Technical report

Safety Function of Cementitious Materials and the Analytical Assessment of Long-Term Evolution of Cement-Bentonite Interface for Geological Disposal in Japan

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Abstract
Cementitious materials used in geological disposal repositories are expected to have various functions for construction, operation and closure of the high-level radioactive waste (HLW)/TRans-Uranic (TRU) waste repositories and they also have functions for safety. In the long term after closure of the repositories, cementitious materials are expected to reduce the release of radionuclides from the waste. However, the expected performance of cementitious materials may decrease in the long term because of their gradual dissolution/alteration. In addition, there is a concern that the high pH groundwater due to alkaline ions leached from cementitious materials may degrade the safety functions of other components (buffer, backfill, host rock) of the repositories. Therefore, in order to understand how the expected safety functions of the cementitious materials and other components can be achieved in the post-closure period, NUMO carried out the analytical evaluation of the evolution of each component. The results showed that most of the cementitious materials and other components will remain during a long-term post-closure period. At present, we are aiming to improve the reliability of the analytical model and to develop a more realistic nuclide migration model that reflects the effect of cementitious materials on reducing mass transfer.

1. Introduction

In Japan, it is legally defined that long-lived radioactive wastes are disposed in host rock at depth greater than 300 m below surface by the Designated Radioactive Waste Final Disposal Act (Enacted at 2000). The wastes for geological disposal include high-level radioactive waste resulting from reprocessing spent fuel and TRU wastes containing over a certain amount of long-lived radionuclides, produced from the operation and de-commissioning of spent fuel reprocessing facilities and MOX fuel fabrication plants (NUMO 2021a). Because the waste will remain highly radioactive for a very long time, it is necessary to isolate it from the human living environment for over tens of thousands of years. Nuclear Waste Management Organization of Japan (NUMO) is responsible for implementing geological disposal of HLW and TRU.

In order to protect humans and the environment from the potential hazards of HLW and TRU wastes, safety functions are expected to cementitious materials in the repositories. In both HLW and TRU repositories, mechanical stability of tunnels is expected during construction and operational phase. In the TRU repository, radiation shielding and prevention of leakage of radionuclides from the waste are also expected during construction and operational phase. Cementitious materials are expected to reduce releases of radionuclides by restricting of groundwater flow and sorption of radionuclides onto these materials in the TRU repository after closure. The state of the cementitious materials will change as a result of the interaction with groundwater and with the other engineered barrier components after closure of the repository. In addition, the change of groundwater chemistry in contact with the cementitious materials will affect the state of the engineered barriers. Therefore, it is important to understand long-term evolution of the cementitious materials and the components affected by the cementitious materials in order to show how the safety functions of the repositories perform after closure.

This technical report outlines the role of cementitious materials used in the HLW and TRU repositories and presents the results of an analytical evaluation of evolution of the cementitious materials and the bentonite buffer. In addition, this report shows the current status of NUMO’s R&D activities related to the cementitious material.

Most of the content of this paper is based on the ‘NUMO Safety Case’ published in February 2021 (NUMO 2021b). At the present moment, repository site has not been selected, so the repository design described in this technical report is an example and may change according to the progress of the repository project.

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2. Overview of geological disposal

2.1 Basic concept of geological disposal
The objective of geological disposal of radioactive waste is to provide isolation and containment of the radionuclides in the waste from the biosphere (IAEA 2011). The basic concept related to “isolation” is to isolate radioactive waste from the human living environment. The long-term stability of the geological environment is evaluated by extrapolating site-specific geological evolution from the past to the present to avoid future natural phenomena such as volcanic activity, fault movement and significant uplift/erosion which disturb around the repositories. The depth of disposal is also a factor of “isolation,” and disposal at sufficient depth is required to avoid the influence by conventional civil engineering activities at the time when there is no longer institutional control of the disposal site. In addition, the geological environment, selected by excluding areas containing mineral resources that might be exploited in the future, should contribute to reduction of the risk of accidental anthropogenic perturbations. As for “containment,” the basic concept is long-term containment of radioactive materials in deep underground and retardation of radionuclide migration. Containment of radionuclides are achieved by the robust engineered barriers which consist of waste package, container, filler, buffer, backfill, and plugs. And retardation of the radionuclide migration is also provided by the natural barrier with low hydraulic conductivity and suitable geochemical conditions (e.g., pH higher than 4.8 and carbonate chemical species concentration less than 0.5mol/dm$^3$ in the groundwater). The release of radioactive materials is limited by the combination of the engineered barriers and natural barrier properties (NUMO 2021b).

2.2 Overview of radioactive wastes
Vitrified waste is produced by vitrifying high-level radioactive liquid waste with borosilicate glass. Borosilicate glass has a cancellous chemical structure which is capable of incorporating different sizes constituent homogeneously and has a stable property due to extremely low dissolution rate. Therefore, borosilicate glass delays the time of release of radionuclides to groundwater.

TRU wastes are low-level radioactive wastes generated mainly in the reprocessing process, which are sealed in drums or other containers or solidified with mortar. TRU wastes include various types of wastes such as metals, mortars, asphalts, etc., which differ in their forms and radionuclides contained. They are classified into four groups, Group 1 to Group 4 (NUMO 2021b), depending on their thermal output and chemical characteristics. Furthermore, Group 4 is divided into two types from the viewpoint of thermal output, hence there are five types of TRU wastes (Table 1 shows the details of the waste Group1 to Group4). The TRU wastes are stored together in metal containers, and the gaps in the containers are filled with mortar. The waste, the metal container, and mortar filler are referred to as the “waste package” as a whole.

2.3 Overview of repositories
Components of the repositories are classified into engineered barriers, underground facilities, and surface facilities, shown in Fig. 1. Underground facilities constructed in the rock mass at depths of 300 m or more consist of a series of tunnels for various purposes, such as access tunnels (ramps/shafts) from the surface to the underground facilities, disposal tunnels for embedding waste packages such as vitrified waste with other engi-

Table 1 The details of the waste Group1 to Group4 (NUMO 2021b).

| Group | Description |
|-------|-------------|
| Group1 | Group (Gr.) 1, the silver adsorbent used to capture off-gas iodine, is immobilized with mortar and has extremely low heat output. It contains a large amount of radioactive iodine (I-129), a long-lived nuclide with high solubility and low sorption. |
| Group2 | Gr.2 comprises compressed metal cladding stripped from SF (hulls and ends), sealed in a stainless steel canister. Its thermal output is relatively high and contains a large amount of radioactive carbon (C-14), which is a relatively long-lived nuclide with high solubility and low sorption. |
| Group3 | Gr.3 results from the solidification of liquid waste containing nitrate generated during reprocessing. It is conditioned in a matrix of bitumen or mortar and has a relatively low heat output. It contains nitrate, which can affect the engineered barrier and host rock properties. |
| Group4 | Gr.4 consists of miscellaneous wastes generated during reprocessing and MOX fuel fabrication, which are solidified or encapsulated in mortar, etc. There are two types of wastes: one is relatively low thermal output (Gr.4L) and the other is relatively high thermal output (Gr.4H). |

Fig.1 Image of underground facility (NUMO 2021b, English translation is by the author).
3. Cementitious materials in underground facility

An overview of the components of underground facility is shown in Figs. 2(a)-2(f). The functions of the components made of cementitious materials in the underground facility are shown in Table 2.

As shown in Table 2, some components of cementitious material are required to have safety functions during operational period, and some are required to have functions after closure of the repositories. The former components should be designed based on conventional design requirements as engineering structure. The latter components should be evaluated how their state evolves with time and the results should be used in the long-term safety assessment. Cementitious materials have potentials to degrade performance of other engineering barriers such as buffer and natural barrier, due to high-alkaline components leaching from cement. Therefore, it is critically important in the long-term safety assessment to evaluate the long-term impact on
engineering barriers and natural barriers causing by cementitious materials. Since the performance of natural barriers depends on the geological and environmental conditions of the site, there is great uncertainty in the performance of natural barriers. On the other hand, the performance of the engineered barriers can be designed to have the required performance in accordance with the site conditions. Thus NUMO put priority on the technological development of the engineered barriers (Masuda et al. 2015). Therefore R&Ds on evaluation of impact on buffer by cementitious materials are of priority. In the next section, an example of evaluation of cement-bentonite interaction for TRU waste repositories is shown.

### 4. Analytical evaluation of evolution of cementitious materials and buffer during a long-term post-closure period

This section introduces the analysis conducted to understand the long-term evolution of cementitious materials and buffer. The objective of the analysis is to evaluate the expected safety functions of cementitious materials and buffer.

Note that this section is based on R&D results as described in Supporting Report 6-8 of the NUMO Safety Case (NUMO 2021b).

### 4.1 Long-term condition change of cementitious materials and buffer

Cement-bentonite interaction can be summarized as follows based on a previous study (Yokoyama et al. 2006).

When cementitious materials come into contact with groundwater, the alkaline components (sodium and potassium) contained in the cement are leached to form pore water of around pH 13. When the pore water reaches buffer, the buffer minerals start to dissolve and precipitation of secondary minerals. After the alkaline component in the cement is completely dissolved, the dissolution of portlandite \((\text{Ca}_2\text{O}_6\text{Si}_2\text{O}_{10}2\text{H}_2\text{O})\) begins. Then, the pH of the cement leachates falls to about 12.5, with equilibrium to portlandite. Calcium leached to the pore water exchanges with the interlayer cations of montmorillonite. When cementitious materials come into contact with groundwater, the alkaline components (sodium and potassium) contained in the cement are leached to form pore water of around pH 13. When the pore water reaches buffer, the buffer minerals start to dissolve and precipitation of secondary minerals. After the alkaline component in the cement is completely dissolved, the dissolution of portlandite \((\text{Ca}_2\text{O}_6\text{Si}_2\text{O}_{10}2\text{H}_2\text{O})\) begins. Then, the pH of the cement leachates falls to about 12.5, with equilibrium to portlandite. Calcium leached to the pore water exchanges with the interlayer cations of montmorillonite. Once the portlandite is completely dissolved, the incongruent dissolution of calcium silicate hydrate \((\text{C-S-H})\) begins. Dissolution of the buffer minerals and precipitation of secondary minerals continue while these reactions occur. It is suggested to take several hundred thousand years to complete the dissolution of portlandite from the calculation based on the assumption that the dissolution proceeds in
proportion to the amount of groundwater flowed through the cementitious materials (Atkinson 1988). As a result, the porosity, pore structure and the mass transport properties of the cementitious materials and buffer will change. For example, the porosity decrease due to precipitation of minerals will cause the decrease of effective diffusion coefficient (De) and hydraulic conductivity (Kw). On the other hand, the porosity increase due to dissolution of minerals will cause the increase of De and Kw. Figure 3 shows a schematic diagram of the above reactions.

4.2 Analytical System

This technical report presents the analysis cases conducted in NUMO Safety Case (NUMO 2021b) for TRU waste repository in Neogene sedimentary rocks, where the largest amount of cement is used in the TRU repositories.

The cross section of the Group 2 waste disposal tunnel was used for the analysis as a representative of Groups 1, 2, and 4H, which are similar in shape. The analysis system is shown in Fig. 4. This is a one-dimensional analytical system for the vertical section of the tunnel in order to evaluate the interactions between the shotcrete, invert concrete, buffer, concrete cell, waste form, filler within waste package, filler between waste packages, buffer and backfill (the red box in Fig. 4). In the green box, waste form, filler within waste package and filler between waste packages are collectively referred to as “waste, mortar” here. Both ends of the lower buffer are in contact with cementitious material: the invert concrete and the concrete cell. For the upper buffer, a bottom end is in contact with cementitious material: the filler between waste package and the concrete cell. As an initial condition, all the components are saturated. Boundary conditions of the cementitious materials in contact with the host rock were established to express the groundwater flow parallel to the disposal tunnel axis, which is called mixing cells. Groundwater was assumed to flow into the mixing cell at a constant flux. Alteration of the host rock may be involved in this reaction, but chemical reactions in the mixing cells were not considered in this analysis. Mass transport within the tunnel and between the mixing cell and the tunnel is considered as diffusion only because groundwater flow is slow in deep underground, and low permeability is kept by the buffer and backfill.
The analysis code was QPAC (Quintessa 2013), which can solve the coupling of chemical reaction and mass transport. The design specifications of NUMO Safety Case (NUMO 2021b) were applied for setting the thicknesses, densities, porosities and initial mineral compositions of cementitious materials, buffer, and backfill. The groundwater chemistry was set based on the nationwide data because the repository site has not been selected in Japan.

The chemical reaction model and parameters, such as the reaction rate and the reactive surface area of minerals, were referred to the previous study for the Japanese geological disposal system with the thermodynamic database developed in Japan. The analysis considered the change in diffusion in response to the porosity changes with the dissolution and precipitation of minerals.

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### 4.4 Analysis results and discussion

Figure 5 shows the results of TRU waste Group 2 under the condition of Neogene sedimentary high Cl− groundwater.

In the very close region (a few centimeters) near the cement-bentonite boundary, the dissolution of portlandite in the cement proceeds due to the influx of relatively low pH (7-9) bentonite porewater with cement porewater. After several thousand years, the portlandite is completely dissolved and the CSH begins to dissolve. On the other hand, in the further inner region, although the dissolution of portlandite progresses gradually, even after 100000 years, the portlandite does not completely dissolve and the dissolution of C-S-H does not occur. In addition, the present analysis assumes that the initial sulfur in the cement exists as monosulfate, but it changes to ettringite and Friedel’s salt due to the thermodynamic stability in pore water chemistry.

In the waste package infills, C-S-H which contributes to sorption of radionuclides was partly dissolved and a considerable amount of it remained after 100000 years. Therefore, it was expected that radionuclides are captured by waste package infill for Groups 1, 3, and 4L. Note that the sorption was not expected in Groups 2 and 4H because the heat generation from these wastes may alter the cement hydrate and reduce the sorption performance, although the heat generation of the wastes was not taken into account in this analysis.

In the buffer, montmorillonite were dissolved by high pH pore water caused by cementitious materials. This reaction caused the precipitation of zeolite, such as phillipsite and hydrotalcite, and calcite. The area where the residual amount of montmorillonite was less than 10 vol.% was limited in 5 cm from the boundary with the cementitious material. As a result, more than 80% of the

| Temperature | Considering the reliability of the thermodynamic database, temperature was set at 25°C. |
| Reactive surface | The value used in the previous study was used (JAEA 2015a). |
| Thermodynamic database | JAEA-J-TDB v1.0 (JAEA 2017) |
| Mineral composition | The mineral composition of buffer and backfill were according to the mixing ratio of bentonite and silica sand depending on the design specifications. For concrete, ordinary Portland cement is used and the mineral composition was according to the amount of water, cement and aggregate. (Section 4 of NUMO Safety Case (NUMO 2021b)) Note that the less reactive minerals of bentonite and the aggregates of cementitious materials were considered as non-reactive in this analysis. Therefore, in the analysis results shown in Fig. 5, the sum of each mineral’s volume fraction is not 100%. |
| Cation exchange capacity and ion exchange selectivity coefficient of buffer | The cation exchange capacity and ion exchange selectivity coefficients referred to Oda et al. (2001), Kamei et al.(1999) and Suzuki et al.(1992). |
| Secondary minerals | Multiple scenarios for mineral alteration were used based on the cases of natural high-alkaline environments (JAEA 2015b). |
| Kinetic rate | The dissolution formula for montmorillonite was based on the Marty equation (Marty et al. 2015), which expresses the rate as a function of pH. The precipitation formulae for secondary minerals were based on SIT model with reference to JAEA (2015b). |
| Diffusion coefficient | The effective diffusion coefficient of the buffer assumed to be changed due to the amount of montmorillonite and the porosity. The formula considering the smectite content and porosity (Mihara and Sasaki 2005) was used. For cementitious materials, the formula regarding to porosity (Mihara and Sasaki 2005) was used. |
| Porosity | The initial porosities of cementitious materials, buffer and backfill were calculated from the mineral compositions and the densities. When the porosity becomes very small due to the precipitation of secondary minerals, the effective diffusivity becomes small, and the analysis does not converge. Therefore, a lower limit of the porosity was set as the analytical limitation. |
| Groundwater | Since repository site has not yet been identified, model water chemistries of low Cl− and high Cl− groundwater were established for three rock types, which were plutonic rock, Neogene sedimentary rock, and Pre-Neogene sedimentary rock, using national scale groundwater chemistry data (NUMO 2021b). |
montmorillonite remained in more than 20 cm away from the interface even after 100000 years. Since the altered range remains in a limited area from the interface between cementitious materials and bentonite, it is considered that the decrease in density as the buffer and the decrease in swelling pressure can be ignored.

The precipitation of secondary minerals at the buffer/cement interface suppressed the mass transfer between cementitious materials and buffer and resulted in a large amount of montmorillonite remaining in the buffer. In laboratory tests and underground laboratories, it has been reported that calcite and C-S-H precipitated at the interface between cement and clay, resulting in porosity reduction (Cuevas et al. 2016; Fernández et al. 2016, 2017; Nakarai et al. 2021). The secondary minerals identified in the analysis, such as C-S-H and zeolite, is consistent with several natural case studies where clay minerals were in contact with highly alkaline groundwater for a long period (Oda et al. 2005). Moreover, the long-term stability of the buffer was shown in other analytical studies using different analysis codes and thermodynamic data (FEPC and JAEA 2005; Gaucher and Blanc 2006; RWMC 2013; Oda et al. 2013). The results showed that most of the cementitious materials and buffer remain for 100000 years after closure. Based on this result, one of the bases to argue that safety functions of cementitious materials and buffer will be maintained after closure was achieved.

Fig. 5 Temporal transition of mineral composition for each component of TRU waste Group 2 under the condition of Neogene sedimentary high Cl− groundwater (NUMO 2021b).
5. Future issues and latest initiatives

This technical report gives an overview of concept of geological disposal and shows the components cementitious materials are applied in the underground facility with their safety functions. In safety assessment for geological disposal, it is important not only to ensure the safety functions of the cementitious materials themselves, but also to consider the impact on other engineered barriers caused by highly alkaline components from the cementitious materials. As an example of such impact evaluation, reactive transport modeling on cement-bentonite interaction is shown.

Cement-bentonite interaction is generally consistent with laboratory experiments and observation of natural systems. Even though, there are still some uncertainties regarding the scenarios of secondary mineral occurrences leading to clogging and the mass transport properties of the clogging zone. Also, the analysis results have uncertainties because the analysis model includes settings based on several hypotheses for mass balance and analysis conditions. Therefore, to improve reliability of the assessment, NUMO conducts long-term laboratory tests (Fig. 6) to acquire the data on secondary minerals and mass transport properties of the altered zone to improve analysis model. The tests will be designed to simulate the state of cement-bentonite boundary expected in the repository as closely as possible. Nevertheless, uncertainties remain, such as the observable secondary minerals are limited due to limitation of test period. Future studies should analyze multiple analysis cases that take into account the uncertainties existing in the analysis model and parameters, and to combine multiple lines of evidence such as observation of natural systems.

The migration behavior of radionuclides in the cementitious materials was modeled based on a conservative simplification that ignores the migration of radionuclides near the waste package, taking into account the uncertainty of the evolution of the cementitious materials and the migration behavior of radionuclides in the cementitious materials over a long period after a repository closure (NUMO 2021b). Radionuclides are instantaneously mixed uniformly in the region from the waste package to the concrete cell when migration starts in the model. In other words, the current model does not take into account the pore structure in the cement, its long-term state evolution, and nuclide migration pathways. Therefore, NUMO is currently working on acquiring the migration behavior data of radionuclides in cementitious materials by laboratory experiments and in-situ experiments (Fig. 7) at the underground laboratory in Switzerland in order to improve the radionuclide migration model in cementitious materials (NUMO 2020).

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