

REVIEW AND COMPILATION OF DATA ON RADIONUCLIDE MIGRATION AND RETARDATION FOR THE PERFORMANCE ASSESSMENT OF A HLW REPOSITORY IN KOREA

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In this study, data on radionuclide migration and retardation processes in the engineered and natural barriers of High-Level Radioactive Waste (HLW) repository have been reviewed and compiled for use in the performance assessment of a HLW disposal system in Korea. The status of the database on radionuclide migration and retardation that is being developed in Korea is investigated and summarized in this study. The solubilities of major actinides such as U, Th, Am, Np, and Pu both in Korean bentonite porewater and in deep Korean groundwater are calculated by using the geochemical code PHREEQC (Ver. 2.0) based on the KAERI-TDB(Korea Atomic Energy Research Institute-Thermochemical Database), which is under development. Databases for the diffusion coefficients (D_e^b values) and distribution coefficients (K_d^b values) of some radionuclides in the compacted Korean Ca-bentonite are developed based upon domestic experimental results. Databases for the rock matrix diffusion coefficients (D_e^r values) and distribution coefficients (K_d^r values) of some radionuclides for Korean granite rock and deep groundwater are also developed based upon domestic experimental results. Finally, data related to colloids such as the characteristics of natural groundwater colloids and the pseudo-colloid formation constants (K_{pc} values) are provided for the consideration of colloid effects in the performance assessment.

KEYWORDS : Performance Assessment, Radionuclide Migration and Retardation, Database Development, Solubility, Diffusion Coefficient, Distribution Coefficient, Rock Matrix Diffusion, Colloid Formation and Migration

1. INTRODUCTION

Presently, spent fuels (SFs) generated from nuclear power plants (NPPs) in Korea are considered a HLW in the Korean radioactive waste management policy. In fact, a direct disposal policy has been applied in Korea without any consideration for the reuse of SFs, despite the recent emergence of the pyro-process as a nuclear non-proliferation method for the reuse of SFs generated from Pressurized Water Reactor (PWR). Since 1997, KAERI has been developing the Korean reference disposal concept for the permanent disposal of HLWs based on the direct disposal of SFs generated from both PWR and CANDU (CANada Deuterium Uranium) [1].

Fig. 1 shows a schematic drawing of the Korean Reference HLW disposal System (KRS). As shown in Fig. 1, the KRS is based upon a multi-barrier system composed of an engineering barrier system (EBS) and a natural barrier system (NBS) [2]. Fig. 2 also shows a

schematic illustration of the engineering barrier system of the KRS, which consists of disposal containers encapsulating the SFs as well as buffer and backfill. The SFs from PWR and CANDU are encapsulated separately in different disposal containers. After the emplacement of a container, the gap between the container and the wall of a borehole is filled with the buffer material and then the room spaces of the tunnels are filled with the backfill material. The present design concept includes the use of domestic Ca-bentonite and its mixture with crushed rock as a buffer and a backfill, respectively.

Fig. 3 shows the release pathways of radionuclides and the multi-barrier concept for the deep geological disposal of the HLW. The buffer material serves as a low-permeability barrier, allowing only a diffusive solute transport. The microscopic pore structure of the buffer is assumed to filter out colloids, microorganisms and natural organic substances with high molecular weights. In many countries, bentonite has been considered as a

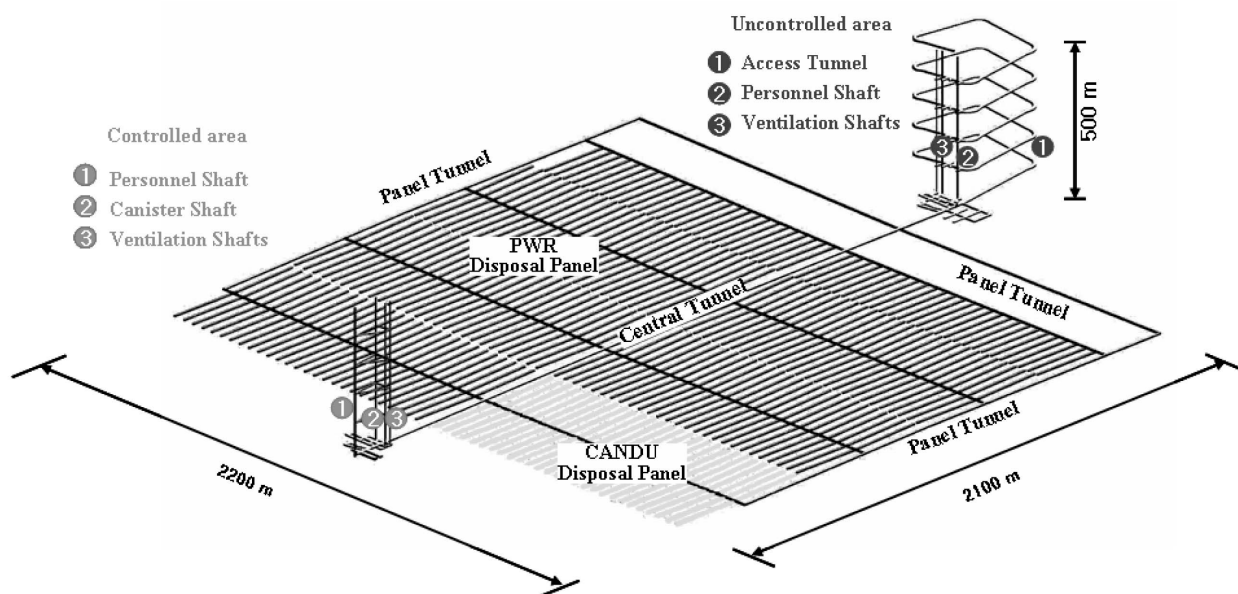


Fig. 1. A Schematic Drawing of a Korean Reference HLW Disposal System (KRS)

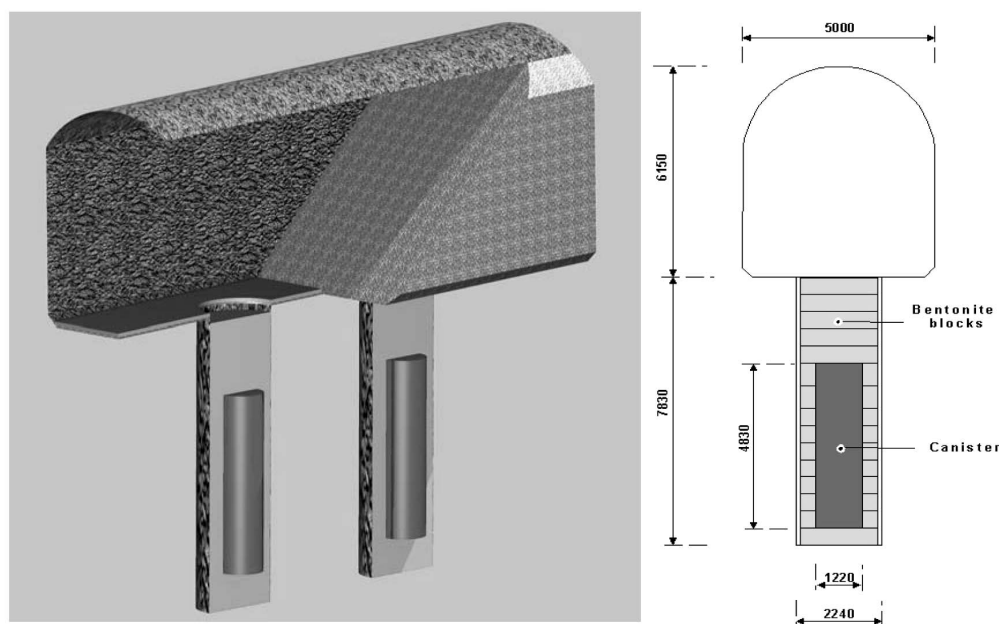


Fig. 2. A Schematic Illustration of the Engineered Barrier System of the KRS

candidate buffer material because of its low hydraulic conductivity, high sorption capacity, self-sealing characteristics, and durability in a natural environment [3, 4]. The Korean Ca-bentonite referred to as ‘Gyeongju bentonite’ has been considered as a possible candidate buffer material for the KRS [1, 2]. Basic physicochemical

and mechanical properties of the Korean bentonite have also been investigated extensively [5]. As shown in Fig. 1, the NBS considered in the KRS is mainly a crystalline rock surrounding the repository [6]. The repository is conceived to be constructed in bedrock several hundred meters (usually about 500 meters) below the ground surface.

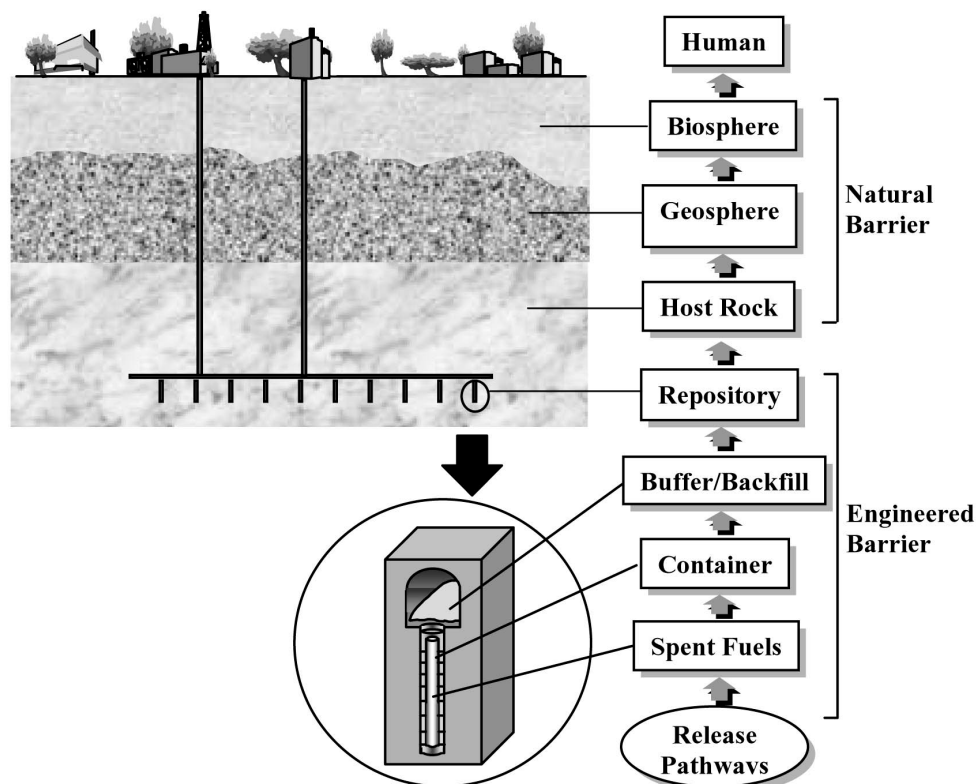


Fig. 3. The Release Pathways of Radionuclides and the Multi-Barrier Concept for the Deep Geological Disposal of HLW

Radionuclides migrate in the buffer material through diffusion driven by concentration gradients whose upper value is limited by the solubility partitioned among the various isotopes. The radionuclides are retarded by sorption onto the mineral constituents of the buffer material in the course of a migration. The flow of groundwater and transport of radionuclides in the fractured rock mass occur in the heterogeneous network of permeable fractures. Once radionuclides are leached out of the buffer, they flow into the rock fractures and are transported as solutes by advection and dispersion.

Radionuclides also diffuse into the rock matrix adjacent to fractures and are retarded by the sorption on pore surfaces in the matrix depending on their sorption properties. Radionuclides reaching major faults transport as solutes due to the advection and dispersion, and are retarded by the matrix diffusion and sorption. Radionuclides, especially actinides, can be released as colloidal species in natural aquatic systems, definitely under near-field geochemical conditions [7]. Radionuclides with a higher sorption capacity may also be associated with natural aquatic colloidal matter and then form radioactive pseudo-colloids. However, the migration of radionuclides as colloids is more complicated than that of dissolved radionuclides, and the processes involved are less well

understood [7].

In general, the main objective of researches regarding HLW disposal is to present the technical reliability of the geological disposal of HLW by considering national geological environments. Therefore, in this study, data on the migration and retardation processes both in the EBS and NBS have been reviewed and compiled for use in a performance assessment of the Korean HLW disposal system.

2. SOLUBILITY AND SPECIATION DATA

Solubility and speciation data of radionuclides in groundwater are very important for understanding radionuclide migration through the EBS and NBS in a HLW repository. Solubilities are usually used as upper limits of the radionuclide concentrations in a given chemical condition and the speciation is used to determine or discuss the fundamental reactions for the sorption characteristics of different geological materials [8]. Thus, the solubility can define the source term for the release of radionuclides and the transport processes such as sorption and colloid formation. The solubilities of radionuclides can be calculated from thermodynamic databases using a

geochemical code and/or obtained directly from reliable solubility measurements.

In order to estimate the solubilities of radionuclides, several geochemical codes such as PHREEQC [9], EQ3/6 [10], and MUGREM [11] have been developed. For the calculation of the radionuclide solubility using a geochemical code, thermodynamic data and the solubility-limiting solid phases (SLSPs) of the radionuclide, the composition and temperature of the solution, the activities of aqueous species in equilibrium, and so on are required. In this study, the solubilities of major actinides with a long lifetime were calculated by using PHREEQC (Ver. 2.0) and thermodynamic databases for both the domestic compacted Ca-bentonite porewater and the domestic granitic groundwater.

2.1 Thermodynamic Database

Critically reviewed thermodynamic databases with a reliable data selection are rare, and ongoing projects are making slow progress [8]. International efforts have been conducted, e.g., by the International Atomic Energy Agency (IAEA) and the Nuclear Energy Agency (NEA). The IAEA project “The Chemical Thermodynamics of Actinide Elements and Compounds” was terminated after its last publication in 1992 [12]. The thermochemical database (TDB) project of the Organization for Economic Co-operation and Development/Nuclear Energy Agency (OECD/NEA) has a high-quality review standard and includes international reviewers, but its progress is slow. The first database was published in 1992 on uranium [13] and the second one in 1995 on americium [14]. Other databases published by the TDB project include databases on technetium in 1999 [15], neptunium and plutonium in 2001 [16], updates on uranium, neptunium, plutonium, americium and technetium in 2003 [17], nickel in 2005 [18], selenium in 2005 [19], compounds and complexes of U, Np, Pu, Am, Tc, Se, Ni and Zr with selected organic ligands in 2007 [20], and solid solutions in 2007 [21].

Recently, a domestic thermodynamic database for major actinides considered to be important in HLW, the KAERI-TDB, has begun to be developed. The general thermodynamic data for actinide compounds and

complexes of Am, Np, U, Th, and Pu were developed by adopting thermodynamic data from other studies [17, 22] and domestic data [23], respectively. The reactions and stability constants used for the solubility calculation of the actinides were also developed. The equilibrium reaction constants at 25°C with the ionic strength of $I = 0$ and a pressure of 0.1 MPa were determined. Data on other important elements such as C, Cl, Co, Ni, Se, Sr, Zr, Tc, Pd, Sn, I, Cs, Ra, Sm, Eu, Ac, Pa, and Cm will continue to be added to the KAERI-TDB to update the database.

2.2 Solubilities of the Major Actinides in the Bentonite Porewater

The solubilities of major actinides such as U, Am, Th, Np, and Pu in the porewater of a compacted Ca-bentonite (i.e., Gyeongju bentonite) were calculated by using the geochemical code PHREEQC (Ver. 2.0) with an appropriate thermodynamic database of the KAERI-TDB by assuming the most important solid phase as a solubility-limiting solid phase [24]. The density of the bentonite porewater was assumed as 1.0 g/cm³ for the solubility calculation of the actinides. The composition of the bentonite porewater was measured [25] and is shown in Table 1. The solubility-limiting solid phases, calculated solubilities, and the percentages of the aqueous species of U, Am, Th, Np, and Pu are given in Table 2.

2.3 Solubilities of the Major Actinides in the Groundwater

For the calculation of the solubilities of radionuclides in the deep granitic groundwater, the composition of the groundwater must be defined. A natural granitic groundwater was sampled from borehole YS-01, located inside KAERI (in Korea), using a multi-packer system at a depth of 457.5 m from the surface. The geochemical conditions and composition of the groundwater are shown in Table 3. The solubilities of the major actinides such as U, Am, Th, Np, and Pu in the granitic groundwater were also calculated by using the PHREEQC (Ver. 2.0) and the KAERI-TDB [24]. Table 4 shows the solubility-limiting solid phases, calculated solubilities, and the percentages of the aqueous species for U, Am, Th, Np, and Pu.

Table 1. The Composition of the Bentonite Porewater

Temp(°C)	pH	Eh(mV)	Na(mg/L)	SiO ₂ (mg/L)	Ca(mg/L)	Mg(mg/L)	Mn(mg/L)	Zn(mg/L)
25	8.75	-194*	243	20.4	2.0	0.4	0.01	0.01
K(mg/L)	P(mg/L)	Al(mg/L)	Fe(mg/L)	CO ₃ + HCO ₃ (mg/L)	Cl(mg/L)	F(mg/L)	SO ₄ (mg/L)	NO ₃ (mg/L)
2.1	1.1	0.54	0.24	422	26	6.4	161	0.1

*Redox potential value of the porewater was taken from the groundwater.

Table 2. The Calculated Solubilities of Major Actinides in the Bentonite Porewater [24]

SLSP	Solubility (M)	Main species in solution	Percent of species (%)
Uraninite	8.4×10^{-7}	$\text{UO}_2(\text{CO}_3)_3^{4-}$	~ 99
$\text{UO}_2(\text{am,hyd})$	4.0×10^{-4}		
$\text{Am}(\text{OH})_3(\text{am})$	1.0×10^{-4}	$\text{Am}(\text{CO}_3)_2^-$	88.3
$\text{Am}(\text{OH})_3(\text{cr})$	5.5×10^{-6}	$\text{Am}(\text{CO}_3)_3^{3-}$	7.2
$\text{AmCO}_3\text{OH}(\text{am})$	1.8×10^{-6}	AmCO_3^+	4.3
$\text{Th}(\text{OH})_4(\text{am})$	2.2×10^{-7}	$\text{Th}(\text{OH})_2(\text{CO}_3)_2^{2-}$	54.3
$\text{Th}(\text{OH})_4(\text{cr})$	2.2×10^{-13}	$\text{Th}(\text{OH})_3\text{CO}_3^-$	44.0
$\text{NpO}_2(\text{am, hyd})$	6.6×10^{-8}	$\text{Np}(\text{OH})_3\text{CO}_3^-$	86.9
$\text{NpO}_2(\text{cr})$	6.6×10^{-15}	$\text{Np}(\text{OH})_4\text{CO}_3^{2-}$	9.1
$\text{NpO}_2\text{OH}(\text{am, aged})$	6.8×10^{-8}	$\text{Np}(\text{OH})_2(\text{CO}_3)_2^{2-}$	2.5
		$\text{Np}(\text{OH})_4(\text{aq})$	1.5
$\text{Pu}(\text{OH})_4(\text{am})$	1.6×10^{-9}	$\text{Pu}(\text{OH})_3\text{CO}_3^-$	52.1
$\text{PuO}_2(\text{am, hyd})$	2.4×10^{-9}	$\text{Pu}(\text{OH})_2(\text{CO}_3)_2^{2-}$	44.2
$\text{PuO}_2(\text{cr})$	5.1×10^{-15}	$\text{Pu}(\text{OH})_4\text{CO}_3^{2-}$	2.8

SLSP: Solubility limiting solid phase

Table 3. The Geochemical Properties of the Groundwater Sampled from Borehole YS-01

Depth		457.5 m	
Temperature		25 °C	
Eh (mV)		-194	
pH		9.92	
TDS (total dissolved solid)		104.9 mg/L	
DOC (dissolved organic carbon)		1.23 mg/L	
DO (dissolved oxygen)		9.6 mg/L	
Alkalinity		650 mg/L	
Element	Concentration (mg/L)	Element	Concentration (mg/L)
Na	35.7	Cl	4.2
SiO_2	18.5	NO_3	0.02
Ca	2.7	SO_4	2.3
K	0.33	Al	0.0791
Mg	0.05	Mn	0.0008
Li	0.118	Fe	0.00155
CO_3+HCO_3	17.3	Sr	0.059
Cl	4.2	Pb	0.00029
F	12.7	Zn	0.0063

Table 4. The Calculated Solubilities of Major Actinides in the Groundwater [24]

SLSP	Solubility (M)	Main species in solution	Percent of species (%)
Uraninite UO ₂ (am,hyd)	9.7 × 10 ⁻¹⁰ 2.2 × 10 ⁻⁵	UO ₂ (OH) ₃ ⁻	65~70
		UO ₂ (CO ₃) ₃ ⁴⁻	29~32
		UO ₂ (CO ₃) ₂ ²⁻	1.4~2.4
Soddite	2.2 × 10 ⁻⁵	UO ₂ (OH) ₃ ⁻	56.9
		UO ₂ (CO ₃) ₃ ⁴⁻	21.3
		UO ₂ (CO ₃) ₂ ²⁻	19.5
Am(OH) ₃ (am)	1.5 × 10 ⁻⁸	Am(OH) ₂ ⁺ Am(CO ₃) ₂ ⁻	51.6 37.4
Am(OH) ₃ (cr)	7.7 × 10 ⁻¹⁰		
AmCO ₃ OH(am)	1.2 × 10 ⁻⁷		
Th(OH) ₄ (am)	5.9 × 10 ⁻⁹	Th(OH) ₄ (aq)	51.7
Th(OH) ₄ (cr)	6.1 × 10 ⁻¹⁵	Th(OH) ₃ CO ₃ ⁻	46.3
NpO ₂ (am,hyd)	3.4 × 10 ⁻⁹	Np(OH) ₃ CO ₃ ⁻	56.7
NpO ₂ (cr)	3.4 × 10 ⁻¹⁶	Np(OH) ₄ (aq)	29.7
NpO ₂ OH(am, aged)	1.2 × 10 ⁻⁹	Np(OH) ₄ CO ₃ ²⁻	13.6
Pu(OH) ₄ (am)	4.3 × 10 ⁻¹¹	Pu(OH) ₃ CO ₃ ⁻	66.7
PuO ₂ (am, hyd)	6.3 × 10 ⁻¹¹	Pu(OH) ₄ (aq)	23.6
PuO ₂ (cr)	1.4 × 10 ⁻¹⁶	Pu(OH) ₄ CO ₃ ²⁻	8.03

SLSP: Solubility limiting solid phase

3. MIGRATION AND RETARDATION DATA FOR THE EBS

The compacted bentonite buffer serves as a low-permeability barrier because radionuclides only migrate through diffusion driven by concentration gradients whose upper value is limited by the radionuclide solubility. Also, they are retarded by sorption onto the mineral constituents of the buffer material in the course of migration. The microscopic pore structure of the buffer is usually assumed to filter out colloids, microorganisms and natural organic substances. In this study, the data development has mainly focused on the sorption and diffusion of radionuclides in the engineered barrier composed of the domestic Ca-bentonite buffer. Fig. 4 schematically shows the migration and retardation processes of radionuclides in the compacted bentonite buffer.

3.1 Sorption Data for the EBS

In general, the sorption of radionuclides onto bentonite is a main retardation process in the compacted

bentonite buffer, and it is usually represented by the distribution coefficient K_d^b (m³/kg) defined as follows [26]:

$$K_d^b = \frac{S}{C} = \frac{(C_0 - C)}{C} \cdot \frac{V}{M} \quad (1)$$

where S is the concentration of the radionuclide sorbed onto bentonite per unit mass of the bentonite solid (mol/kg), C_0 is the initial concentration of the radionuclide loaded to the system (mol/m³), C is the concentration of the radionuclide in the solution per unit volume of the solution (mol/m³), M is the mass of the bentonite solid (kg), and V is the volume of the solution (m³). Table 5 shows the K_d^b values of some radionuclides obtained from domestic sorption studies [27-29]. As shown in Table 5, K_d^b values of I and Tc show lower values, while Ni, U, Am, and Cs show higher K_d^b values.

3.2 Diffusion Data for the EBS

Diffusion coefficients in the compacted bentonite

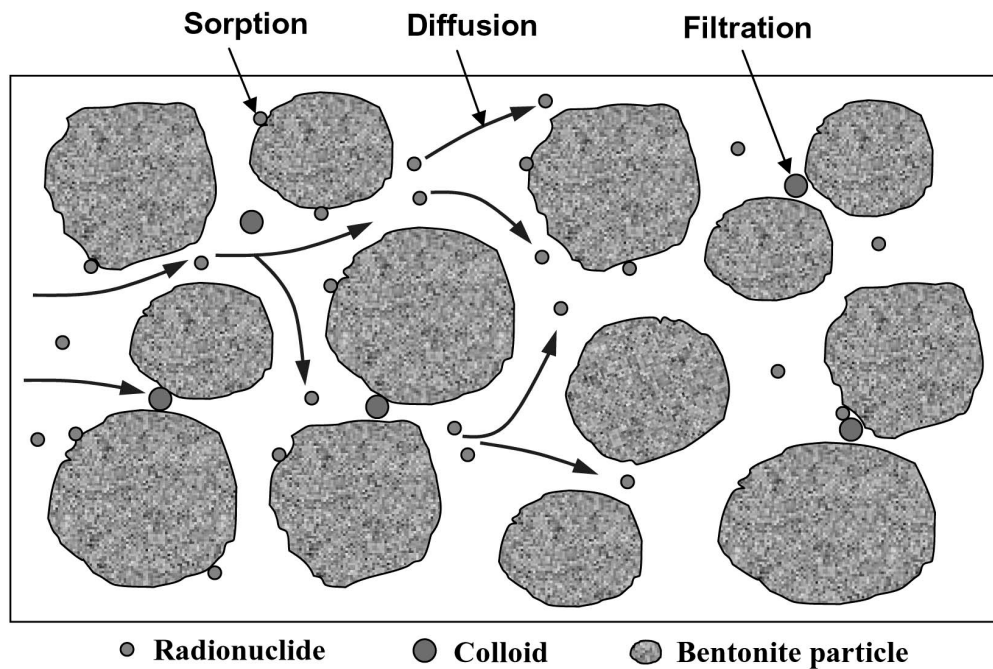


Fig. 4. Radionuclide Migration and Retardation Processes in the Engineered Barrier System (EBS)

Table 5. Radionuclide Sorption Data for Bentonites

Nuclides	Bentonite type	K_d^b (m ³ /kg)	Ref.
Sr	Na-Bentonite	7.6×10^{-2}	[27]
	Ca-Bentonite	1.2×10^{-2}	[27]
I	Na-Bentonite	3.4×10^{-3}	[27]
	Ca-Bentonite	7.4×10^{-3}	[27]
Ni	Ca-Bentonite	7.0×10^{-1}	[28]
Tc	Ca-Bentonite	0.3×10^{-3}	[28]
U	Ca-Bentonite	1.7×10^{-1}	[28]
Am	Ca-Bentonite	7.3×10^{-1}	[28]
Cs	Na-Bentonite	6.6×10^{-1}	[29]

buffers usually depend on the chemical species, and thus they should be determined individually for each chemical species considered. However, available data is very limited due to a wide range of parameters and conditions, such as the mineralogical composition and bentonite density, redox conditions, porewater chemistry, and temperature.

The diffusion coefficient can be expressed as an effective diffusion coefficient D_e^b (m²/s) defined as follows [30]:

$$D_e^b = D_x \cdot \tau \cdot \varepsilon \quad (2)$$

where D_x is the molecular diffusion coefficient of the radionuclide in free water (m²/s), and τ and ε are the tortuosity and porosity of the medium, respectively. However, an apparent diffusion coefficient D_a^b (m²/s) is usually adapted to represent the overall diffusion process by considering the retardation capacity of the medium, and it is defined as follows [30]:

$$D_a^b = \frac{D_e^b}{\varepsilon + K_d^b \cdot \rho_d} \quad (3)$$

where K_d^b is the distribution coefficient of the radionuclide (m³/kg) on the diffusing medium and ρ_d is the dry density of the medium (kg/m³).

Table 6 shows the diffusion data obtained from domestic studies with the domestic Gyeongju bentonite [27, 28, 31-35]. As shown in Table 6, Am shows the lowest D_a^b value of 7.3×10^{-15} m²/s, and HTO shows the highest value of 3.4×10^{-10} m²/s. In the KRS concept, it was recommended that the apparent diffusion coefficients of the cationic radionuclides (excluding strontium) and actinides should be less than 10^{-11} m²/s in the buffer material. On the other hand, D_a^b values should be less than 10^{-10} m²/s and 10^{-9} m²/s for the Sr and anionic radionuclides such as I, respectively [2].

Table 6. Radionuclide Diffusion Data for Compacted Bentonites

Nuclide	Bentonite type	ρ_b (Mg/m ³)	D_e^b (m ² /sec)	D_a^b (m ² /sec)	Ref.
Cl	Ca-Bentonite	1.6	-	9.5×10^{-11}	[31]
HTO	Ca-Bentonite	1.4	-	3.4×10^{-10}	[28]
Tc	Ca-Bentonite	1.4	-	9.5×10^{-11}	[28]
Ni	Ca-Bentonite	1.4	-	2.0×10^{-13}	[28]
Am	Ca-Bentonite	1.4	-	7.3×10^{-15}	[28]
U	Ca-Bentonite	1.4	-	1.8×10^{-13}	[28]
Co	Ca-Bentonite	1.4	-	3.4×10^{-13}	[32]
C	Ca-Bentonite	1.6	-	7.6×10^{-11}	[31]
Sr	Na-Bentonite	1.3	2.4×10^{-11}	8.5×10^{-12}	[27]
	Ca-Bentonite	1.4	1.9×10^{-11}	4.1×10^{-12}	[33]
I	Na-Bentonite	1.3	7.5×10^{-12}	4.8×10^{-11}	[27]
	Ca-Bentonite	1.4	9.5×10^{-12}	5.7×10^{-11}	[33]
Cs	Na-Bentonite	1.4	7.0×10^{-12}	5.7×10^{-13}	[34]
	Ca-Bentonite	1.4	1.1×10^{-11}	1.4×10^{-12}	[35]

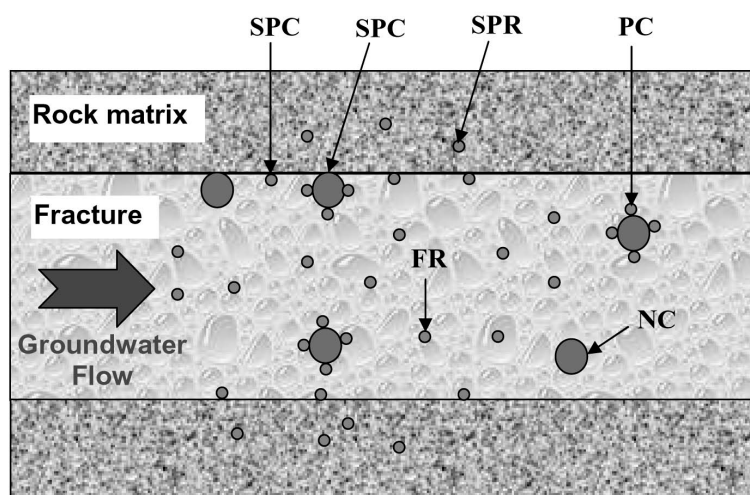
D_e^b : effective diffusion coefficient in the compacted bentonite

D_a^b : apparent diffusion coefficient in the compacted bentonite

4. MIGRATION AND SORPTION DATA FOR THE NBS

In the performance assessment of a radioactive waste repository, it is necessary to understand how radionuclides

migrate through the surrounding geological formation. The main migration mechanism of radionuclides in the rock surrounding a waste repository would be migration via groundwater flowing through rock fractures. Dissolved



FR: dissolved mobile radionuclide in the fracture

NC: mobile natural colloids

PC: mobile radioactive pseudo-colloid

SFR: immobile radionuclide sorbed on the fracture surface

SPR: immobile radionuclide sorbed on rock matrix pore surfaces

SPC: immobile pseudo-colloid sorbed on the fracture surface

Fig. 5. Radionuclide Migration and Retardation Concept in the Natural Barrier System (NBS)

Table 7. Recommended Rock Matrix Diffusion Coefficients of Radionuclides

Nuclide	D_x (m ² /s)	D_e (m ² /s)	D_a (m ² /s)	Ref.
I	2.0×10^{-9}	$8.3\text{--}230 \times 10^{-14}$	5×10^{-11}	[23, 36, 37]
Cl	2.0×10^{-9}	8.3×10^{-14}	2×10^{-11}	[23, 36, 37]
Cs	2.1×10^{-9}	8.8×10^{-14}	6×10^{-16}	[23, 36, 38]
Sr	7.9×10^{-10}	$3.3\text{--}27 \times 10^{-14}$	6×10^{-14}	[23, 36, 38]
U	$4.3\text{--}10 \times 10^{-10}$	3.6×10^{-14}	3×10^{-18}	[23, 39, 40]
Th	1.5×10^{-10}	6.3×10^{-15}	5×10^{-19}	[23, 39, 40]
Am	1×10^{-9}	4×10^{-14}	5×10^{-18}	[40-42]
Pu	1×10^{-9}	4×10^{-14}	3×10^{-18}	[40-42]
Pa	1×10^{-9}	4×10^{-14}	1×10^{-17}	[40-42]
Ac	1×10^{-9}	4×10^{-14}	5×10^{-18}	[40-42]
Cm	1×10^{-9}	4×10^{-14}	5×10^{-18}	[40-42]
Ra	8.9×10^{-10}	3.7×10^{-14}	7×10^{-16}	[40-42]
Np	1×10^{-9}	$4\text{--}25 \times 10^{-14}$	3×10^{-18}	[40-42]
C	1.2×10^{-9}	5×10^{-14}	2×10^{-14}	[40-42]
Ni	6.8×10^{-10}	2.8×10^{-14}	5×10^{-16}	[40-42]
Se	1×10^{-9}	4×10^{-14}	1×10^{-14}	[40-42]
Zr	1×10^{-9}	4×10^{-14}	1×10^{-17}	[40-42]
Nb	1×10^{-9}	4×10^{-14}	1×10^{-17}	[40-42]
Tc(IV)	1×10^{-9}	4×10^{-14}	1×10^{-17}	[40-42]
Tc(VII)O ₄ ⁻			8×10^{-12}	[40-42]
Pd	1×10^{-9}	4×10^{-14}	1×10^{-15}	[40-42]
Sn	1×10^{-9}	4×10^{-14}	1×10^{-14}	[40-42]
Sm	1×10^{-9}	4×10^{-14}	7×10^{-18}	[40-42]

D_x = molecular diffusion coefficient in water

D_e = effective diffusion coefficient in the rock matrix

D_a = apparent diffusion coefficient in the rock matrix

radionuclides may be in solution, or they may be associated with fine particulate matter (colloids). The migration of dissolved radionuclides may depend upon the advection-dispersion, and interactions with the rock matrix. However, data related to groundwater flow and geological media were not considered in this study since this study is mainly focused on database development for radionuclide behaviors. Thus, this section mainly focuses on data development for the sorption and rock matrix diffusion of radionuclides in the fractured granite rock. Fig. 5 schematically shows the migration and retardation processes of radionuclides and colloids in the fractured rock.

4.1 Diffusion Data for the Rock Matrix

Diffusion and sorption are the main retarding

mechanisms in the migration of radionuclides through geological media. Thus, many countries have measured the diffusivities of radionuclides in rock matrix. The rock matrix diffusion coefficients (D_e values) of the major radionuclides in the crystalline rocks are listed in Table 7 [23, 36-42]. The rock matrix diffusion coefficients listed in Table 7 are recommended values based on the experimental data from Korean rocks. As shown in Table 7, the diffusivities of the actinides show values with wide ranges, since the properties and chemical species of the actinides strongly depend on individual geochemical environments [43].

4.3 Sorption Data for the Granitic Rock

The equilibrium partition constant usually called a

Table 8. Radionuclide Sorption Data for Granite [25]

Nuclide	Concentration (M)	Groundwater	K_d range (m ³ /kg)	K_d^* (m ³ /kg)
Co	10 ⁻⁵	GW-1	0.004 – 0.033	0.005
Ni	10 ⁻⁷ ~ 10 ⁻⁵	GW-2	1.706 – 5.667	1
Se	10 ⁻⁷ ~ 10 ⁻⁵	GW-2	0 – 0.0024	0
Sr	10 ⁻⁶ ~ 10 ⁻⁵	GW-1	0.0035 -0.0 69	0
Zr	10 ⁻⁷ ~ 10 ⁻⁵	GW-2	0.028 – 1.077	0.2
Pd	10 ⁻⁷ ~ 10 ⁻⁵	GW-2	0.030 – 2.965	0.03
Sn	10 ⁻⁷ ~ 10 ⁻⁵	GW-2	0.021 – 0.337	0.02
I	10 ⁻⁷ ~ 10 ⁻⁵	GW-1	0	0
Cs	10 ⁻⁷ ~ 10 ⁻⁵	GW-2	0.052 – 0.073	0.05
Sm	10 ⁻⁷ ~ 10 ⁻⁵	GW-2	0.748 – 15.139	1
Eu	10 ⁻⁷ ~ 10 ⁻⁵	GW-1	0.049 – 0.173	0.07
Th	10 ⁻⁷ ~ 10 ⁻⁵	GW-2	4 – 7.4	5
U	10 ⁻⁷ ~ 10 ⁻⁵	GW-2	0.0032 – 0.0045	0.005

* Representative K_d value

GW-1: Natural groundwater sampled from borehole YS-01, pH=9.79, Eh=0.3mV

GW-2: Natural groundwater sampled from borehole YS-01, pH=9.94, Eh=-161mV

distribution coefficient (K_d) has been used to quantify a sorption that is kinetically fast and reversible [26]. However, the application of K_d in the performance assessment of a radioactive waste repository has been limited since this value is only valid for the conditions studied. Many factors such as the amount and type of minerals, the composition of the aqueous phase, pH and Eh play important but widely different roles in the sorption [26]. Thus, the development of a methodology for the selection of suitable sorption data from a vast range of experimentally determined K_d values is pivotal for the performance assessment of a radioactive waste repository.

The NEA Sorption Data Base (SDB) was developed to provide comprehensive sorption data for the performance assessment of radioactive waste repositories [26]. The use of the SDB, however, has been limited because of the complexity of its manipulation and the absence of an analysis tool for the selection of suitable sorption data. The Japan Atomic Energy Agency (JAEA) also developed a sorption database (JNC-SDB) in 1999, and started a web-version service in 2005 [44]. Recently, Germany developed a digitized version of the thermodynamic sorption database, the RES T-Rosendorf expert system [45]. The recorded data comprises surface complexation reactions. However, the application of surface complexation models has been limited due to a lack of data.

KAERI has also established a sorption database for evaluating the performance of a radioactive waste

repository. The software developed at KAERI, SDB-21C, is a graphical user interface (GUI) program that provides efficient and user-friendly tools for evaluating a large amount of sorption data. The SDB-21C provides many functions such as data addition, retrieval, and graphic representation in order to effectively analyze the attained sorption data [46]. For the improvement of the simple K_d approach, a parametric model was also included in the SDB-21C. The SDB-21C was established to fulfill two functions. One is to develop an integrated sorption data management tool and the other is to acquire relevant sorption data in a consistent manner. The database of K_d compiled in the program contains about 11,000 NEA data entries and more than 4,000 KAERI data entries.

Table 8 shows the distribution coefficient K_d values obtained from domestic experimental studies for domestic granite and groundwater [25]. The groundwater was also sampled from the borehole YS-01, as shown in Table 3. The granite used in the sorption experiments was taken from a declined borehole at a depth of 157 m at the KAERI Underground Research Tunnel (KURT). The granite mainly consists of quartz (32.13%), plagioclase (47.07%), biotite (7.2%), microcline (6.8%), chlorite (6.4%), and muscovite (0.4%).

5. COLLOID MIGRATION AND RETARDATION

5.1 Properties of Natural Colloids

Table 9. Comparison of the Characteristics for Natural Colloids

Sampling site	Concentration($\mu\text{g/L}$)	Size (nm)	Composition	pH	Depth (m)	Ref.
Sweden						
Non-saline	43 ± 43	50~500	clay, silica $\text{Fe}(\text{OH})_3$	8.0	275	[50]
Saline	20 ± 20	50~500	clay, silica $\text{Fe}(\text{OH})_3$	7.9	500	
Switzerland						
Leuggern	20	10~1000	illite, silica	7.9	1650	[50]
Zurzach	10	10~1000	illite, silica	8.0	500	
Canada	300 ± 300	5~1000	clay, organic calcite	~ 8.0	500	[50]
Canada ¹⁾	~ 264	1~450	organics carbonates aluminosilicates	~ 7.0	240	[53]
Korea	~ 200	400	aluminosilicates	~ 9.9	450	[54]
Yuseong ²⁾			Fe-oxides calcite			

¹⁾ Conducted jointly by KAERI and AECL²⁾ Conducted jointly by KAERI and INE

Radionuclides, especially actinides, can migrate or discharge not only as simply dissolved molecules but also as colloids or complexes. It has been reported that the formation of colloids by the sorption of radionuclides on negatively charged naturally occurring colloidal matter can increase drastically radionuclide migration [47-49]. The main conclusion of these studies is that colloids can accelerate the migration of radionuclides in groundwater. In terms of the safety and performance assessment of a radioactive waste repository, therefore, the roles of colloids in radionuclide migration should be estimated by an appropriate quantitative method. However, the exact mechanisms of the colloid migration and the contribution of colloids to the radionuclide migration have not been accurately determined.

Many field studies [50-52] have been performed in order to investigate the characteristics of natural colloidal particles such as their composition, size distribution, and concentration in subsurface systems. The physicochemical properties of natural colloids sampled from a deep granitic groundwater in the Canadian Shield were carried out jointly by KAERI and Atomic Energy of Canada Limited (AECL) to characterize the behaviors of natural colloids in deep granitic groundwater [53]. Several years ago, the groundwater colloids sampled from a borehole in the Yuseong area of Daejeon, Korea were characterized by the Laser-Induced Breakdown Detection (LIBD) and

Inductively-Coupled Plasma-Mass Spectrometry (ICP-MS) methods in combination with the Asymmetrical Flow Field-Flow Fractionation (AsymFFFF) method as a joint study with the Institut für Nukleare Entsorgung, Forschungszentrum Karlsruhe (INE/FZK), of Germany [54].

In Table 9, the characteristics of natural colloids obtained from domestic granitic groundwater and foreign granitic groundwaters are compared. As shown in Table 9, the characteristics of the domestic natural groundwater colloids are very similar to those of the Canadian colloids.

5.2 Colloid Migration and Sorption

If radionuclides were to become attached to mobile colloidal particles to form pseudo-colloids, the transport behaviors of those radionuclides would be different from the behaviors of dissolved radionuclides. To assess the importance of the mobile colloids on the radionuclide migration at a particulate disposal site, information is required on both the total mobile load of the radionuclide and colloids as well as the distribution of the radionuclide between the colloidal and dissolved phases. Sorption properties of radionuclides onto bentonite colloids and natural colloids can provide information about the colloid migration and sorption in the engineered and natural barriers.

Generally, the sorption coefficient of the radionuclide onto colloids called a pseudo-colloid formation constant (K_{pc}) can be given as [50, 52, 53]:

$$K_{pc} = \frac{PC}{C \cdot C_c} \quad (4)$$

where C is the concentration of the radionuclide in the solution (mol/L), C_c is the concentration of the colloids in the solution (kg/L), PC is the concentration of the radionuclide sorbed on the colloids per unit volume of the solution (mol/L), and K_{pc} is the equilibrium pseudo-colloid formation constant (L/kg).

Pseudo-colloid formation experiments for some radionuclides such as Sr, Am, and U were conducted with natural groundwater colloids [55], artificial silica [56], and bentonite colloids [57], respectively, and the results are shown in Table 10. It can be concluded that for strongly sorbing radionuclides, their association with colloids could have a significant influence on their migration processes if the colloid concentration is sufficiently high and the colloids are mobile. Thus, the pseudo-colloid formation of radionuclides with colloids has been given the highest priority because pseudo-colloids formed in the geosphere could migrate faster than the dissolved radionuclides.

Although various migration models for fractured rock media [58-60] have recently been introduced to analyze and predict the migration behaviors of colloidal particles and their effects on radionuclide migration in subsurface systems, a unified approach has not yet been established.

Table 10. Pseudo-Colloid Formation Constants of Radionuclides with Colloids

Nuclide	Colloid	Solution	K_{pc} (mL/g)	Ref.
Am	Natural colloids	Granite	7×10^4	[55]
		groundwater	$\sim 7 \times 10^5$	
Sr	Natural colloids	Granite	3×10^2	[55]
		groundwater	$\sim 1 \times 10^3$	
U	Silica fumed	pH 4~10 Carbonate	$10^4 \sim 10^5$	[56]
U	Ca-bentonite colloids	NaClO ₄	$10^4 \sim 10^7$	[57]

6. CONCLUSIONS

In this study, the status of the development of a database on radionuclide migration and retardation for the performance assessment of a HLW repository in Korea has been reviewed. The solubilities of major actinides such as U, Th, Am, Np, and Pu both in Korean

compacted Ca-bentonite porewater and in deep Korean groundwater were calculated by using the geochemical code PHREEQC (V.2) based on the KAERI-TDB, which is being developed.

Data for the diffusion coefficients and distribution coefficients of some radionuclides in the EBS, i.e., in the compacted Korean Ca-bentonite, were compiled based upon domestic experimental results, although some data were not sufficient. In addition, data on the rock matrix diffusion coefficients and the distribution coefficients of some radionuclides in the NBS, i.e., in Korean granite rock and deep groundwater, were also compiled on the basis of domestic experimental results in spite of some insufficient data. Finally, data related to colloids such as the characteristics of natural groundwater colloids and the pseudo-colloid formation constants were presented for the consideration of colloid effect on the performance assessment of a HLW repository in Korea.

This study clarifies the research status of inquiries into the radionuclide migration and retardation processes important for the performance assessment of HLW repositories. Additionally, the compiled radionuclide migration and retardation data will provide technical information and relevant data necessary for the performance assessment of the KRS for the safe disposal of HLW in Korea.

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