Research Paper

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A Study About Radionuclides Migration Behavior in Terms of Solubility at Gyeongju Low- and Intermediate-Level Radioactive Waste (LILW) Repository

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A safety assessment of radioactive waste repositories is a mandatory requirement process because there are possible radiological hazards owing to radionuclide migration from radioactive waste to the biosphere. For a reliable safety assessment, it is important to establish a parameter database that reflects the site-specific characteristics of the disposal facility and repository site. From this perspective, solubility, a major geochemical parameter, has been chosen as an important parameter for modeling the migration behavior of radionuclides. The solubilities were derived for Am, Ni, Tc, and U, which were major radionuclides in this study, and on-site groundwater data reflecting the operational conditions of the Gyeongju low and intermediate level radioactive waste (LILW) repository were applied to reflect the site-specific characteristics. The radiation dose was derived by applying the solubility and radionuclide inventory data to the RESRAD-OFFSITE code, and sensitivity analysis of the dose according to the solubility variation was performed. As a result, owing to the low amount of radionuclide inventory, the dose variation was insignificant. The derived solubility can be used as the main input data for the safety assessment of the Gyeongju LILW repository in the future.

Keywords: Radioactive waste disposal facility, Safety assessment, Geochemical parameter, Solubility

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1. Introduction

Securing the safety of the radioactive waste repository is essential owing to the potential hazard from radioactive waste. The possible migration of radionuclides from the repository in the long-term time scales to the nearby ecosystem is inevitable and the inability of conducting experimental tests that can cover this time scale will require a simulation study to minimize this migration and ensure the safety concept of the designed repository. Especially understanding the migration behavior of radionuclide is crucial because radionuclides can migrate to the surroundings under various geochemical interactions in the natural groundwater systems. Solubility is one of the key geochemical parameters that can describe the migration/retardation process of radionuclides. Solubility can be affected significantly by the groundwater conditions and geochemical environment, so it is crucial to derive the solubility by applying the groundwater and geochemical conditions that reflect site-specific characteristics of the repository. South Korea's first radioactive waste repository was constructed and started operation in 2015. The first phase of the radioactive waste disposal facility is under operation now and construction for the next phases, the second, third, and fourth phase disposal facilities are planned in sequence. Radioactive wastes in South Korea are classified as high-level waste (HLW), intermediate-level waste (ILW), low-level waste (LLW), very-low-level waste (VLLW), and exempt waste (EW) according to the Nuclear Safety and Security Commission's Notice 2017-65. The radioactive waste will be disposed of at an appropriate disposal facility depending on the level of contamination. Currently, the Gyeongju LILW repository does not include radioactive waste from the nuclear power plant (NPP) decommissioning to stock estimation. However, if the decommissioning of Kori Unit 1 progresses, various levels and a large amount of radioactive waste will be generated and need to be disposed of at the Gyeongju LILW repository. Therefore, research related to the safety assessment of the Gyeongju LILW repository which includes NPP radioactive wastes is an urgent and important task. Studies on the solubility of the Gyeongju LILW repository are being conducted [1, 2]. In addition, a few studies are being actively conducted on the effect of solubility to a domestic environment [3-8]. Among the studies related to solubility, a study about solubility that reflects the operational conditions of the disposal facility has not been reported. Therefore, in this study, the solubility of the interested radionuclides to the groundwater system of the Gyeongju LILW repository was investigated. A program can simulate geochemical calculation, PHREEQC code was used for this purpose. PHREEQC code is a computational code developed by the U.S. Geological Survey [9]. This code is based on the ion-association aqueous model and generally used to calculate hydrochemical behavior such as solubility [10]. The groundwater system of the Gyeongju LILW repository was established by using actual data of the first-phase disposal facility operation period. The solubility derived in this study is a site-specific solubility that can reflect the operational conditions of the Gyeongju LILW disposal facility and can be used as a main geochemical data for deriving site-specific radiation dose results for safety assessment.

2. Input data for PHREEQC code

2.1 Thermodynamic database

It is known that radionuclides are interacting with the minerals in the strata during the migration and the migration of radionuclides can be significantly influenced by the chemical properties of the aqueous solution. Therefore, for successful evaluation of the migration behavior of radionuclides, it is crucial to understand the chemical properties of aqueous solutions such as pH, redox potential, and ionic strength. Besides, to evaluate the complicated and various chemical behaviors among numerous radionuclides in the groundwater, computational code that can simulate speciation, reaction-path, advective-transport, and inverse geochemical calculation should be used. Therefore, in this

Table 1. Thermodynamic databases analyzed in this study

Thermodynamic database	Development agency	Release date of latest version	Number of Radionuclides
TDB of JAEA [11]	Japan Atomic Energy Agency	2019.03	129
ThermoChimie of Andra [12]	French National Radioactive Waste Management Agency	2018.09	125
TDB of Nagra/PSI [13]	Laboratory for Waste Management of the Nuclear Energy and Safety Research Department of the Paul Scherrer Institute and National Cooperative for the Disposal of Radioactive Waste	2015.06	85

study, PHREEQC computational code was adopted. For the computational code, it is necessary to secure a chemical and thermodynamic database as input data, for this study, various thermodynamic databases from several institutes around the world were secured and analyzed. The Japan Atomic Energy Agency (JAEA) has established an independent thermodynamic database (TDB) for the safety assessment of radioactive waste repositories. This database is called TDB of JAEA and is continuously maintained and updated. The TDB of JAEA considered reactions between groundwaters, cement, clay, and minerals under the radioactive waste repository site condition. The TDB of JAEA was first developed for use as the basis for the safety assessment of radioactive waste disposal facilities in Japan. Also, it is being continuously improved and updated to reflect the additional collected or potential future data [11]. In addition to TDB of JAEA, ThermoChimie which is the thermodynamic database of the French National Radioactive Waste Management Agency (Andra) [12], and TDB of Nagra/PSI were considered [13]. After analyzing the thermodynamic database of JAEA, Nagra/PSI, and Andra, the TDB of JAEA was selected as the input database of the PHREEQC computational code. Because TDB of JAEA contains the most extensive radionuclide database and employs specific ion-interaction theory (SIT) as the ionic strength correction model and it also provides the latest thermodynamic data. In Table 1, information about the thermodynamic databases is represented.

2.2 Ionic strength correction model

In general, equilibrium constants are used for studying system equilibrium or reaction, but it is not possible to estimate the equilibrium between substances under ideal standard state conditions. Therefore, in a realistic system, chemical and thermodynamic databases containing aqueous species related to activity coefficients can be used. Because the equilibrium constant can be derived at a constant ionic strength condition, but this value varies depending on the experimenter and the conditions of the system. Therefore, the equilibrium constants are valid only under specific conditions and the equilibrium constant is of limited use. The research related to hydrolysis, complex formation, redox, and solubility equilibrium is actively being conducted [14]. It is started from the identification of the activity of free ions and chemical species in an aqueous solution. Therefore, information related to the activity coefficients of reactants and products is crucial. The experimental data on the free concentration of the chemical species are needed and the thermodynamic database based on activity is essential to establish a methodology for finding the relationship between the concentration of radionuclides and activity. Various methodologies have been proposed to solve the problem of extrapolating data from different ionic media to an infinite dilute aqueous solution [14]. To consider the ionic strength of the system, various ionic strength correction models such as the Debye-Hückel model, Davies model, SIT, and Pitzer equation were

Table 2. Ionic strength correction models analyzed in this study

Ionic strength correction model	Equation	Note
Debye-Hückel model [18]	$log\gamma = -Az^2 \frac{\sqrt{I}}{1 + Ba\sqrt{I}}$	A = 0.51, z = charge, B = 0.33, a = ion size parameter, I = ionic strength
Davies model [15]	$log\gamma = -Az^2 \left(\frac{\sqrt{I}}{1+\sqrt{I}} - 0.3I\right)$	A = 0.51, z = charge, I = ionic strength
Specific ion-interaction theory [16]	$log\gamma = -z^2 D + \sum_j B_{ij} Im_j$	z = charge, D = Debye-Hückel term, B_{ij} = specific interaction term between ions i and j, I = ionic strength, m_j = molality of j
Pitzer equation [17]	$log\gamma_{\pm} = f^{\gamma} + mB^{\gamma} + m^2 C^{\gamma}$	$f^{\gamma} = -A_{\varphi}[I^{1/2}/(1 + bI^{1/2}) + (2/b)\ log(1 + bI^{1/2})]$ $B^{\gamma} = 2\beta^{(0)} + 2\beta^{(1)}\{[1-\exp(-\alpha I^{1/2})(1 + \alpha I^{1/2}-1/2\ \alpha^2 I)]/(\alpha^2 I)\}$, $C^{\gamma} = 1.5C_{\varphi}$

considered and analyzed. In Table 2, characteristics of each ionic strength correction model were shown. In this study, SIT was selected as the ionic strength correction model in this study [15-18]. Because the SIT model is formulated in linear terms, the accuracy of the fitted coefficients is high, and the uncertainty is relatively insignificant compared to others. The SIT model can estimate the reasonably accurate value of the ion interaction coefficient. Therefore, the SIT model can obtain the reliability of both the estimated and the experimentally determined values of the ion interaction coefficient. As a result, this model can be applied to a relatively wide range of ionic strengths. For the SIT model, in the range of ionic strengths ($I \leq 0.1$ mol·kg^{-1}), the uncertainty of the SIT coefficient is negligible. Therefore, a more accurate estimation of the activity coefficient can be accomplished. Because the ionic strength range ($I \leq 0.1$ mol·kg^{-1}) of the SIT model is relatively well-matched with the groundwater conditions of South Korea, so the general application of the SIT model is chosen for appropriate for this study.

2.3 Radionuclides inventory

The radionuclide inventory database adopted in this study were the results of the previous study [1]. According to the previous study, radionuclide inventory was analyzed from the perspectives of radioactivity and mass to derive the radionuclide of concern. As a result, ^{14}C, ^{59}Ni, ^{63}Ni, ^{99}Tc, ^{137}Cs, ^{238}U, and ^{241}Am were selected as the main radionuclides. For gross alpha, although it has not been accurately identified, ^{241}Am and ^{238}U which is representative actinide was selected as the representative radionuclides of gross alpha. If the radionuclide list and fraction of gross alpha are determined, additional information related to gross alpha can be applied to the methodology established in this study to obtain a more accurate safety assessment result. The radionuclide inventory database is shown in Table 3.

Table 3. Radionuclide inventory adopted in this study

Radionuclide	Inventory (Bq)	Half-life (year)
^{14}C	1.66×10^{14}	5.73×10^{3}
^{59}Ni	3.78×10^{13}	7.60×10^{4}
^{63}Ni	2.71×10^{15}	1.00×10^{2}
^{99}Tc	7.85×10^{11}	2.11×10^{5}
^{137}Cs	1.21×10^{14}	3.02×10^{1}
Gross alpha	6.22×10^{11}	-

2.4 Groundwater system

The solubility can be significantly affected by the groundwater and geochemical conditions. Therefore, it is essential to apply the site-specific groundwater and the

Table 4. Groundwater composition of the Gyeongju LILW repository used in this study (averaged groundwater data from 2015 to 2016)

Parameter	Values
pH	7.2 ± 0.8
Eh	181 ± 39 mV
Components	Concentration ($mg \cdot L^{-1}$)
Na	1,819.2
K	76.3
Ca	404.6
Mg	265.4
SiO_2	31.7
Cl^-	3,769.7
SO_4^{2-}	645.4
Alkalinity as HCO_3^-	80.6
Br^-	21.8
Fe	20.4

geochemical conditions of the Gyeongju LILW repository to derive site-specific solubility. For site-specific groundwater data, the safety analysis report of the first phase disposal facility was analyzed and adopted. Currently, the first phase disposal facility is the only operating disposal facility, the groundwater data of the first phase disposal facility is available and was applied in this study as the main groundwater system data. Groundwater system data were collected through several boreholes at the Gyeongju LILW repository, provided by the Korea Radioactive Waste Agency (KORAD), and includes enormous amounts of data for each borehole categorized by period, time, and depth. To consider the operational influence on the groundwater system by the disposal facility at the Gyeongju LILW repository, the average value of groundwater data during the operation period was adopted. For the applicability of the thermodynamic database, the temperature was set to 25°C, and data below 5 ppm were excluded because its effect on the system is considered insignificant. In Table 4, the groundwater composition applied in this study is represented.

The main subject of this study is to derive the site-specific solubility of the Gyeongju LILW repository, but radiation dose assessment was performed using the RESRAD-OFFSITE code to evaluate the effect of solubility on the radiation dose change.

3. Results and discussion

3.1 Solubility

For reliable safety assessment of a radioactive waste disposal facility, the migration behavior of radionuclides should be evaluated. To derive the site-specific solubility of the Gyeongju LILW repository reflecting its groundwater conditions, various data such as thermodynamic database, groundwater data, and radionuclide data were entered into the PHREEQC code. The saturation index and the solid phase of each radionuclide were considered, and the information of chemical species present in the aqueous solution was also evaluated. Following this procedure, the migration behavior of radionuclides was evaluated reflecting the Gyeongju LILW repository's characteristics. Especially the groundwater data during the operation of the disposal facility was considered. In addition, a realistic and complex groundwater system was developed by simultaneously applying all related radionuclides. Therefore, the interaction between radionuclides and groundwater and the interactions between the individual radionuclides could be considered. Cs and C, which represents high solubility in the current condition, the solubility limit was not established to represent the well-soluble characteristics for conservative assessment. After fixing the dominant solid phase of each radionuclide, the solubility was derived by varying the pH to observe the solubility variation trend according to pH in the current groundwater conditions. The adopted solubility product constant includes deviation, and this deviation was also considered. Fig. 1 shows the solubility considering the pH. The dominant solid phase

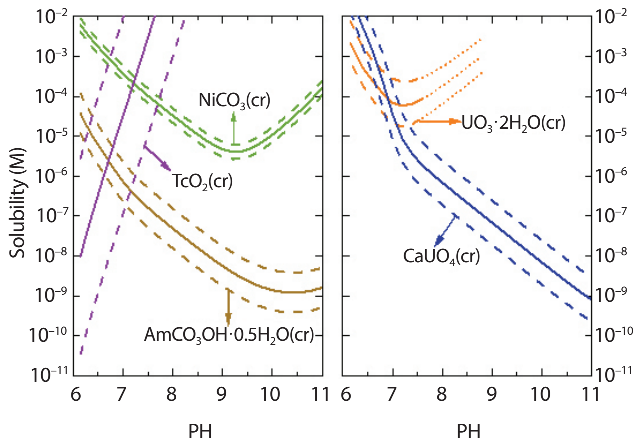

Fig. 1. Solubility variation according to pH.

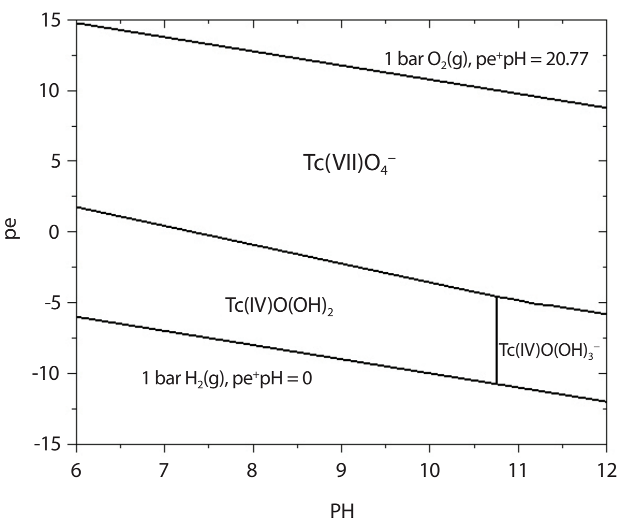

Fig. 2. Pourbaix diagram of Tc.

of Am is $AmCO_3OH \cdot 0.5H_2O$(cr); of Ni, $NiCO_3$(cr); of Tc, TcO_2(cr); of U, $CaUO_4$(s). As shown in Fig. 1, in the case of U, changes of the solubility limiting phase from $CaUO_4$(s) to $UO_3 \cdot 2H_2O$(cr) was observed at around pH 6.8, inducing a further decrease of U solubility under neutral to weakly acidic condition. The solubility of Ni and Am tends to decrease and then increase in the pH range of from 6 to 11. The solubility of U tends to decrease according to pH increase compared to the solubility of Tc increases rapidly as the pH increases.

To analyze the reason for the large and rapid increase in solubility of Tc, Pourbaix diagram of Tc in the current groundwater system was derived and the results are shown in Fig. 2. In general, Tc(IV) dominates the solubility of Tc in the system. But in the current groundwater condition of pH 7.2, Eh 181 mV, and pe 3, Tc(VII) dominates the Tc solubility of the system instead of Tc(IV). The TcO_4^- is known to highly soluble and form significant aqueous complexes. Therefore, it has been confirmed that the value of Tc solubility is relatively high and increases rapidly.

3.2 Effect of solubility on radiation dose

The radiation doses derived in this section are the results by only applying the site-specific solubility and radionuclide inventory data. Since the site-specific parameters related to the Gyeongju LILW disposal facility and the repository site were not applied, the derived radiation dose is not exactly the site-specific radiation dose of the Gyeongju LILW repository. However, the radiation dose was derived to estimate the degree of variation in radiation dose owing to the change in solubility which reflects the influence of the operation of the Gyeongju LILW repository. For the radiation dose calculation, the RESRAD code that is generally used to derive the radiation dose was adopted. This code was developed by the Argonne National Laboratory (ANL) to perform a safety assessment of the nuclear facilities [19]. The RESRAD code considers the various site-specific parameters and possible exposure pathways such as external, inhalation, ingestion. By considering the migration of radionuclides from the source to the surrounding, the environmental media is contaminated and people can be exposed to the radiation dose. Based on the solubility and radionuclide inventory, radiation dose was derived using the RESRAD-OFFSITE code. According to the previous studies, the RESRAD code has been used to derive radiation dose for the safety assessment of nuclear power plants and radioactive waste repository [20-24]. Solubility was varied by 0.1 and 10 times compared to the reference

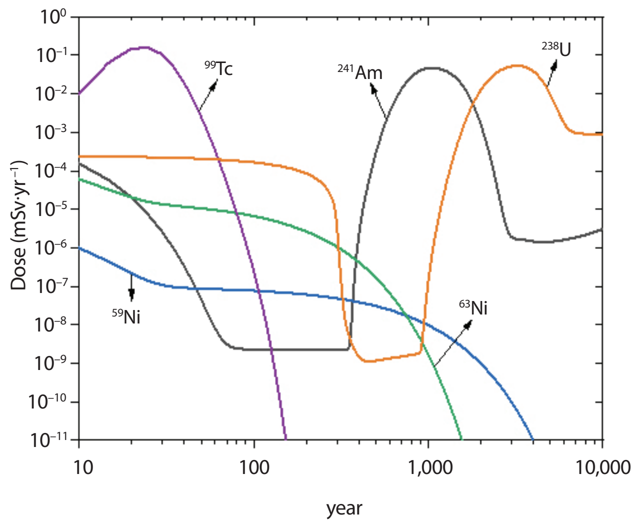

Fig. 3. Radiation dose estimation using the derived solubility.

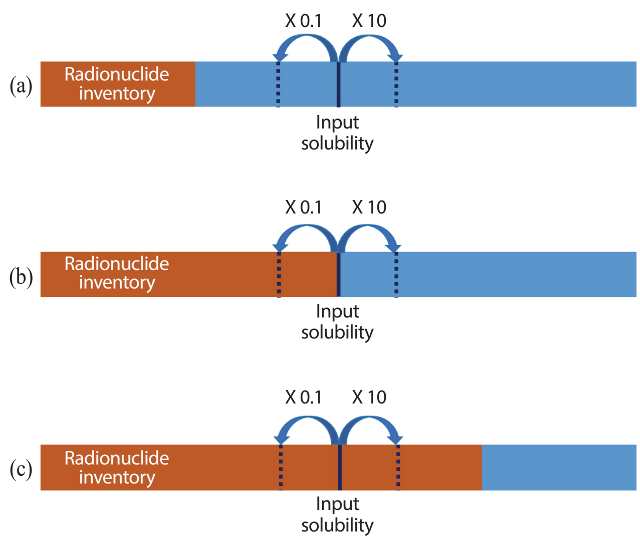

Fig. 4. The conceptual model for the relationship between radionuclide inventory and input solubility.

value to evaluate the radiation dose sensitivity. Derived radiation dose results are shown in Fig. 3. The radiation doses of ^{99}Tc, ^{59}Ni, and ^{63}Ni show a tendency toward decreasing over time. The radiation doses of ^{241}Am and ^{238}U decrease at first but increase at the end. This increase in radiation doses results from the radiation dose variation in progeny. ^{241}Am and ^{238}U were not composed of a single radiation dose, and the radiation dose results were related to the radiation dose of progeny. Therefore, the radiation dose does not decrease gradually but has an inflection point.

It was confirmed that the radiation dose change according to the solubility variation is insignificant. The reason for this observation can be explained by comparing the amount of radionuclide inventory to the input solubility value. Fig. 4 represents the correlation of solubility on the radionuclide inventory. As shown in Fig. 4(a), when the amount of radionuclide inventory is relatively lower, the variation of solubility will not result in a discernible change in the amount of radionuclide dissolved. Therefore, the variation of radiation dose according to solubility change is negligible. In the case of Fig. 4(b), if the amount of radionuclide inventory is relatively high, the amount of radionuclide which is dissolved in the groundwater changes according to the solubility decrease. Therefore, the concentration of radionuclides in the groundwater changes, resulting in a radiation dose variation. As shown in Fig. 4(c), if the amount of radionuclide inventory is high enough, the amount of radionuclide that is dissolved in the groundwater is sensitively changed according to the increase or decrease of solubility. Because radionuclide can be dissolved in the groundwater according to solubility change. Therefore, the radiation dose will vary sensitively according to the solubility change under the conditions of Figure 4(c). The sensitivity analysis of radiation dose represents that the effect of solubility variation to the radiation dose is insignificant, and this is because of relatively low amount of radionuclide inventory such as Fig. 4(a).

4. Conclusions

In this study, the site-specific solubility of the Gyeongju LILW repository was derived and the effect of solubility on the radiation dose was evaluated. This solubility was derived by applying groundwater data reflecting the operational history of the Gyeongju LILW disposal facility, radionuclide data, and thermodynamic database to the

PHREEQC computational code. The main characteristic of the derived solubility is that the selected major radionuclides were applied simultaneously to the groundwater system to consider not only the interactions of each radionuclide but also the effects between the radionuclides and the groundwater system. Under the current conditions, it was confirmed that the effect of solubility on radiation dose was insignificant because the amount of the radionuclide inventory is relatively low. If additional site-specific information is secured, the more site-specific solubility can be quickly derived using the established methodology in this study. In addition, the derived solubility is expected to be used as a major geochemical parameter for the safety assessment of radioactive waste repository and can be a comparative basis.

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