Database Development
of Chemical Thermodynamics of Protactinium
for Performance Assessment
of HLW Geological Disposal System

March , 1998

TOKAI WORKS

POWER REACTOR AND NUCLEAR FUEL DEVELOPMENT CORPORATION

Ⓚ

Database Development
of Chemical Thermodynamics of Protactinium
for Performance Assessment
of HLW Geological Disposal System

Tomoki Shibutani*, Sanae Shibutani* and Mikazu Yui*

ABSTRACT

In the performance analysis of geological disposal system of high-level radioactive waste (HLW), solubilities of radioactive elements are estimated by thermodynamic calculation. The reliable thermodynamic database (TDB) is needed for solubility estimation. In this report, thermodynamic data for protactinium solid and aqueous species for performance assessment were selected.

For the refinement of previous PNC in house thermodynamic database (PNC-TDB), existing literatures data were surveyed and reliable thermodynamic data were selected under consideration of the scientific defensibility and the consistency with the whole PNC-TDB. The estimated solubility using refined PNC-TDB was higher than measured value. We have confirmed the refined data-set of Pa to be conservative for solubility estimation of performance assessment.

*; Geological Isolation Technology Section, Waste Technology Development Division,
   Tokai Works, Power Reactor and Nuclear Fuel Development Corporation,
   Tokai-mura, Ibaraki-ken, 319-1112, JAPAN

高レベル廃棄物地層処分システムの性能評価のための
プロトアクチニウムに関する熱力学データ整備

報告者；澁谷朝紀*、
　　　　澁谷早苗*、
　　　　油井三和*

要　旨

　高レベル放射性廃棄物の地層処分システムの性能評価においては、元素の溶解度を熱力学計算により評価している。この計算には信頼性の高い熱力学データベースが必要とされる。本報においては、プロトアクチニウム(Pa)についての熱力学データ整備を行ったので報告する。

　整備においては、まず、Paの固相と水溶性化学種についての熱力学データを文献より収集した。収集されたデータについては、科学的信頼性について検討され、動燃の熱力学データベース(PNC-TDB)全体との整合性についてもチェックされた後、PNC-TDBに取り込まれた。更新されたPNC-TDBを用いて計算したPaの溶解度は、実験値よりも高くなったが、地層処分安全評価においては、保守的な評価となるため、適当なデータセットと判断された。

---

*; 動力炉・核燃料開発事業団　東海事業所
　　環境技術開発部　地層処分開発室

CONTENTS

TABLE

FIGURE

1. INTRODUCTION

The equilibrium constants for chemical reaction of protactinium are important for performance assessment of geological isolation system of high-level radioactive waste. These values were compiled from various studies. The related databases are shown as follows.

(1) CHEMVAL database (Falck, 1992)
(2) HATCHES ver.10 database (Harwell/Nirex,1997)
(3) PNC Thermodynamic Database (PNC-TDB) (Yui et al., 1992).

The thermodynamic data of protactinium have already been collected in the PNC-TDB (Yui et al.,1992), and these data were cited from Cross et al.(1987). It is necessary to confirm the reliability of thermodynamic data and to revise the data based on recent studies. In this paper, the thermodynamic data of protactinium for solubility estimation were selected. The comparison were also presented between experimental solubility measurements in the literature and the calculation results using a geochemical code PHREEQE (Parkhurst et al., 1980) with the refined PNC-TDB.

2. COLLECTION OF DATA

In order to keep the quality and reliability of the database used for performance assessment of the radioactive waste disposal, individual data should be checked. The reliability of individual sources and experimental data were checked in accordance with the following guideline.

(1) the identification of the solid phase is appropriately carried out or not ?
(2) the atmospheric condition (O_2, CO_2 concentration) is clarified or not?
(3) the ionic strength is clarified or not?
(4) the pH, Eh, cation, anion concentration are measured or not?
(5) the reducing reagent is appropriately used or not?
(6) the solid-liquid separation methods such as the filtration are clarified or not?
(7) the temperature is clarified or not?
(8) the redox state and speciation are clarified or not?

Furthermore, the reliable thermodynamic data should be selected under consideration of the consistency with the whole PNC-TDB. The thermodynamic data were selected from the following view point as possible.

(1) consistency with chemical analogy and theoretical consideration

(2) conservatism for solubility estimation of performance assessment

(3) validity for experimental data

(4) comprehensiveness by comparison with the other international database

3. CHEMISTRY OF PROTACTINIUM

The solution chemistry of protactinium has been reviewed in several studies (Guillaumont et al.,1968, Baes and Mesmer,1976, Fuger and Oetting,1976, Kirby,1986). Two oxidation states, Pa(V) and Pa(IV), have been definitely confirmed in aqueous solution, but the existence of Pa(III) has not been observed in aqueous solution (Kirby,1986). Pentavalent protactinium differs from the other actinides of which pentavalent state forms the well known MO_2^+ species. In the case of dilute protactinium ($\leq 10^{-5}M$), non-complexing solutions such as perchloric media, or in the presence of weakly complexing anions at pH$\leq$0, the predominant species is probably $PaOOH^{2+}$ (Fuger and Oetting,1976). At $H^+<1M$, $PaO(OH)_2^+$ begins to form and becomes predominant at pH~3. At higher pH values, the neutral species, $Pa(OH)_5(aq)$, is formed. At tracer levels, these species are in equilibrium, but, at concentrations of Pa(V) close to saturation, polymers are formed rapidly and the process is irreversible. At pH 5-6, the hydrous oxide is precipitated. In alkaline solution (ionic strength μ=0.1), $Pa(OH)_6^-$ are formed slightly, and at $H^+>3M$, the existence of PaO_3^+ has been suggested (Kirby,1986). In more concentrated protactinium solutions or less actinide media, polymerization occurs, with the final step being the precipitation of hydrous oxide. This difference in behavior of protactinium as compared to the other pentavalent actinides is probably due to the difference in availability of the 5f orbitals for forming M-O bonds.

Protactinium hydrolysis also occurs in complexing media such as hydrochloric, sulfuric or nitric acid.

The hydrolysis and complexation behavior of tetravalent protactinium, although less complicated, has been studied. From a study at the tracer level (Guillaumont et al.,1968), tetravalent protactinium appears distinctly more acidic than the heavier actinides with a pK of 0.4 (μ=3, perchloric medium) for the first hydrolysis reaction of $Pa(OH)^{3+}$. The trivalent protactinium is unstable in aqueous solution, and standard oxidation potential (E^0) of Pa^{4+}/Pa^{3+} was estimated very high values as -1.8V ~ -2.1V (Inova et al.,1992, Nugent et al.,1973). Therefore, Pa(III) has not been identified in the aqueous solution.

4. OXIDATION AND REDUCTION REACTION

4.1 Entropy, enthalpy and free energy of formation

Thermodynamic data of protactinium such as entropy, enthalpy and free energy of formation have been compiled by Fuger and Oetting(1976). These data are shown in Table 4.1.

Table 4.1 Entropy, enthalpy and free energy of formation

	ΔH_f (kJ/mol)	ΔG^0_f (kJ/mol)	S^0 (J/K/mol)
Pa^{4+}	-619.96	-565.12	-397.68
$PaOOH^{2+}$	-1113.50	-1050.71	-20.93

4.2 The standard oxidation potential

The standard oxidation potential of Pa has been obtained for Pa^{3+}/Pa^{2+}, Pa^{4+}/Pa^{3+} and Pa^{5+}/Pa^{4+} couple by electrochemical technique and theoretical calculation. These value are shown in Table 4.2.

Table 4.2 Standard oxidation potential

	E^0 (V)	References
Pa^{3+}/Pa^{2+}	-2.59	Mikheev et al., 1994
Pa^{4+}/Pa^{3+}	-2.0	Bratsch and Lagowski ,1986
Pa^{4+}/Pa^{3+}	-1.8	Inova et al., 1992
Pa^{4+}/Pa^{3+}	-2.1	Nugent et al., 1973
Pa^{5+}/Pa^{4+}	-0.1	Fried and Hindman ,1964

4.3 Oxidation and reduction

(1) Pa^{3+}/Pa^{4+}

The redox reaction equilibrium constant between Pa^{3+} and Pa^{4+} was estimated by the following reaction.

$$Pa^{4+} + e^- \leftrightarrow Pa^{3+} \qquad \log K=2.03 \qquad (4.3.1)$$

The log K value in Cross et al.(1987) was significantly different from that calculated from standard oxidation potential. The value of Cross et al.(1987) is not likely appropriate, because experimentaly observed standard oxidation potential value is very high as shown in Table 4.3.1.

Species of Pa(III) should not be included in the revised PNC-TDB.

Table 4.3.1 The redox equilibrium constant between Pa^{3+} and Pa^{4+}

References	logK
Cross et al., 1987	2.03
Bratsch and Lagowski,1985	33.90
Inova et al.,1992	30.51
Nugent et al.,1971	35.59

(2) PaO_2^+/Pa^{4+}

The redox equilibrium constant between PaO_2^+ and Pa^{4+} was estimated by following reaction.

$$Pa^{4+}+2H_2O \leftrightarrow PaO_2^++4H^++e^- \tag{4.3.2}$$

The equilibrium constant for equation (4.3.2) was estimated from dissolution of $Pa_2O_5(s)$ by Baes and Mesmer (1976). However, Guillaumont et al. (1968) reported that the existence of PaO_2^+ was not confirmed from the solvent extraction. PaO_2^+ was not considered in the review report of Kirby (1986). The equilibrium constant of PaO_2^+ for equation (4.3.2) are shown in Table 4.3.2.

Table 4.3.2 The redox equilibrium constant between PaO_2^+ and Pa^{4+}

References	logK
Cross et al.,1987	7.79
HATCHES ver. 10	2.36

Cross et al.,1987 selected the value from Gmelin Handbook (Brown et al.,1977). HATCHES ver.10 adopted the value from Baes and Mesmer (1976). However, PaO_2^+ was not identified in the experimental system, so the equilibrium constant of PaO_2^+ described by equation (4.3.2) is not included in the refined PNC-TDB.

5. HYDROLYSIS REACTION

5.1 Hydrolysis reaction of Pa(IV)
5.1.1 Solid phase
(1) $PaO_2(c)$

The solubility product of $PaO_2(c)$ was estimated by following reaction.

$$PaO_2(c)+4H^+ \leftrightarrow Pa^{4+}+2H_2O \qquad (5.1.1)$$

Guillaumont et al. (1968) reported an upper limit of $10^{-5}M$ for the concentration of Pa(IV) in 3M $HClO_4$. If $PaO_2(c)$ was precipitating phase, this would suggest a value of logK for equation (5.1.1), -6.9. Baes and Mesmer (1976) expected much higher value of logK from an interpolation of the estimated value for ThO_2 and UO_2 assuming that logK changed linearly as the reciprocal of the lattice parameter of these oxides. CHEMVAL, HATCHES ver.10, and Cross et al. (1987) are refered the value from Baes and Mesmer (1976) for solubility product of $PaO_2(c)$ for equation (5.1.1).

Table 5.1.1 The solubility product of $PaO_2(c)$

References	logK
Guillaumont et al.,1968	-6.9
Baes and Mesmer,1976	0.6

The log K value for equation (5.1.1) is selected,0.6 for the refined PNC-TDB.

5.1.2 Aqueous phase

The hydrolysis of Pa(IV) has been examined in perchlorate solution by Guillaumont et al. (1968) and by Lundqvist (1974) by extracttion method using thenoyltrifluoroacetone (TTA) and acetylacetone in benzene, respectively. Baes and Mesmer (1976) calculated the equilibrium constant of hydrolysis reactions of Pa(IV) based on the data from Guillaumont et al. (1968). Theoretical consideration was also applied for hydrolysis reactions of Pa(IV) (Brown and Wanner, 1987). The equilibrium constant for hydrolysis reaction of Pa(IV) are compiled in Table 5.1.2. HATCHES ver.10 and Cross et al. (1987) are refered the value from Baes and Mesmer (1976).

Table 5.1.2 The equilibrium constant for hydrolysis reaction of Pa(IV)

	PaOH^{3+}	Pa(OH)$_2^{2+}$	Pa(OH)$_3^+$	Pa(OH)$_4$
Baes and Mesmer,1976	0.84	-0.02	-1.5	
CHEMVAL database	0.66	0.8	-1.5	no data
Brown and Wanner,1987	-5.37	-2.41	-8.78	

For realistic solubility estimation, experimental results should be preferred more than theoretical calculation. The equilibrium constant for hydrolysis reactions of Pa(IV) reported by Baes and Mesmer (1976) is selected for the refined PNC-TDB.

5.2 Hydrolysis reaction of Pa(V)

5.2.1 Solid phase

(1) $Pa_2O_5(s)$

Guillaumont et al. (1968) reported that the solubility of Pa(V) in perchlorate media below pH=3 is about 10^{-5}M. The solubility of well-crystalline $Pa_2O_5(s)$ is not known but it is likely to be considerably lower than this value (Baes and Mesmer, 1976). The solubility product of $Pa_2O_5(s)$ for equation (5.2.1) is shown in Table 5.2.1

$$Pa_2O_5(s)+10H^++2e^- \leftrightarrow 2Pa^{4+}+5H_2O \tag{5.2.1}$$

Table 5.2.1 The solubility product of Pa_2O_5 (s)

References	logK
HATCHES ver. 10	-8.72
Cross et al.,1987	-19.58

The solubility product from HATCHES ver. 10 did not agree with that from Cross et al.(1987), because these values were calculated by using different value for PaO_2^+/Pa^{4+}.
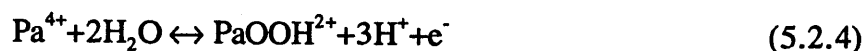
Baes and Mesmer (1976) estimated the equilibrium constant for equation (5.2.2) as logK$\leq$2.

$$1/2Pa_2O_5(s)+H^+ \leftrightarrow PaO_2^++1/2H_2O \tag{5.2.2}$$
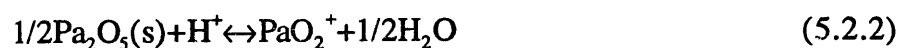
Baes and Mesmer,1976 also estimated the equilibrium constant for equation (5.2.3) as logK=0.5.

$$PaOOH^{2+} \leftrightarrow PaO_2^+ + H^+ \qquad\qquad (5.2.3)$$

The formation constant of $PaOOH^{2+}$ for equation (5.2.4) can be calculated as logK=1.86 from free energy of formation (Table 4.1).

$$Pa^{4+} + 2H_2O \leftrightarrow PaOOH^{2+} + 3H^+ + e^- \qquad\qquad (5.2.4)$$

The hydrolysis constant of Pa_2O_5 for equation (5.2.1) can be calculated as logK≤-8.72 from equation (5.2.2), (5.2.3) and (5.2.4). This maximum value is selected for the refined PNC-TDB.
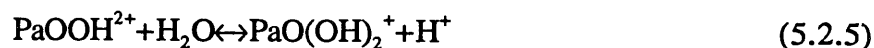
5.2.2 Aqueous phase
(1) PaOOH^{2+}

The formation constant of $PaOOH^{2+}$ for equation (5.2.4) is calculated as logK=1.86 from free energy of formation (Table 4.1) as the foregoing method.

(2) PaO(OH)$_2^+$

Guillaumont et al. (1968) studied the dissolution behavior of Pa(V) in 3M $LiClO_4$ and Fuger (1992) estimated the formation constant of $PaO(OH)_2^+$ for equation (5.2.5) as logK=-1.05.
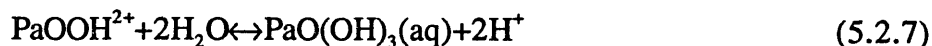
$$PaOOH^{2+} + H_2O \leftrightarrow PaO(OH)_2^+ + H^+ \qquad\qquad (5.2.5)$$

The formation constant of $PaO(OH)_2^+$ for equation (5.2.6) is calculated from equation (5.2.4) and (5.2.5) as logK=0.85.

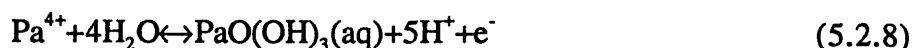$$Pa^{4+} + 3H_2O \leftrightarrow PaO(OH)_2^+ + 4H^+ + e^- \qquad\qquad (5.2.6)$$
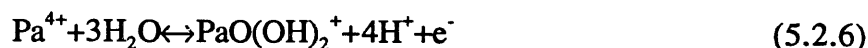
(3) PaO(OH)$_3$(aq)

Guillaumont et al. (1968) studied the dissolution behavior of Pa(V) in 3M $LiClO_4$ and Fuger (1992) estimated the formation constant of $PaO(OH)_3(aq)$ for equation (5.2.7) as logK=-5.5.

$$PaOOH^{2+} + 2H_2O \leftrightarrow PaO(OH)_3(aq) + 2H^+ \qquad\qquad (5.2.7)$$

The formation constant of $PaO(OH)_3(aq)$ for equation (5.2.8) can be calculated from equation (5.2.7) and (5.2.4) as logK=-3.6.

$$Pa^{4+} + 4H_2O \leftrightarrow PaO(OH)_3(aq) + 5H^+ + e^- \qquad\qquad (5.2.8)$$

The equilibrium constant of hydrolysis reaction of Pa(V) are compiled in Table 5.2.2. In this table, the formation constants of $PaOOH^{2+}$ from Fuger and Oetting (1976), $PaO(OH)_2^+$ and $PaO(OH)_3(aq)$ from Fuger (1992) are selected for the refined PNC-TDB, and those of $PaO_2(OH)(aq)$, $Pa(OH)_4^+$ and $Pa(OH)_5(aq)$ are not considered because the reliability of data was unknown.

Table 5.2.2 The formation constant for Pa(V) hydoxo-species from Pa^{4+}

	PaO_2OH	$Pa(OH)_4^+$	$Pa(OH)_5$	$PaOOH^{2+}$	$PaO(OH)_2^+$	$PaO(OH)_3(aq)$
[1]					0.85	-3.6
[2]				1.9		
[3]	3.29			7.29		
[4]		11.9	7.01	1.92		
[5]	-2.14			1.86		

[1]Fuger,1992, [2]Fuger and Oetting,1976, [3]Cross et al.,1987,
[4]CHEMVAL database, [5]HATCHES ver.10

6. CHLORIDE COMPLEX

Thermodynamic data of $PaCl_4(c)$ and $PaCl_5(c)$ were compiled in NBS table (Wagmann et al.,1982). In this report, the free energy of formation of $PaCl_4(c)$ and $PaCl_5(c)$ were -953kJ/mol and -1034.2 kJ/mol, respectively. The solubility product of $PaCl_4(c)$ and $PaCl_5(c)$ for equation (5.2.7) and (5.2.8) is calculated from the free energy of formation as logK=24.01 and 32.85, respectively.

$$PaCl_4(c) \leftrightarrow Pa^{4+} + 4Cl^-$$ (5.2.7)

$$PaCl_5(c) \leftrightarrow Pa^{5+} + 5Cl^- + e^-$$ (5.2.8)

The solubility product for equation (5.2.7) and (5.2.8) calculated from the thermodynamic data of $PaCl_4(c)$ and $PaCl_5(c)$ in NBS table (Wagmann et al., 1982) are selected for the refined PNC-TDB. The solubility product of $PaCl_4(c)$ are compiled in Table 6.1.

Table 6.1 The solubility product of $PaCl_4(c)$

References	logK
NBS table	24.01
Cross et al.,1987	24.10

7. THERMODYNAMIC DATABASE OF Pa FOR PNC-TDB

Thermodynamic database of Pa selected for the refined PNC-TDB are shown in Table 7.1 and 7.2.

Table 7.1 Thermodynamic database of Pa solid for the refined PNC-TDB

Solid	Reaction	logK	original data from
$PaO_2(c)$	$PaO_2(c)+4H^+ \leftrightarrow Pa^{4+}+2H_2O$	0.6	Baes and Mesmer,1976
$Pa_2O_5(s)$	$Pa_2O_5+10H^++2e^- \leftrightarrow 2Pa^{4+}+5H_2O$	-8.72	Fuger and Oetting,1976 +Baes and Mesmer,1976
$PaCl_4(c)$	$PaCl_4(c) \leftrightarrow Pa^{4+}+4Cl^-$	24.01	Fuger and Oetting,1976 +Wagman et al.,1982
$PaCl_5(c)$	$PaCl_5(c) \leftrightarrow Pa^{4+}+5Cl^-+e^-$	32.85	Fuger and Oetting,1976 +Wagman et al.,1982

Table 7.2 Thermodynamic database of Pa aqueous species for the refined PNC-TDB

Species	Reaction	logK	original data from
$PaOH^{3+}$	$Pa^{4+}+H_2O \leftrightarrow PaOH^{3+}+H^+$	0.84	Baes and Mesmer,1976
$Pa(OH)_2^{2+}$	$Pa^{4+}+2H_2O \leftrightarrow Pa(OH)_2^{2+}+2H^+$	-0.02	Baes and Mesmer,1976
$Pa(OH)_3^+$	$Pa^{4+}+3H_2O \leftrightarrow Pa(OH)_3^++3H^+$	-1.5	Baes and Mesmer,1976
$PaOOH^{2+}$	$Pa^{4+}+2H_2O \leftrightarrow PaOOH^{2+}+3H^++e^-$	1.86	Fuger and Oetting,1976
$PaO(OH)_2^+$	$Pa^{4+}+3H_2O \leftrightarrow PaO(OH)_2^++4H^++e^-$	0.85	Fuger and Oetting,1976 +Fuger,1992
$PaO(OH)_3(aq)$	$Pa^{4+}+4H_2O \leftrightarrow PaO(OH)_3(aq)+5H^++e^-$	-3.6	Fuger and Oetting,1976 +Fuger,1992

8. SOLUBILITY CALCULATION

The solubility was calculated using existing experimental data in order to confirm the reliability of the refined PNC-TDB. The PHREEQE code (Parkurst et al., 1980) has been used to calculate protactinium solubilities.

8.1 Solubility experiment

Solubility of protactinium was studied by Berry et al. (1989). In their study, [233]Pa and [231]Pa were used. The solubility was measured from over saturation in 0.1M NaCl. Solubility limiting solid was not identified. The solubility experiment was carried out under N_2 atmosphere at the room temperature. Experimental period was 2 hours, pH was adjusted by NaOH and HCl. The solid liquid separation was conducted by using 25000MWCO filter. Dominant oxidation state was suggested Pa(V).

8.2 Solubility calculation

Solubility was calculated by using the refined PNC-TDB described in section 7. In this calculation, solubility limiting solid and ionic media were assumed $Pa_2O_5(s)$ and 0.1M NaCl, respectively. The effect of O_2 and CO_2 gas was not considered. The temperature was assumed 25°C.

8.3 Result

Calculated results are shown in Fig.8.1. In the results, solubility of $Pa_2O_5(s)$ was predicted 3.15×10^{-7}M. Experimental value didn't agree with calculated value. The reliability of the revised PNC-TDB was not able to be confirmed as shown in Fig.8.1.
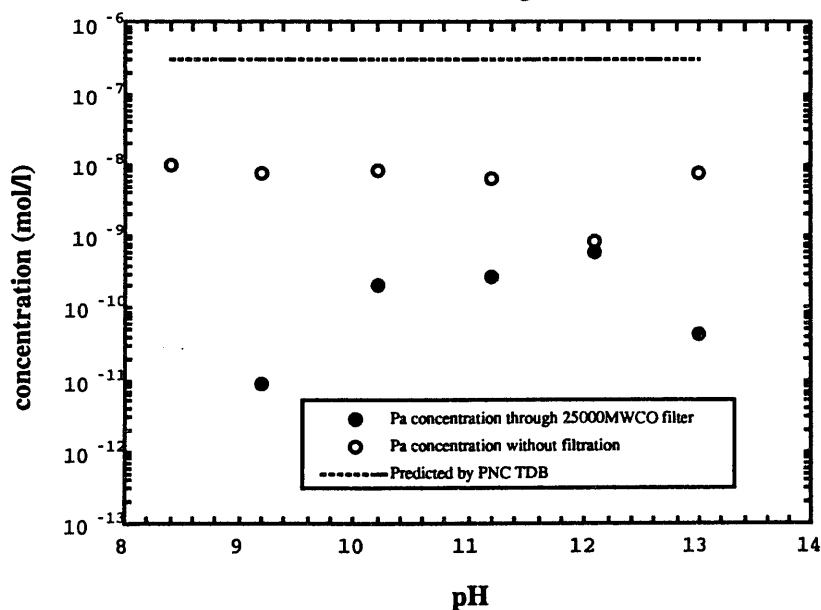

Fig. 8.1 Experimental and calculated results on solubility of Pa in cement equilibrated water

9. DISCUSSION

There were various values of Pa thermodynamic data. However, the available data for performance assessment were too rare, because most of the data were obtained under unappropriate experimental conditions such as very low pH, high ionic strength, moreover identification of solid phase and aqueous species was not carried out. The disagreement between experimental and calculated values might be unequilibrium of the system because experimental period was too short (i.e. the two hours test period). Although the calculated solubility was higher than experimental value, the former value can apply as the conservative solubility estimation for performance assessment of geological disposal system of high level radioactive waste. Therefore, the refined Pa dataset was considered to be available for performance assessment.

10. CONCLUSION

In this report, the thermodynamic data of redox reaction and hydrolysis of Pa was discussed, and the data in the previous PNC-TDB was refined. There were little reliable data for redox reaction and hydrolysis of Pa. The Pa database in the refined PNC-TDB is available for performance assessment, by the comparison between experimental and calculated values.

REFERENCES

Baes,C.F. and Mesmer,R.E. (1976); The Hydrolysis of Cations. John Wiley & Sons, New York

Berry,J.A., Hobley,J., Lane,S.A., Littleboy,L.A., Nash,M.J., Oliver,P., Smith-Briggs,J.L. and Williams,S.J. (1989); Solubility and Sorption of Protactinium in the Near-field and Far-field Environments of a Radioactive Waste Repository. ANALYST, Vol.114, pp.339-347

Bratsch,S.G. and Lagowski,J.J. (1986); Actinide Thermodynamic Predictions. 3. Thermodynamics of Compounds and Aquo Ions of the 2+ 3+, and 4+ Oxidation States and Standard Electrode Potentials at 298.15K. J. Phys. Chem., Vol.90, pp.307-312

Brown,D., Gantner,E., Kanellakopulos,B. Munzel,H. and Seidel,A. (1977); Protactinium, in Gmelin Handbuch der Anorganishen Chemie

Broun,P.L. and Wanner,H. (1987); Predicted Formation Constants Using the Unified Theory of Metal Ion Complexation. OECD/NEA

Cross,J.E., Ewart,F.T. and Tweed,C.J. (1987); Thermochemical Modelling with Application to

Nuclear Waste Processing and Disposal, AERE-R12324

Falck,W.E., (1992); CHEMVAL Project; Critical Evaluation of the CHEMVAL Thermodynamic Database with Respect to its Contents and Relevance to Radioactive Waste Disposal at Sellafield and Dounreay -Thermodynamic Database and Geochemical Modeling(ver.3.0).

Fried,S. and Hindman,J.C. (1964); J. Am. Chem. Soc., 76, 4863-4867

Fuger,J. (1992); Thermodynamic Properties of Actinide Aqueous Species Relevant to Geochemical Problems. Radiochimica Acta, 58/59, pp.81-91

Fuger,J. and Oetting,F.L. (1976); The Chemical Thermodynamics of Actinide Elements and Compounds Part2, The Actinide Aqueous Ions. IAEA, Vienna

Guillaumont,R., Bouissieres,G. and Muxart,R. (1968); Chimie du Protactinium. I. Solutions Aqueuses de Protactinium Penta et Tetravalent. Actinides Rev. Vol.1, pp.135-163

Harwell/Nirex (1997); HATCHES Ver.10, The Harwell/Nirex Thermodynamic Database Release10, Available from the Nuclear Energy Agency Data Bank

Inova,G.V., Pershina,V., Johnson,E.. and Fricke,C. (1992); in: Abstracts of Papers of the 22iemes Journees des Actinides, Meribel Savoire, France, p.137

Kirby,H.W. (1986); Protactinium, in Katz,J.J., Seaborg,G.T. and Morss,L.R., The Chemistry of Actinide Elements, 2nd Ed., Vol.2, Chapman and Hall

Lundqvist,R. (1974); Acta. Chem. Scand. A28, 243

Mikheev,N.B., Kamenskaya,A.N., Rumer,I.A., Kulyukhin,S.A. and Auerman,L.N. (1994); Reduction of Protactinium to the Bivalent State in the Melt and Determination of the Pa^{3+}/Pa^{2+} Oxidation Potential. Translated from Radiokhimiya, pp.512-514

Nugent,L.J., Baybarz,R.D. and Burnett,J.L. (1973); Electron-Transfer and f-d Absorption Bands of Some Lanthanide and Actinide Complexes and the Standard (II-III) Oxidation Potential for Each Member of the Lanthanide and Actinide Series. J. Phys. Chem. Vol.77, pp. 1528-1535

Parkurst,D.L., Thorstenson,D.C. and Plummer,L.N. (1980); PHREEQE-A Computer Program for Geochemical Calculations. U.S. Geological Survey, Water-Resources Investigations 80-96

Yui,M., Takeda,S., Komuro,K., Makino,H., Shibutani,T., Umeki,H., Ishiguro,K., Takase, H. and Neyama,A. (1992); "Kou-reberu haikibutu chisou-syobun shisutemu no seinou-hyouka ni okeru chikasui no chikyu-kagaku-teki tokusei ni kansuru moderuka" (Modelling of groundwater evolution for performance assessment of high-level radioactive waste disposal). PNC TN8450 92-166 (in Japanese)

Wagman,D.D., Evans,W.H., Parker,V.B., Schumm,R.H., Halow,I., Bailey,S.M., Churney, K.L. and Nuttall,R.L. (1982); The NBS Tables of Chemical Thermodynamic Properties. Selected Values for Inorganic and C1 and C2 Organic Substances in SI Units. Journal of Physical and Chemical Reference Data, Vol.11