: Smectite void ratio

Figure 8 shows the change in hydraulic conductivity over time, as calculated by this expression. In the case of the non-coupled analysis

(the reference case), the distribution of hydraulic conductivity shows a gradual increase after about 3500 years at a location near the boundary with the cementitious material. On the other hand, in the case of the weakly coupled analysis, hydraulic conductivity remained small compared with the values from the reference case.

Figure 9 shows how the hydraulic conductivity changes over time for meshes that range from mesh 1 on the rock side to mesh 8 on the waste side. Mesh number 8 on the waste side had the largest value, and the hydraulic conductivity grew in the areas where it came into contact with the cementitious material. Also, the hydraulic conductivity increased smoothly in the case of chemical analysis only. In the other case in which mechanical behavior was also considered, the hydraulic conductivity shows inflection points because data are being passed into the model.

When performance as a buffer material is evaluated, the equivalent hydraulic conductivity is generally considered as an index. The equivalent hydraulic conductivity coefficient was calculated by the formula in Fig. 10. Figure 11 shows its

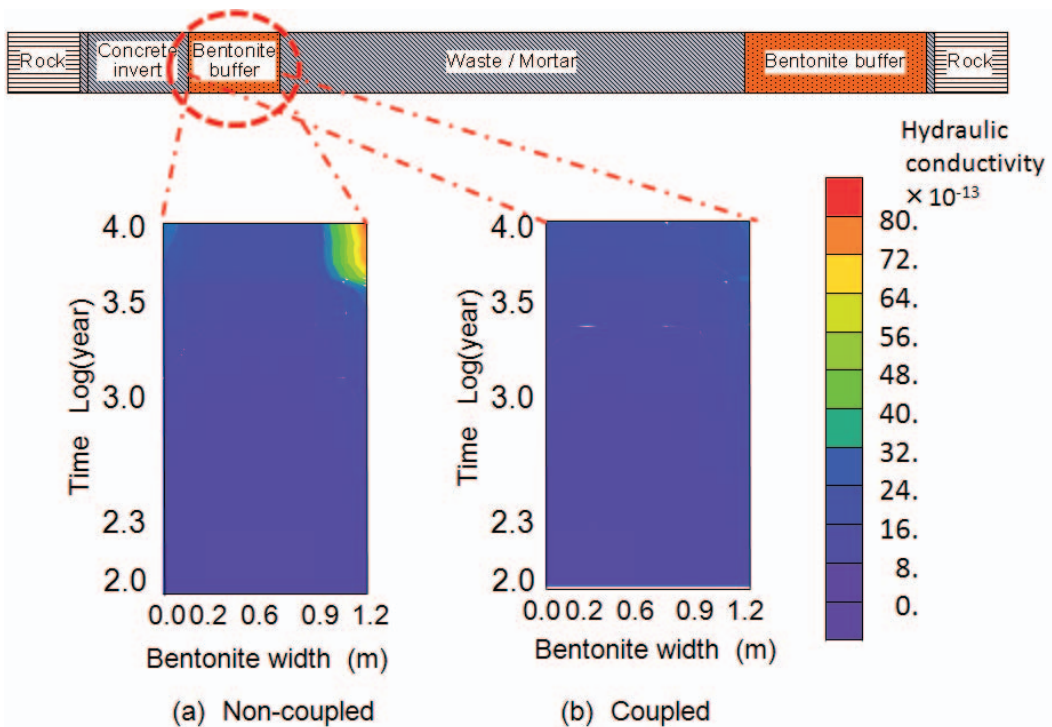


FIG. 8. Changes in the distribution of hydraulic conductivity over time.

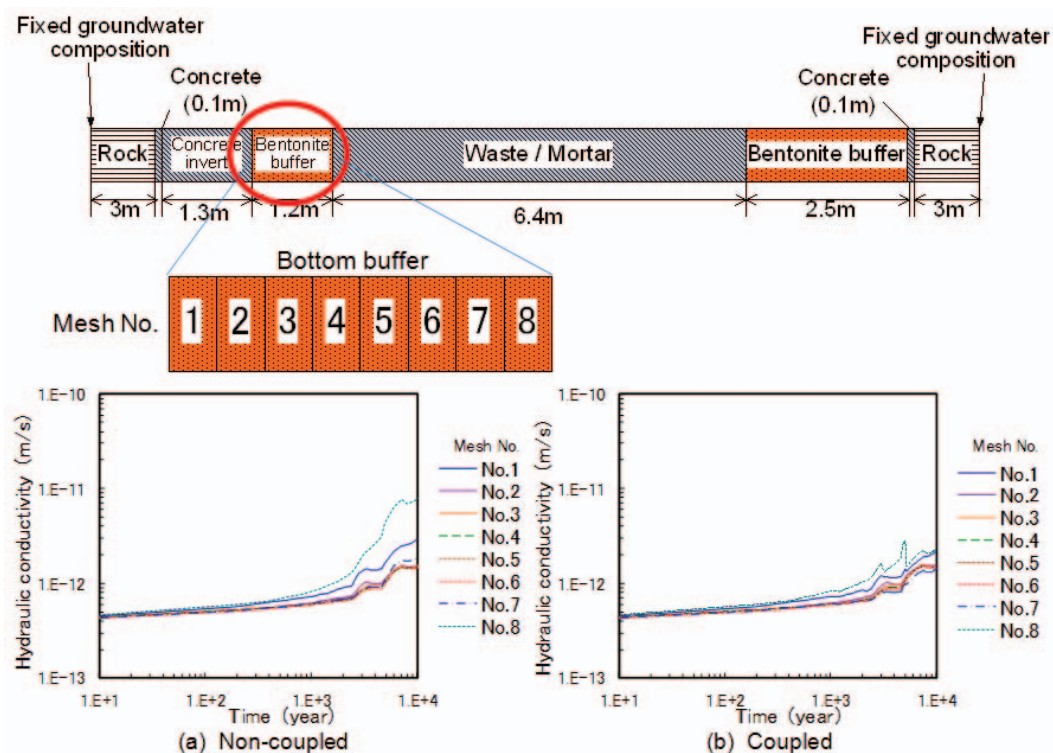
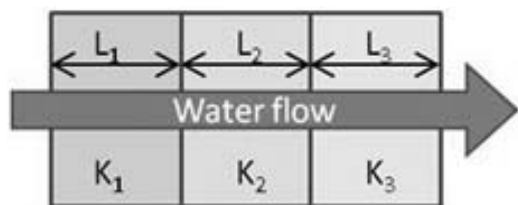


FIG. 9. Changes of hydraulic conductivity in each mesh.

evolution with time. The data represented by the dotted line in Fig. 11 were obtained from the case of chemical analysis only, and those represented by the solid line were obtained from the weakly coupled analysis. The two sets of data show no significant differences.



$$K_w = \frac{L_1 + L_2 + L_3}{L_1/K_1 + L_2/K_2 + L_3/K_3}$$

FIG. 10. Formula for computation of equivalent hydraulic conductivity coefficient.

### THE REALIZATION OF A FULLY COUPLED APPROACH

The evaluation of hydraulic/mechanical effects produced by chemical alteration of bentonite in the one-dimensional model presented in this study was small as stated above. However, it is necessary to examine the behaviour by using models in two or more dimensions in order to evaluate the uncertainty. Moreover, some problems were identified in this study, such as:

The basis for calculating the hydraulic conductivity differs between the chemical and hydraulic/mechanical models;

In the present hydraulic/mechanical analysis, it has not been considered that the soil grain density and the water density change when the montmorillonite alters;

Unsaturated conditions have not been considered in the chemical analysis;

For simulations in two dimensions, the speed of calculation in the chemical model is insufficient.

It would be necessary to solve the above problems in order to make advances in the

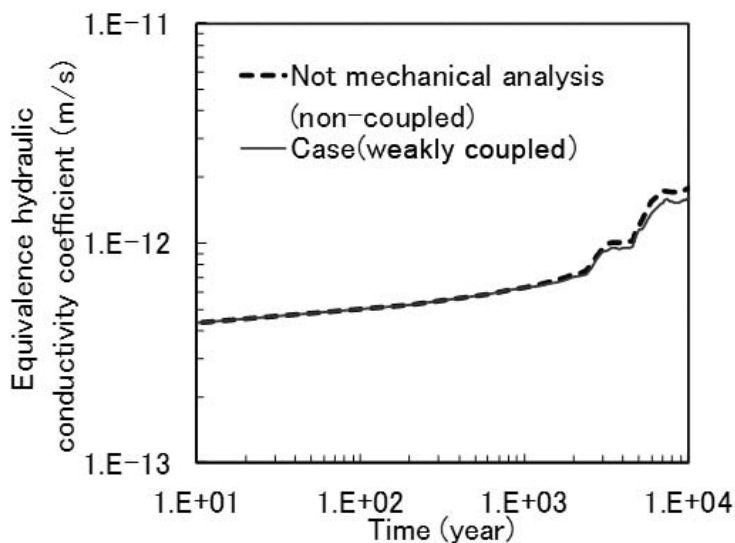


FIG. 11. Comparison of the equivalent hydraulic conductivity coefficient.

coupled analysis of chemical and hydraulic/mechanical processes. The first problem can be solved by setting a common coefficient. However, the effects will need to be examined. Addressing the second problem requires the types of mechanical changes be reviewed through a theoretical examination. The third and fourth problems are those that concern the details of the chemical model. New numerical approaches must be introduced or developed in order to solve these problems, although these do not appear to be achievable in the short-term.

Thus, to evaluate the performance of the engineered barrier in the long-term, it is generally recognized that more attention needs to be given to the synergistic effects of the chemical and hydraulic/mechanical processes. However, some difficult problems need to be solved in order to realize a fully-coupled simulation. It is therefore necessary to ascertain these effects using a more effective approach.

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#### REFERENCES

- Kanazawa S. (2010) *Mechanical evaluation of compacted soil structures based on mathematical modeling of unsaturated soils*. PhD thesis, Kobe University, Kobe-shi, Japan.
- Kobayashi I., Owada H. & Ishii (2011) Hydraulic/mechanical modeling of smectite materials for HMC analytical evaluation of the long term performance of TRU geological repository. *Proceedings of the 14<sup>th</sup> International Conference on Environmental Remediation and Radioactive Waste Management ICM2011*.
- Xie M., Bauer S., Kolditz O., Nowak T. & Shao (2006) Numerical simulation of reactive processes in an experiment with partially saturated bentonite. *Journal of Contaminant Hydrology*, **83**, 122–147.
- Zheng L., Samper J., Montenegro L. & Fernández A.M. (2010) A coupled THMC model of a heating and hydration laboratory experiment in unsaturated compacted FEBEX bentonite. *Journal of Hydrology*, **386**, 80–94.