

Solubilities of Actinides in a Domestic Groundwater

S. S. Kim, K. C. Kang, M. H. Baik, J. W. Choi
 HLW Disposal Research Center, Korea Atomic Energy Research Institute
 1045 Daedeok-daero, Yusong-gu, Daejeon, 305-353, nsskim@kaeri.re.kr

1. Introduction

Solubility limits and speciation data of nuclides in a groundwater are important for understanding an aqueous radionuclide transport through a geosphere in a high level waste repository. They define the source term for transport retardation processes such as a sorption and colloid formation, and are useful in verifying the validity of geochemical codes that are part of the predictive transport models [1].

In this study, the solubilities of actinides with a long lifetime in a domestic granitic ground water were calculated by PHREEQC (V.2), and the results are compared with the values reported in other literatures.

2. Calculation Method

For the calculation of the solubility of a nuclide with a geochemical program, the thermodynamic data and a solubility limiting solid phase (SLSP) of the nuclide, the composition and temperature of a solution, the activities of the nuclide aqueous species in equilibrium and etc. are required.

2.1 Geochemical Program

PHREEQC (V.2) is designed to perform a wide variety of low-temperature aqueous geochemical calculations. This program is based on an ion-association aqueous model and has capabilities for a speciation and saturation-index calculation, batch reaction and one-dimensional transport calculation [2]. This is one of the well known geochemical programs with EQ3/6 for the calculation of a solubility and migration of actinides in a nuclear waste repository.

2.2 Thermodynamic Data

As the thermodynamic input data for the reactions of actinides, the data in the fifth volume of the series "Chemical Thermodynamics" edited by the OECD Nuclear Energy Agency (NEA) in 2003 were mainly selected. These data are update with the critical reviews of chemical thermodynamics about actinides published successively in 1992, 1995, 1999 and 2001 [3]. The reaction constant values in the condition of 296.15 K, 0.1 MPa of a pressure and "0" of an ionic strength were used for the calculation of an actinide's solubility without any correction.

2.3 Solubility Limiting Solid Phase (SLSP)

While SLSP is a pure radionuclide bearing solid or a solid solution having more than two end-members, the former is generally used for the calculation of a solubility. When a solid solution is used, the modeling is too complicated to calculate its solubility and its formation in the solution has to be verified. Moreover, since the solubilities of pure radionuclide bearing solids are greater than those of solid solutions, the use of a pure radionuclide bearing solid can produce a conservative solubility of a radionuclide [4].

For the selection of SLSP, its kinetics as well as thermodynamics in a solution has to be considered. An amorphous precipitate formed initially in a solution turns into a crystal form slowly with a lower solubility. Therefore, if the amorphous form of a nuclide is used as a SLSP, its solubility would be a conservative value [4]. In this study, the solubilities of actinides for both amorphous and crystal forms known as SLSP in a granitic groundwater were calculated [5-6].

2.4 Composition of a Groundwater

A natural groundwater sampled from the YS-01 borehole, located inside KAERI at Yuseong, Korea, at a 457.5 m depth from the surface by a multi-packer system was analyzed for its composition. This composition is shown in Table 1. The gravity of the groundwater was assumed as 1.0.

Table 1. The geochemical conditions of the groundwater sampled from the borehole YS-01

Element	Na	K	Ca	Mg	HCO ₃
Concentration (mg/L)	35.7	20.33	2.7	0.05	88
Element	F	Fe	Mn	Li	SiO ₂
Concentration (mg/L)	12.7	0.016	8x10 ⁻⁴	0.12	18.5
Element	Cl	Sr	SO ₄	NO ₃	Al
Concentration (mg/L)	4.2	0.059	2.3	0.01	0.079
Eh (mV) ^{a)}	pH				
-194	9.92				

3. Results and Discussion

The solubilities of the actinides were calculated with PHREEQC (V.2) by inputting thermodynamic reaction data, a groundwater composition and SLSP at 296.15 K and the results are shown in Table 2. When these values

are compared with other results in a similar groundwater condition, the difference between them is not great except for uraninite.

Table 2 Comparison of the solubilities of actinides between this and other studies

SLSP ^a	PFREEQ C ^{ab}	MUGRE M ^c	EQ3/6 ^d
	(mole/L)		
Am(OH) ₃ (am)	1.5 x 10 ⁻⁸	-	-
Am(OH) ₃ (c)	7.7 x 10 ⁻¹⁰	3.5 x 10 ⁻¹⁰	-
AmCO ₃ OH(am)	1.2 x 10 ⁻⁷	-	-
Np(OH) ₄ (am)	3.4 x 10 ⁻⁹	4.4x10 ⁻⁹	-
NpO ₂ (c)	3.4 x 10 ⁻¹⁶	-	5 x 10 ⁻¹⁸
Pu(OH) ₄ (am)	6.3x10 ⁻¹¹	8.6x10 ⁻¹¹	-
PuO ₂ (c)	9.4x10 ⁻¹¹	-	-
PuO ₂ (am)	6.3x10 ⁻¹¹	-	-
UO ₂ (am)	7.5 x 10 ⁻⁵	1.7 x 10 ⁻⁴	-
Uraninite	2.0 x 10 ⁻⁷	-	4.0 x 10 ⁻¹⁰

^aSolubility Limiting Solid Phase
^bThis study results
^cReference 5, p.88, pH=9.9. Eh=-194mV
^dReference 6, pH=10.3. Eh=-435mV

4. Conclusion

The solubilities of actinides with a long lifetime in a domestic granitic ground water condition were calculated by PHREEQC (V.2), one of the well known geochemical codes. From the calculation, the maximum solubilities of Am, Np, Pu and U in the ground water were obtained as 1.2 x 10⁻⁷, 3.4 x 10⁻⁹, 9.4x10⁻¹¹ and 7.5 x 10⁻⁵, respectively. These values are similar to the results calculated by other geochemical codes, MUGREM and EQ3/6.

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