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Database development of glass dissolution and radionuclide migration for performance analysis of HLW repository in Japan

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Abstract

Japan Nuclear Cycle Development Institute (JNC, the successor of Power Reactor and Nuclear Fuel Development Corporation (PNC)) has published the second progress report for high-level radioactive waste (HLW) disposal in Japan (H12 report) in November, 1999. This report is important to obtain the confidence of HLW disposal system and to establish the implementation body in 2000. JNC has developed databases of glass dissolution and radionuclide migration for performance analysis of the engineered barrier system (EBS) and the geosphere for H12 report. The databases developed for H12 report are of dissolution rates of high-level radioactive vitrified waste, thermochemical data of radioactive elements (JNC-TDB), sorption/diffusion data in the EBS and the geosphere. The database development has been focused on the repository conditions; reducing conditions and compacted/intact system, e.g., actinide (IV)/(III), derivation of sorption coefficients from diffusion experiments rather than batch sorption experiments. The JNC-TDB and sorption database have been developed under the auspices of international experts. The quality of these databases has been checked through independent individual experiments; glass leaching, solubility, batch sorption, diffusion experiments and through coupled leaching experiments by using the fully high-level radioactive glass and plutonium-doped glass which were sandwiched between compacted bentonite saturated with water. The maximum concentration of insoluble elements dissolved from the glass has also been investigated to check the quality of the JNC-TDB by comparison with solubility prediction. Based on these studies, JNC has determined the transport parameters for H12 report; dissolution rate of glass for a soluble radioactive element (Cs), solubility for insoluble radioactive elements (e.g., actinides, Tc), distribution coefficients and effective diffusion coefficients in the EBS and the geosphere.

1. Introduction

The objective of H12 report [1] for a high-level radioactive waste (HLW) geological disposal in Japan is specified to present the technical reliability of HLW geological disposal based on the progress made since the publication of the first progress report (H3 report) [2] in 1992. The H12 report has also provided scientific and technical basis for site selection and formulation of the regulatory infrastructures. The report consists of a Project Overview Report and three Supporting Reports that cover the three major fields; (1) evaluation of the geological environment, (2) repository design and engineering technology, (3) performance assessment. The performance assessment in the H12 report requires data for dissolution rate of vitrified waste, thermodynamic database and sorption/diffusion database of radioactive elements, which are reliable enough to analyze the performance of the engineered barrier (vitrified waste, bentonite) and natural barrier (rocks) under expected repository conditions.

This paper presents the status of database development for glass dissolution and radionuclide migration analysis and examples of studies on the quality check of databases, which are focused on the EBS. Since the databases in the geosphere have been developed in a

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similar way [3] as the EBS, except for sorption, no detailed information is supplied in this paper.

2. Concept of HLW geological disposal in Japan

JNC has emphasized that the multi-barrier system is the most appropriate approach to HLW disposal in Japan. The multi-barrier system consists of the EBS; the vitrified waste form, overpack, buffer material, and natural barrier (geosphere) (Fig. 1). The EBS should be located in a stable geological environment to perform robustly against possible perturbations and to reduce the role in the natural barriers which have their spacial heterogeneity. It is possible to enhance the overall performance of the multi-barrier system by selecting an appropriate site that satisfies the following conditions:

- Small groundwater flux.
- Reducing groundwater chemistry with pH neutral to slightly alkaline.
- Large retardation of radionuclide transport.
- Long groundwater travel path with large dilution.

3. Data for dissolution rate of the vitrified waste

The vitrified waste (glass) is surrounded by buffer material, compacted bentonite, and the release rate of dissolved silica from the glass is limited by molecular diffusion and sorption process in bentonite. The glass dissolution rate depends on the concentration of dissolved silica in the vicinity of the glass, which increases with reaction progress. Therefore, it is reasonable to set the value for the performance analysis by results from long-term leaching experiments. For a reference case analysis, the glass dissolution rate at 60°C was defined to be 1×10^{-3} g m^{-2} d^{-1} which was derived by a linear regression to the temperature dependence of the dissolution rate obtained from the long-term leaching experiments (Fig. 2). Based on a glass hydration model (e.g., [4]), the glass dissolution rate decreases over time.

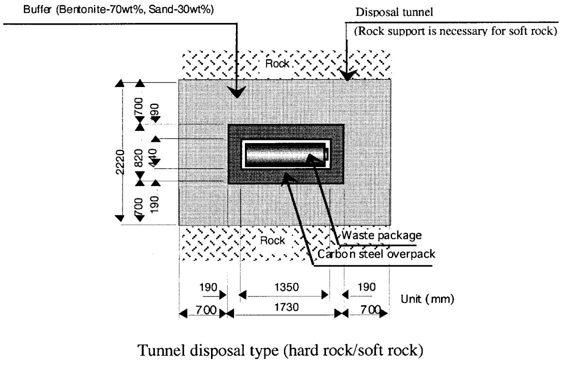

Fig. 1. HLW disposal concept (design example adopted in H12 report).

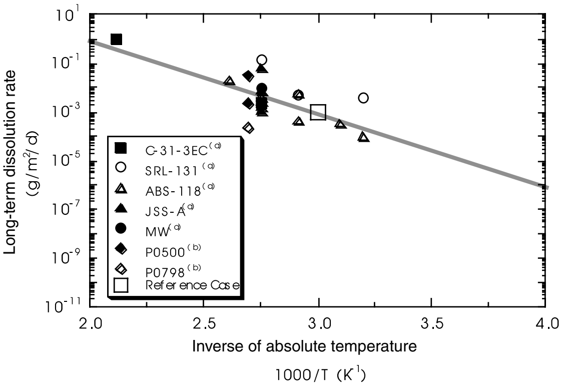

Fig. 2. The dissolution rates derived from long-term leaching experiments for some selected waste glasses. (Arrhenius plots of long-term dissolution rates are presented by Ohe et al. [34] originally. The linear regression curve is drawn as a best fit to: (a) data cited in the paper by Ohe et al. [34]; (b) data reported by Ishiguro and Masuda [35].)

The glass surface area is increased by cracking during cooling and fragmentation due to expansion of carbon–steel overpack corrosion products. The surface area of the fractured glass available for leaching was estimated to be five times that of the monolithic glass block based on experimental results [5].

4. Thermodynamic database and solubilities of radioactive elements

The solubilities of radioactive elements except for Cs were derived from thermodynamic database for radioactive elements (JNC-TDB) and reliable solubility measurements. For the concept of the HLW disposal in Japan, it is important to set solubilities/speciation of radioactive elements in porewater of the highly compacted bentonite and their speciation in rocks. Solubilities were used as upper limits of concentrations of radioactive elements, and speciation was used to determine or discuss the fundamental reactions for adsorptive characteristics of different geological materials.

4.1. JNC-TDB development

Critically reviewed thermodynamic databases with a traceable data selection are rare, and ongoing projects are slow to make progress. International efforts have been undertaken, 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’ (e.g., [6]), has been closed after the last publication in 1992. The OECD/NEA-TDB project has a high quality review standard and includes international reviewers, but its progress is slow. The first database was published in 1992 on uranium [7], the second one in 1995 on americium [8]. Databases on technetium, neptunium and plutonium are close to completion, but were not available in time for H12 report. JNC decided to start its own project to develop a thermodynamic database for radioactive elements (JNC-TDB). International experts have assisted JNC with the aim of developing critically reviewed database for a large number of elements: Ac, Am, Cm, Np, Pu, Sm, Th, U, Bi, Nb, Pb, Pd, Sb, Sn, Cs, Ni, Pa, Po, Ra, Se, Tc, and Zr. These elements were selected on the basis of their potential hazard index [5]. Key reactions in the solubility estimation of radioactive elements for the performance analysis of the geological disposal system are oxidation/reduction, hydrolysis, and complexation or precipitation with anions as carbonate, sulfate, phosphate, chloride. In H3 report for HLW disposal in Japan [2], a thermodynamic database for radioactive elements (PNC-TDB_H-3, [9]) was used to estimate their solubilities.

The procedure for the JNC-TDB development to focus on activity coefficient correction method is shown in Fig. 3. In all cases, an effort was made to compile all the available data.

In the case of trivalent (Ac(III), Am(III), Cm(III), Np(III), Pu(III), and Sm(III)) and tetravalent (Th(IV), U(IV), Np(IV), and Pu(IV)) actinides and rare earths, decisions were made primarily based on recent experimental data obtained in simple systems and validated in mixed and complex systems where the Pitzer model (e.g., [10]) was used. In the case of pentavalent actinides (Np(V) and Pu(V)), recently obtained data for Np(V) (e.g., [11]) using the Pitzer model approach were used as the basis for both Np(V) and Pu(V). In the case of hexavalent actinides (U(VI), Np(VI), Pu(VI)), recently available U(VI) data [7] were the primary reference source. In the case of Bi, Nb, Pb, Pd, Sb, and Sn, all available experimental data were evaluated, and selected experimental data were extrapolated to zero ionic strength using the Specific Ion Interaction Theory (SIT) model which has been described in the literature (e.g., [7,8]). Data for the remaining elements were compiled by JNC staff using primarily the existing compilations. For Ra and Po, the thermodynamic data sets are the same as the PNC-TDB_H-3, because no new thermodynamic data have been reported since 1992. For Tc, the OECD/NEA database will be adopted in the JNC-TDB in the near future. For Ni, Zr and Se, the thermodynamic data reported by [12] were the primary reference data, which are the same as the PNC-TDB_H-3.

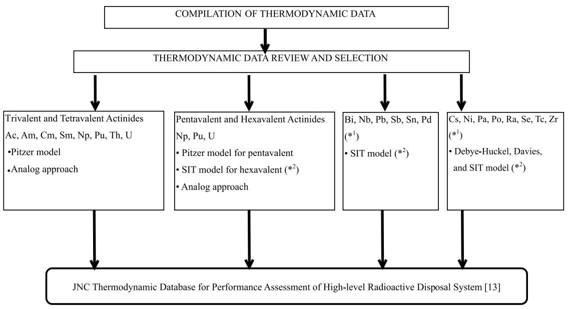

Fig. 3. Procedure for developing the thermodynamic database (JNC-TDB).

The data selection of the JNC-TDB was determined based on the following three criteria: (i) the reliability and traceability of thermodynamic data could be confirmed, (ii) data selections were carried out by using an available method for activity correction and (iii) in the case of data selection from other thermodynamic databases, the data were reviewed by experts.

The frozen JNC-TDB [13] has been improved of the traceability of the origin of the thermodynamic data and of its scientific reliability by checking the original studies from the following points of view:

- Identification of the solubility limiting solid phase.
- Description of pH, redox conditions, and concentrations of the ligands.
- Ionic strength and electrolyte media used in the experiments.
- Extrapolation of activity to zero ionic strength.

Moreover, the latest information has been incorporated as described in [13]. Consequently, the JNC-TDB has been improved mainly in relation to the following points:

- Traceability of all data was assured except for a few elements.
- Carbonate and hydroxy-carbonate complexes of the tetravalent actinides have been incorporated, which have been recently reported (e.g., [14,15]) and are important for high carbonate systems like porewater in compacted bentonite. This development procedure for tetravalent actinides is an extension of the Th thermodynamic data used for the safety assessment of the Waste Isolation Pilot Plant (WIPP) in the United States;
- Thermodynamic data for americium are mostly based on the OECD/NEA database [8] whose high reliability has been internationally recognized.
- Thermodynamic data for palladium, lead, and tin were reevaluated from original experimental values basically following the procedure of the NEA-TDB [16].
- For the trivalent lanthanide (Sm) and actinides (Ac, Pu and Cm), available thermodynamic data are limited and reliability of the data has not been established. Therefore, they are represented by the data for Am(III) based on chemical analogy.

In developing the JNC-TDB, the equilibrium constants at 25°C with the ionic strength of $I = 0$ were determined. However, the parameters concerning activity corrections have not yet been sufficiently established. Therefore, the Davies activity coefficient formula, which does not depend on the type of soluble chemical species, has been adopted. This formula is applicable up to an ionic strength of appropriately 0.5 molal [17]. It is thus applicable to the fresh-reducing-high pH groundwater (a reference groundwater in H12 report) whose ionic strength is 0.02 molal.

4.2. Solubilities of radioactive elements in bentonite porewater

The solubility of radioactive elements in porewater of compacted bentonite is determined by the following procedure:

- Estimate the solubility by using the geochemical code PHREEQE [18] with an appropriate thermodynamic database of the JNC-TDB and assuming a solubility-limiting solid phase.
- Values measured in experiments are used where they are available and greater than the calculated values.
- Effects of co-precipitation are considered for a relevant element (Ra).

The solubilities of safety-relevant elements in porewater determined for fresh-reducing-high pH groundwater are listed in Table 1 [19]. In the selection of solubility-limiting solid phases, thermodynamic stability and observations in solubility experiments are taken into account.

5. Diffusion data for radioactive elements in the EBS

Effective diffusion coefficients were determined considering selecting conservative value within the realistic range based on measured values. Effective diffusion coefficients in bentonite depend on the chemical species, and thus ideally they should be determined for each chemical species individually. The dependence of effective diffusion coefficients on bentonite density is shown in Fig. 4 [20–22]. However, the available data are limited considering the wide range of parameters and conditions, such as the mixing ratio of sand and density of the bentonite, redox conditions, porewater chemistry, and temperature. Therefore, an effective diffusion coefficient for each element has been determined by the following procedure:

1. When measured data for the relevant elements are available, the effective diffusion coefficient is determined based on the measured data. This is the case for Cs.
2. The effective diffusion coefficient is determined based on chemical analogy when measured data are unavailable, the chemical species in the porewater is

Table 1
Solubilities of safety-relevant elements in fresh-reducing-high pH porewater for the reference case

Element	Solubility (mol l^{-1} at 25°C)	Limiting solid[a]	Dominant species[a]
Se	3×10^{-9}	$FeSe_2$(cr)	HSe^-
Zr	1×10^{-6} [b]	ZrO_2(am)	$Zr(OH)_4$(aq)
Nb	1×10^{-4} [b]	Nb_2O_5(am)	$Nb(OH)_6^-$
Tc	4×10^{-8}	$TcO_2 \cdot 2H_2O$(am)	$TcO(OH)_2$(aq)
Pd	1×10^{-9} [b]	Pd(cr)	$Pd(OH)_2$(aq)
Sn	5×10^{-6} [b]	SnO_2(am)	$Sn(OH)_5^-$, $Sn(OH)_4$(aq)
Cs	Soluble	–	Cs^+
Sm	2×10^{-7}	$SmOHCO_3$(cr)	$Sm(CO_3)_2^-$, $Sm(CO_3)_3^{3-}$, $SmCO_3^+$
Pb	2×10^{-6}	$PbCO_3$(cr)	$PbCO_3$(aq), $Pb(CO_3)_2^{2-}$
Ra	1×10^{-12} [c]	Ra–Ca–CO_3	Ra^{2+}
Ac	2×10^{-7}	$AcOHCO_3$(cr)	$Ac(CO_3)_2^-$, $Ac(CO_3)_3^{3-}$, $AcCO_3^+$
Th	5×10^{-6}	ThO_2(am)	$Th(OH)_3CO_3^-$
Pa	2×10^{-8}	Pa_2O_5(s)	$PaO(OH)_3$(aq)
U	8×10^{-9}	UO_2(am)	$U(OH)_2(CO_3)_2^{2-}$, $U(OH)_4$(aq)
Np	2×10^{-8}	NpO_2(am)	$Np(OH)_2(CO_3)_2^{2-}$, $Np(OH)_4$(aq)
Pu	3×10^{-8}	PuO_2(am)	$Pu(OH)_2(CO_3)_2^{2-}$, $Pu(CO_3)_2^-$, $Pu(CO_3)_3^{3-}$, $PuCO_3^+$
Am	2×10^{-7}	$AmOHCO_3$(cr)	$Am(CO_3)_2^-$, $Am(CO_3)_3^{3-}$, $AmCO_3^+$
Cm	2×10^{-7}	$CmOHCO_3$(cr)	$Cm(CO_3)_2^-$, $Cm(CO_3)_3^{3-}$, $CmCO_3^+$

[a] (cr): Crystalline, (am): amorphous, (s): solid that cannot be identified as either crystalline or amorphous in the original literature, (aq): aqueous species with neutral charge [13].

[b] Measured value.

[c] The measured values in natural groundwater are adopted as a conservative value, based on the comparison with solubility calculated using simple co-precipitation model of Ra with Ca.

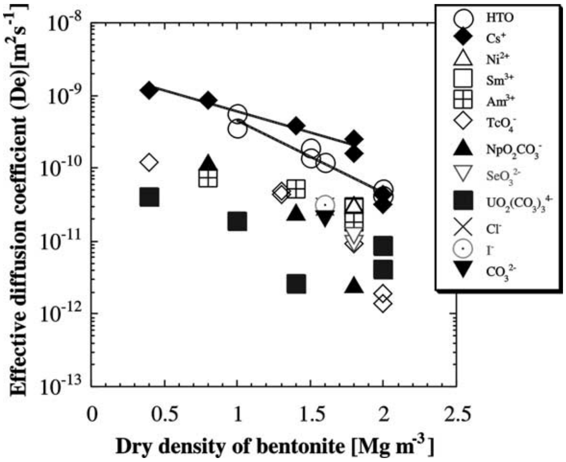

Fig. 4. Effective diffusion coefficients as a function of bentonite dry density.

simple, and a suitable analog is available. This is the case for Se based on chemical analogy to I.

3. The effective diffusion coefficient should be determined based on tritiated water (HTO) data conservatively when measured data are unavailable, the chemical species in the porewater are neutral, or complex and two or more chemical species exist. This is the case for many elements except for Cs and Se.

Table 2 lists the effective diffusion coefficients determined based on the above consideration. Values at a temperature of 60°C were used for the performance analysis. However, those at 25°C are also shown as they were used in determination of the distribution coefficients for lack of data.

6. Sorption database and distribution coefficients of radioactive elements in the EBS

6.1. Sorption database development of batch measurements

Sorption databases for radioactive elements have been internationally developed through the OECD/NEA project: the NEA Sorption DataBase (NEA-SDB) development which replaced the former International Sorption Information Retrieval System initiated since 1981. The database of distribution coefficients (K_d) of radionuclides on various geological materials like NEA-SDB was not very useful for performance assessment because of only availability of the conditions where the data have been obtained, because K_d values depend on various conditions, e.g., pH, Eh, ionic concentrations, rock properties, specific surface, solid/liquid ratio. In the OECD/NEA Workshop on sorption in 1991, some scientists have proposed mechanistic model development in order to understand the sorption process in a generic way [23]. JNC has initiated the sorption program to focus on mechanistic understanding that is consistent with the thermodynamic database of radioactive elements. The program is based on four steps: (i) surface characterization of dominant adsorbents, (ii) mechanistic understanding of sorption and derivation of its constants by batch sorption measurements for simple system, e.g., pure minerals, (iii) validation of mechanistic model/constants by batch measurements for complex system, e.g., bentonite, rocks, (iv) validation of mechanistic model/constants by diffusion measurements for complex systems. The correction of solid/liquid ratio for K_d values in bentonite is not so easy by conversion of batch sorption data to those in highly compacted bentonite due to the microstructural effects of the bentonite on porewater chemistry. On the other hand, it is probably relevant to use batch sorption data directly for an intact rock, because the pore volume is significantly larger than diffusive chemical species (e.g. [24]).

6.2. Selection of K_d values for bentonite

There are two ways to derive the distribution coefficient of compacted bentonite, namely to measure it directly in batch sorption measurements or to derive it from measurements of the apparent diffusion coefficient. As the data selection should be carried out under consideration of the properties of compacted bentonite, it is more relevant to use the latter method. There are also many arguments of reliability of K_d models, however, this model is simple to handle for performance analysis and is applicable under careful consideration of repository conditions. The distribution coefficient is calculated

Table 2
Effective diffusion coefficients for safety-relevant elements in the reference case

Element	Species	Effective diffusivity ($m^2\ s^{-1}$) at 25°C	Effective diffusivity ($m^2\ s^{-1}$) at 60°C
Cs	Cs^+	2.7×10^{-10}	5.4×10^{-10} (6×10^{-10})[b]
Se	HSe^-	6.0×10^{-11}	1.2×10^{-10} (2×10^{-10})[b]
Other elements		1.2×10^{-10} [a]	2.4×10^{-10} (3×10^{-10})[b]

[a] The value for tritiated water (HTO) is adopted.

[b] The temperature correction is based on the activation energy for diffusion (approximately 15.05 KJ mol^{-1}). The values in parentheses are rounded up to one significant figure.

from the apparent and effective diffusion coefficients using the following equation:

$$K_d = \frac{1}{\rho}\left(\frac{D_e}{D_a} - \varepsilon\right),$$

where K_d is the distribution coefficient, D_e is the effective diffusion coefficient, D_a is the apparent diffusion coefficient, ρ the dry density of bentonite, and ε is the total porosity.

Effective diffusion coefficients (D_e) used in these calculations were determined using the same approach as discussed above. For Cs, the measured D_e value for Cs was used. For Se, the D_e value for I^- as an analog element was used. For the other elements where measured values or chemical analog do not exist, the value for HTO was used. Adopting an effective diffusion coefficient, which does not correspond to the actual diffusing species, leads to a distribution coefficient (K_d) which includes effects other than sorption. However, appropriate non-steady-state retardation analysis can be described by using the consistent effective diffusion coefficients with the measured D_a value, and using the effective coefficient of HTO is conservative for the steady-state diffusion case.

Apparent diffusion coefficients for redox-sensitive elements have been derived from measurements with consideration of the expected redox conditions in a repository. The dependence of distribution coefficients on the dry density was also considered. In the case where the distribution coefficient of an element is density-dependent (e.g., Cs, Sm, Ra, Am), an empirical formula representing the relationship between $\log D_a$ and dry density was derived and interpolated or extrapolated to a dry density of 1600 kg m^{-3} (reference design density of bentonite in H12 report). Then, a lower bound distribution coefficient at this density was chosen. For elements whose distribution coefficient is not density-dependent or its density-dependence is not well known (e.g., Se, Zr, Nb, Tc, Pd, Sn, U, Np), a lower bound distribution coefficient was selected directly from all the data. Although the D_a for Pu has been measured only for one value of dry density, 400 kg m^{-3} [25], K_d tendency has been observed to increase with density for another bentonite (Kunipia-F) with higher smectite content [22]. For U and Np, both of which are tetravalent actinides under reducing conditions, the data on apparent diffusion coefficients were discussed simultaneously, and the same values were chosen considering their chemical analogy.

The reference material used is Na-type bentonite produced at Tsukinuno, Yamagata Prefecture for which many reliable data are available.

No major effects of the 30% silica sand content have been observed on the apparent diffusion coefficients of elements, such as Cs, Se, HTO. Therefore, apparent diffusion coefficients taken from the test samples containing no silica sand have been used without correction.

The porewater chemistry measured under laboratory conditions does not necessarily agree with that used in the performance analysis. Porewater under experimental conditions is assumed to have higher ionic strength and slightly lower pH than the reference case porewater assumptions [26]. However, this was judged to be conservative because the laboratory experimental conditions would not facilitate much sorption of cations.

Based on the results of batch measurements, the distribution coefficients of some elements are known to depend on temperature. For example, Baston et al. [27] studied the temperature dependence of Tc, U, Np and Cm under reducing conditions. The distribution coefficient of Tc was found to be one order of magnitude higher at 60°C than at 25°C. However, the temperature dependence of Np, U, and Cm has not been determined. The K_d of Cs is known to decrease as temperature increases. For example, Tukamoto and Ohe [28] have reported that the distribution coefficient of Cs decreased by approximately 40% as the temperature increased from 20°C to 70°C. This can also be calculated from the activation energy of apparent diffusion in bentonite [29] and the activation energy of diffusion in free water. Thus, it has been decided to include the effect of temperature when the distribution coefficient decreases significantly as temperature increases.

For those elements whose measured apparent diffusion coefficients were not available, data have been adopted by chemical analogy among the elements and their dissolved chemical species in porewater derived

Table 3
Distribution coefficients (K_d) for buffer material in the reference case (m^3 kg^{-1})

Element	Distribution coefficient	Note (chemical analogy)
Se	0	
Zr	10	
Nb	1	
Tc	0.1	
Pd	0.1	
Sn	1	
Cs	0.01	
Sm	1	
Pb	0.1	Ni
Ra	0.01	
Ac	1	Sm
Th	1	U, Np
Pa	1	Sn
U	1	
Np	1	
Pu	10	
Am	10	
Cm	10	Am

from thermodynamic calculations. The data for Ni, Sm, Sn, U, Am are related to those for Pb, Ac, Pa, Th, Cm, respectively. Table 3 lists the distribution coefficients for the reference case analysis. The values have been rounded to 1×10^n or 5×10^n (where n is an integer).

When values of K_d derived from apparent diffusion coefficients were compared with those obtained from batch measurements under conditions similar to those in the reference case, the value of the batch measurement K_d was, in general, higher. The lowest values in batch measurements were higher than the values set above in Table 3 for most elements. Therefore, the K_d values set for elements are conservative with respect to experimental measurements, because lower values were chosen from among measured variations. Mechanistic sorption models such as ion-exchange models (e.g. [30]) were also applied to support the data selection.

7. The quality of the databases

Quality of migration databases can be checked through independent experiments. Examples of studies on the quality check are described below for leaching rates of glass, thermodynamic database and K_d model (values). An example of the quality check of thermodynamic data for trivalent actinides, such as Am(III), was performed by comparison with studies by Rai et al. [31] using Am-doped glass. The two following coupled experiments were performed in JNC to check the quality of leaching rates of glass, thermodynamic database and K_d model (values) simultaneously.

Ashida et al. [32] have studied the coupled behavior of Pu released from the Pu-doped borosilicate glass surrounded by the compacted bentonite saturated with distilled water. The experimental results are shown in Fig. 5. Under these conditions, leaching of Pu from the glass, diffusion and sorption of Pu in the compacted bentonite occur simultaneously. The experimental results were compared with blind prediction by transport model using individual parameters; leaching rate of the glass, solubility, sorption and diffusion coefficient of Pu. The predicted diffusion profiles are similar to experimental results as shown in Fig. 5. They have also compared MCC-1 experimental results of Pu with modelling results of glass dissolution and Pu solubility. Ashida et al. [33] have also studied the coupled behavior of Cs released from the fully high-level radioactive waste (HLRW) glass surrounded by the compacted bentonite saturated with distilled water. As shown in Fig. 6, the experimental diffusion profiles are also similar to the modelling results that were conducted in the same way as for Pu, except for the assumption of congruent dissolution of Cs with the glass. These two coupled experimental results increase the confidence in the quality of the individual database and of the models used for performance analysis of the EBS.

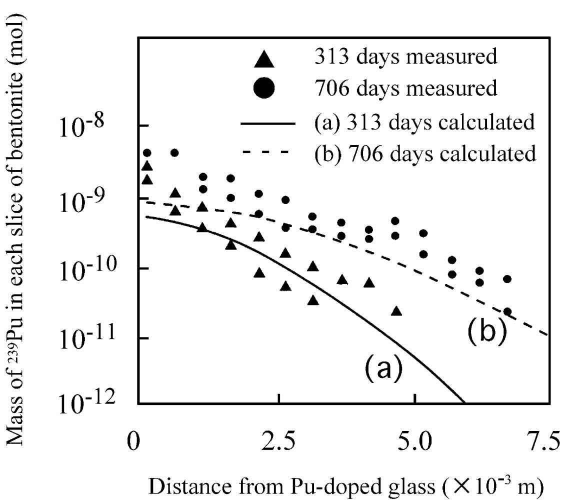

Fig. 5. Measured profiles of quantity of ^{239}Pu in each slice of bentonite and results of independent transport blind predictions (the thickness of each slice: 0.5 mm).

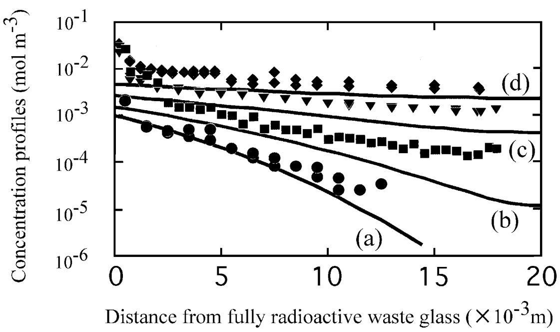

Fig. 6. Measured concentration profiles for ^{137}Cs in compacted bentonite at experimental periods of 15(●), 35(■), 110(▼) and 300(◆) days and modeling results for a dry density of 1000 kg m^{-3}. Concentration profiles by independent calculation are represented as lines (a)–(d) for each period.

8. Conclusion

The database for dissolution rate of vitrified waste, thermodynamic database (JNC-TDB), sorption/diffusion database for radioactive elements in the EBS and the geosphere have been developed for H12 report. The database development has been focused on the repository condition; reducing conditions and compacted/intact system, e.g., actinide (IV)/(III), derivation of sorption coefficients from diffusion experiments rather than batch sorption experiments. The JNC-TDB and sorption database have been developed under the auspices of international experts.

The quality of these databases has been checked through individual experiments e.g., glass leaching, solubility experiments, coupled leaching experiments by using the fully HLRW and Pu-doped glass which were sandwiched by the compacted bentonite saturated with water.

Based on these studies, JNC has determined the sets of transport parameters to be used in the H12 report; dissolution rate of glass for soluble radioactive element (Cs), solubility for insoluble radioactive elements (e.g., actinides, Tc), distribution coefficients and effective diffusion coefficients in the EBS and the geosphere.

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References

[1] Japan Nuclear Cycle Development Institute (JNC), H12 project to establish technical basis for HLW disposal in Japan, Project Overview Report, JNC TN1400 99-010, 1999.

[2] Power Reactor and Nuclear Fuel Development Corporation (PNC), Research and development on geological disposal of high-level radioactive waste, First progress report, PNC Technical Report, TN1410 93-059, 1992.

[3] T. Shibutani, T. Suyama, M. Shibata, Sorption data-base for radionuclides on bentonite and rocks, JNC Technical Report, TN8410 99-050, 1999 (in Japanese).

[4] Y. Inagaki, H. Furuya, K. Idemitsu, S. Yonezawa, J. Nucl. Mater. 208 (1994) 27.

[5] Power Reactor and Nuclear Fuel Development Corporation (PNC), Current status of research and development on geological disposal (FY1989), PNC Technical Report, TN1410 91-009, 1991 (in Japanese).

[6] J. Fuger, I.L. Khodakovsky, E.I. Sergeyeva, V.A. Medvedev, J.D. Navratil, The Chemical Thermodynamics of Actinide Elements and Compounds: Part 12. The Actinide Aqueous Inorganic Complexes, International Atomic Energy Agency, Vienna, Austria, 1992.

[7] I. Grenthe, J. Fuger, R.J.M. Konings, R.J. Lemire, A.B. Muller, C. Nguyen-Trung, H. Wanner, The Chemical Thermodynamics of Uranium, OECD Nuclear Energy Agency, North-Holland, Amsterdam, 1992.

[8] R.J. Silva, G. Bidoglio, M.H. Rand, P.B. Robouch, H. Wanner, I. Puigdomenech, Chemical Thermodynamics of Americium, OECD/NEA, Elsevier, Amstredam, 1995.

[9] M. Yui, H. Makino, T. Ashida, H. Umeki, K. Ishiguro, A. Neyama, Dissolution behavior of vitrified waste and evaluation of radionuclides solubilities in near-field porewater chemistry, PNC Technical Report, TN8410 92-161, 1992 (in Japanese).

[10] K.S. Pitzer, J. Phys. Chem. 77 (2) (1973) 268.

[11] T. Fanghaenel, V. Neck, J.I. Kim, Radiochim. Acta 69 (1995) 169.

[12] J.E. Cross, F.T. Ewart, C.J. Tweed, Thermodynamic modelling with application to nuclear waste processing and disposal, AERE-R 12324, 1987.

[13] M. Yui, J. Azuma, M. Shibata, JNC thermodynamic database for performance assessment of high-level radioactive waste disposal system, JNC Technical Report, TN8400 99-070, 1999.

[14] D. Rai, N.J. Hess, A.R. Felmy, D.A. Moore, M. Yui, Radiochim. Acta 84 (1999) 159.

[15] D. Rai, N.J. Hess, A.R. Felmy, D.A. Moore, M. Yui, Radiochim. Acta 86 (1999) 89.

[16] B. Lothenbach, M. Ochs, H. Wanner, M. Yui, Thermodynamic data for the solubility and speciation of Sn, Sb, Pb, Bi, Nb, and Pd in aqueous solution, JNC Technical Report, TN8400 99-011, 1999.

[17] W. Stumm, J.J. Morgan, Aquatic Chemistry, Chemical Equilibria and Rates in Natural Waters, 3rd Ed., Wiley, New York, 1996, p. 103.

[18] D.L. Parkhurst, D.C. Thorstnsen, L.H. Plumer, PHREEQE - A Computer Program for Geochemical Calculations US Geological Survey Water-Resources Investigations (1980) 80.

[19] J. Azuma, M. Shibata, M. Yui, T. Shibutani, S. Notoya, Y. Yoshida, Solubility and speciation of radioactive elements of high-level radioactive waste disposal system, JNC Technical Report, TN8400 99-071, 1999.

[20] H. Fujiwara, I. Yasutomi, H. Kato, S. Ueta, Proc. 1998 Annual Meeting of the Atomic Energy Society of Japan, L25, 1998 (in Japanese).

[21] H. Sato, T. Shibutani, Study on adsorption and diffusion mechanism of nuclides in buffer material and geosphere, PNC Technical Review, TN8410 94-284 (1994) 71 (in Japanese).

[22] H. Sato, Nuclide diffusion/adsorption data and its mechanism in buffer material and rocks, Proceedings for the Information Exchange Workshop for Performance Assessment, PNC Technical Report, TN1410 91-079 (1991) 275 (in Japanese).

[23] OECD/NEA, Proc. Radionuclide Sorption from the Safety Evaluation Perspective, Proceedings of a NEA Workshop, 1992.

[24] T. Yamaguchi, H. Isobe, S. Nakayama, Radioactive Waste Res. 3 (2) (1997) 99 (in Japanese).

[25] Y. Tachi, Y. Kohara, N. Uchidate, T. Shibutani, diffusion behavior of plutonium and americium in bentonite, JNC Technical Report, TN8400 99-089, 1999 (in Japanese).

[26] C. Oda, M. Shibata, Modelling of bentonite–water interaction experiments under anaerobic conditions, JNC Technical Report, TN8400 99-032, 1999 (in Japanese).

[27] G.H.N. Baston, J.A. Berry, M. Brownsword, T.G. Heath, C.J. Tweed, M. Yui, Mater. Proc. Res. Soc. Symp. 465 (1997) 805.

[28] M. Tukamoto, T. Ohe, in: Papers on Basic Data for Nuclide Migration V, Atomic Energy Society of Japan (Eds.), RWM-90014, 1990, p. 34 (in Japanese).

[29] T. Kozaki, H. Sato, A. Fujishima, S. Sato, H. Ohashi, J. Nucl. Sci. Technol. 33 (1996) 522.

[30] M. Ochs, M. Boonekamp, H. Wanner, H. Sato, M. Yui, Radiochim. Acta 82 (1998) 437.

[31] D. Rai, J.A. Schramke, D.A. Moore, G.L. McVay, Nucl. Technol. 75 (1986).

[32] T. Ashida, Y. Kohara, M. Yui, Radiochim. Acta 66&67 (1994) 359.

[33] T. Ashida, Y. Kohara, T. Shibutani, M. Yui, Migration behavior of cesium released from fully radioactive waste glass in compacted sodium bentonite, PNC Technical Report, TN8410 98-014, 1998.

[34] T. Ohe, M. Tsukamoto, M. Kinoshita, T. Inoue, Waste Management 11 (1999) 191.

[35] K. Ishiguro, S. Masuda, A study of long-term performance of waste glass under repository conditions, PNC Technical Report, PNC TN4410 90-006 (1990) (in Japanese).