

Sorption of Np(IV) on MX-80 in Ca-Na-Cl Type Reference Water of Crystalline Rock

Shinya Nagasaki*

McMaster University, 1280 Main Street West, Hamilton, Ontario, L8S 4L7, Canada

(Received October 24, 2020 / Revised November 3, 2020 / Approved November 18, 2020)

The pH dependence of sorption distribution coefficient (K_d) of Np(IV) on MX-80 in Ca-Na-Cl type solution with the ionic strength of 0.3 M, which was similar to one of the reference groundwaters in crystalline rock, was experimentally investigated under the reducing conditions. The overall trend of K_d on MX-80 was independent of pH at $5 \leq \text{pH} \leq 10$ but increased as pH increased at $\text{pH} \leq 5$. The 2-site protolysis non-electrostatic surface complexation and cation exchange model was applied to the experimentally measured pH dependence of K_d and the optimized surface complexation constants of Np(IV) sorption on MX-80 were estimated. The values of surface complexation constants in this work agreed relatively well with those in the Na-Ca-Cl solution previously evaluated, suggesting that compared to Na^+ , the competition of Ca^{2+} with Np(IV) for surface complexation on MX-80 was not much strong in Ca-Na-Cl solution. The sorption model well predicted the pH dependence of K_d values but slightly overestimated the sorption at the low pH region.

Keywords: Np(IV), MX-80, Sorption, pH dependence, Sorption distribution coefficient, 2 SPNE SC/CE model

*Corresponding Author.

Shinya Nagasaki, McMaster University, E-mail: nagasas@mcmaster.ca, Tel: +1-905-525-9140

ORCID

Shinya Nagasaki

<http://orcid.org/0000-0002-6990-0811>

1. Introduction

The Nuclear Waste Management Organization (NWMO) has a responsibility to implement the Adaptive Phased Management (APM) which was approved by the Canadian federal government for a long-term management plan of used nuclear fuel in Canada [1]. Under the APM plan, used nuclear fuel will be placed within a deep geological repository (DGR) in a suitable rock formation. Crystalline rocks in Canada are being as potential host rocks for a DGR [1].

The NWMO proposes a reference groundwater composition to represent the saline groundwater conditions at the depth of the hypothetical repository (CR-10) [1]. CR-10 is Ca-Na-Cl type water with an ionic strength (I) of about 0.26 M and is under reducing conditions. It was defined by groundwater chemistry at this depth in the crystalline rock under reducing conditions in Canada.

Sorption of radionuclides, including actinides and their daughter products onto host rocks surrounding a DGR and onto materials comprising the engineered barrier system is considered an important mechanism for retarding their subsurface transport from the repository to the biosphere [2-6]. Therefore, it is important to elucidate the sorption behavior of radionuclides not only on host rocks but also on engineered barrier materials such as MX-80 bentonite which is one of the candidate buffer materials in the DGR project.

^{237}Np is one of the key elements to dominate a long-term radiological risk of used nuclear fuel and high-level radioactive waste disposal. Np exists as Np(IV) under reducing conditions. For Np(IV) sorption on bentonite and other clay minerals, some investigations have been carried out [7-14]. However, only three papers studied the Np(IV) sorption at $I > 0.1$ M. Kitamura and Tomura [11] and Ashida et al. [12] studied the effect of carbonate ions on sorption of Np(IV) on smectite at $I = 1.0$ M. Nagasaki et al. [14] studied the sorption of Np(IV) on illite, shale and MX-80 in Na-Ca-Cl type solution with $I = 0.1$ M to 6 M, and showed that K_d (sorption distribution coefficient [15]) values for illite,

shale and MX-80 were independent of the ionic strength at $I \geq 1$ M, but dependent on it at $0.1 \text{ M} \leq I \leq 1 \text{ M}$. However, there is no K_d data of Np(IV) on bentonite like MX-80 in Ca-Na-Cl type solution with $I > 0.1$ M such as CR-10. Both Ca-Na-Cl and Na-Ca-Cl type solutions contain Ca, Na and Cl, but in the former, Ca is richer than Na and, in the latter, vice versa. Although the ionic strength is the same as each other, the sorption competition between Np(IV) and Ca^{2+} may be enhanced in Ca-Na-Cl type solution.

Sorption modelling improves the understanding of sorption processes. The 2-site protolysis non-electrostatic surface complexation and cation exchange model (2 SPNE SC/CE model) has been successfully applied to simulate the sorption of tetravalent actinides such as Th(IV), Np(IV), and Pu(IV) on illite and montmorillonite [16, 17].

In this study, the sorption of Np(IV) on MX-80 in Ca-Na-Cl type solution with $I = 0.3$ M was studied as a function of pH, and the 2 SPNE SC/CE model was applied and the surface complexation constants were estimated and compared to the constants obtained from the Np(IV) sorption in Na-Ca-Cl type solution on MX-80.

2. Experimental

2.1 Materials

All chemicals used were reagent grade and supplied from Fisher Scientific. Deionized water from a Milli-Q Direct 8 was used after being contacted with 10% H_2 + 90% N_2 gas in the presence of iron (electrolytic powder) for three days. After the iron powder was removed by centrifugation, 1×10^{-2} M $\text{Na}_2\text{S}_2\text{O}_4$ was added to the deionized water, and that was then stored in a glove box (GB). Eh value of the water was confirmed to be stable in the range of -250 mV to -170 mV (vs. SHE) for 30 days. Just before use, the water was treated with 10% H_2 + 90% N_2 gas again for several hours without iron powder addition. The concentration of Fe in the solution was smaller than the detection limit of

ICP-MS (Agilent ICP-MS 8800). Hence, it was considered that the iron dissolved in the water did not affect the sorption behaviour of Np(IV).

The solid used was MX-80. The specific surface area was $26.2 \text{ m}^2 \cdot \text{Rg}^{-1}$ [13]. Other information including their mineralogy is described elsewhere [2, 13, 15]. A Precise Controlled Atmosphere GB supplied by Labconco was filled with N_2 gas (> 99.999 %) to exclude CO_2 , and the N_2 gas was left running through the GB. The concentration of O_2 in the running N_2 gas was confirmed to be less than 2 ppm by an oxygen sensor.

The ^{237}Np solution was purchased from Stuart Hunt & Associates Ltd. Np(IV) solution was prepared using the established procedures [18-21]. The tetravalent oxidation state of the prepared Np solution was confirmed by UV-VIS spectroscopy (Agilent 8453 UV-Vis spectrometer) and 0.1 M TTA-xylene extraction. The oxidation of Np(IV) to Np(V) was also confirmed to be negligibly small at least for 30 days after the Np(IV) stock solution was prepared. Np(IV) stock solution was prepared for use just before sorption experiments. The concentration of the Np(IV) stock solution was $1.0 \times 10^{-7} \text{ M}$ in 0.01 M HClO_4 solution.

2.2 Sorption experiment

The CR-10 reference groundwater contains $1,900 \text{ mg} \cdot \text{L}^{-1}$ of Na and $2,130 \text{ mg} \cdot \text{L}^{-1}$ of Ca (Na/Ca molar ratio = 0.64) [1]. Therefore, in this study, Ca-Na-Cl solutions with Na/Ca molar ratio = 0.64 and $I = 0.3 \text{ M}$ was prepared by using NaCl and $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$.

When I is higher than 0.1 M, the molar H^+ concentrations ($\text{pH}_c = -\log c_{\text{H}^+}$) or the molal H^+ concentration ($\text{pH}_m = -\log m_{\text{H}^+}$) should be used instead of the pH values indicated on the pH meter (pH_{meter}). However, since the difference between pH_c and pH_{meter} was confirmed to be small at $I = 0.3 \text{ M}$, the pH values indicated on the pH meter were used in this study.

All sorption experiments were carried out at 25°C in triplicate. The basic procedures of sorption experiments

were the same as those we used in the sorption experiments of Np(IV) in Na-Ca-Cl solutions [14]. In this study, the sorption results were expressed as K_d [$\text{m}^3 \cdot \text{kg}^{-1}$], which is calculated as follows:

$$K_d = \frac{(C_0 - C_e) V}{C_e W} \quad (1)$$

where C_0 (M) is the initial concentration of Np(IV) in the liquid phase, C_e [M] the equilibrium concentration of Np(IV) in the liquid phase, V [m^3] the volume of the liquid phase, and W [kg] the mass of the solid phase.

In this study, assuming that the time required for sorption equilibrium was the same as that in the Na-Ca-Cl solution case (3–7 days) [14], the sorption test period was set 14 days.

The pH dependence of the K_d value of Np(IV) sorption for MX-80 in Ca-Na-Cl solutions ($I = 0.3 \text{ M}$) was measured. During the solid/liquid (MX-80/Ca-Na-Cl solutions ($I = 0.3 \text{ M}$)) pre-equilibration period of 3 to 4 days, 10% H_2 + 90% N_2 gas passed through the solid/liquid mixture for 30 minutes once a day. The solid/liquid mixture was then removed from the GB, and the liquid and solid were separated by centrifugation for 30 minutes at 12,000 rpm. pH and Eh of the solution were measured in the GB to confirm the low Eh values (smaller than -100 mV (vs. SHE)).

Ca-Na-Cl solution ($I = 0.3 \text{ M}$) was added again to the pre-equilibrated solid in the polycarbonate reaction vessel in the GB. A portion of the Np(IV) stock solution was spiked into the reaction vessel in the GB. The pH of the solution was adjusted to the pre-decided values between about 3 and about 10 by the addition of 0.01 M HCl, 0.1 M HCl, 0.1 M NaOH, or 0.01 M NaOH solution in the GB. The reaction vessels were tightly sealed in the GB, transferred from the GB to an incubator at 25°C (Infors HT Electron), and were gently shaken for a pre-decided period of time. The pH of the solution was measured once a day in the GB and re-adjusted to the original pH value if the pH changed by more than ± 0.3 from the original value. Then, the reaction vessels were returned to the incubator. The ini-

Table 1. Surface complexation constants for MX-80 estimated in this work and estimated in Na-Ca-Cl solution at $I = 0.1$ to 4 M [14]

Surface species	$\log {}^sK$ ($I = 0$ M) on strong site in Na-Ca-Cl solution (this work)	$\log {}^sK$ ($I = 0$ M) on strong site in Na-Ca-Cl solution [14]
$\equiv \text{S}^{\circ}\text{ONp}^{3+}$	nd	nd
$\equiv \text{S}^{\circ}\text{ONpOH}^{2+}$	12 ± 0.3	10 ± 0.3
$\equiv \text{S}^{\circ}\text{ONp}(\text{OH})_2^{+}$	5.8 ± 0.3	5.7 ± 0.2
$\equiv \text{S}^{\circ}\text{ONp}(\text{OH})_3^0$	0.15 ± 0.2	0.10 ± 0.1
$\equiv \text{S}^{\circ}\text{ONp}(\text{OH})_4^{-}$	-7.6 ± 0.4	-7.8 ± 0.3

*“nd” refers to value that could not be determined.

tial Np(IV) concentrations were 1.0×10^{-11} M. The solid/liquid ratio was $0.2 \text{ g}\cdot\text{L}^{-1}$ ($2 \times 10^{-4} \text{ kg}\cdot\text{m}^{-3}$). The liquid was separated from the solid by centrifugation (Nalgene Oak Ridge tubes for 120 minutes at 18,000 rpm at 25°C). The concentration of Np in the liquid was measured by ICP-MS.

3. Results and discussion

The pH dependence of K_d value of Np(IV) sorption in Ca-Na-Cl solution is illustrated in Fig. 1. The details of Np(IV) concentration in liquid at sorption equilibrium and the K_d values are summarized in Supplementary Material. Fig. 1 shows that the K_d values of Np(IV) for MX-80 increased with pH from 3 to 5 and were not dependent on pH between 5 and 10. This trend was identical to that measured in Na-Ca-Cl solution at $I = 0.1$ M to 6 M. The absolute values of K_d in Ca-Na-Cl were also close those in Na-Ca-Cl solution. These suggest that the trend of Np(IV) sorption behavior on MX-80 toward pH is similar both in Ca-Na-Cl solution ($I = 0.3$ M) and in Na-Ca-Cl solution ($0.1 \text{ M} \leq I \leq 6 \text{ M}$).

The 2 SPNE SC/CE model was applied to simulate the pH dependence of K_d values of Np(IV) on MX-80. The specific ion interaction theory (SIT [22]) was used to calculate the activity coefficients of aqueous species. Thermodynamic constants for Np(IV) aqueous species were taken

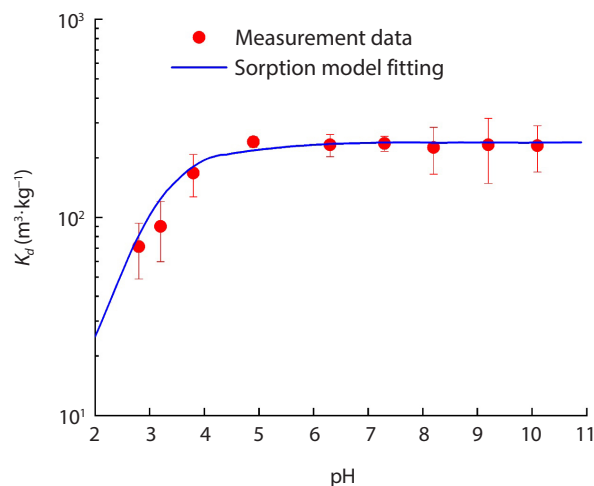


Fig. 1. pH dependence of the K_d values in Ca-Na-Cl solution ($I = 0.3$ M). Solid line is the simulation results using 2 SPNE SC/CE model.

from the NEA thermodynamic database [23]. When the SIT parameters required for the calculation were not reported in the NEA thermodynamic database, the parameters in the SIT database provided with PHREEQC [24] were used. The surface complexation constants estimated for Np(IV) on MX-80 in Na-Ca-Cl solution were used as initial values for fitting. Other parameters for fitting such as protolysis constants were cited from the values used by Bradbury and Baeyens [16, 17] because Np(IV) sorption on MX-80 in Na-Ca-Cl solution was successfully simulated by using those values [14].

The fitting result was also shown in Fig. 1. It was found

Appendix 1. Np(IV) concentrations in liquid phase at sorption equilibrium and K_d values

pH	2.8	3.2	3.8	4.9	6.3	7.3	8.2	9.2	10.1
$C_e(1)$ $\times 10^{-12}$ M	9.32	9.88	9.75	9.55	9.61	9.60	9.65	9.67	9.67
$C_e(2)$ $\times 10^{-12}$ M	9.87	9.80	9.66	9.51	9.54	9.56	9.62	9.60	9.54
$C_e(3)$ $\times 10^{-12}$ M	9.89	9.78	9.61	9.51	9.53	9.53	9.47	9.42	9.47
K_d ($\text{m}^3 \cdot \text{kg}^{-1}$)	71 ± 21	92 ± 31	1.7×10^2 ± 41	2.5×10^2 ± 15	2.3×10^2 ± 27	2.3×10^2 ± 20	2.2×10^2 ± 60	2.3×10^2 ± 79	2.3×10^2 ± 60

that the fitting result was quantitatively consistent with the experimental K_d values at $\text{pH} \geq 5$, but slightly overestimated the experimental data at $\text{pH} \leq 4$. The surface complexation constants obtained are summarized in Table 1 together with those in Na-Ca-Cl solution with $I = 0.1$ to 4 M [14]. The values of surface complexation constants in this work agreed relatively well with those in the Na-Ca-Cl solution. This may suggest that compared to Na^+ , the competition of Ca^{2+} with Np(IV) for surface complexation on MX-80 was not much strong in the Ca-Na-Cl solution.

In this estimation, we neglected the surface complexation reactions on weak sites, because the concentration of Np(IV) was very small and the reactions “ $\equiv \text{S}^{\circ}\text{OH} + \text{Np}^{4+} \leftrightarrow \equiv \text{S}^{\circ}\text{ONp}^{3+} + \text{H}^+$ ” and “ $\equiv \text{S}^{\text{w}1}\text{OH} + \text{Np}^{4+} \leftrightarrow \equiv \text{S}^{\text{w}1}\text{ONp}^{3+} + \text{H}^+$ ” did not contribute to the simulation practically. The sorption capacity of the strong site was set $2.0 \times 10^{-3} \text{ mol} \cdot \text{kg}^{-1}$ for Na-montmorillonite [16]. This meant $4.0 \times 10^{-7} \text{ mol}$ on 0.2 g MX-80. Compared to the initial Np(IV) concentration ($1 \times 10^{-11} \text{ M}$), it was much large. In such a situation, the contribution of weak sites frequently could be negligible [16, 17]. The result in this study is consistent with the previous works [16, 17].

In the 2 SPNE SC/CE model, cation exchange reaction as well as surface complexation reaction is considered. From the simulation, it was found that the fitting result agreed well with the experimental results only by the surface complexation. However, as we pointed out in our previous studies [14, 25], a non-electrostatic model like

the 2 SPNE SC/CE model may not be sufficient when electrostatic effects could play an important role in the cation exchange. This might be related to the overestimation of fitting at $\text{pH} \leq 4$. This will be left for the challenge to be solved in the future.

The surface complexation constants for surface species “ $\equiv \text{S}^{\circ}\text{ONp}(\text{OH})_2^{+}$ ” and “ $\equiv \text{S}^{\circ}\text{ONp}(\text{OH})_3^0$ ” were well consistent with those estimated by the linear free energy relationship (LFER) [16, 17]. On the other hand, the surface complexation constant for surface species “ $\equiv \text{S}^{\circ}\text{ONpOH}_2^{+}$ ” estimated in this work was larger than that by LFER. These consistency and discrepancy were also observed in the Na-Ca-Cl study [14]. Hence, this might suggest a necessity of review of hydrolysis constants of Np(IV). However, in this study, we used the parameter values such as protolysis constants which Bradbury and Baeyens reported. So, we need more investigations.

4. Conclusions

The K_d values of Np(IV) on MX-80 in Ca-Na-Cl solution with $I = 0.3 \text{ M}$ were measured as a function of pH. The overall trend of pH dependence of Np(IV) on MX-80 was that the K_d values increased with pH at $\text{pH} \leq 5$ and were independent of pH at $5 \leq \text{pH} \leq 10$. The trend and the K_d values were almost the same as those observed in the Na-Ca-Cl solution with $I = 0.1$ to 6 M.

The pH dependence of K_d values was simulated by the 2 SPNE SC/CE model well. The surface complexation constants for Np(IV) sorption on MX-80 were estimated by fitting the 2 SPNE SC/CE.

Acknowledgement

This study was funded by the Discovery Grant Program of Natural Science and Engineering Research Council of Canada (RGPIN-2014-05732). An author wishes to acknowledge Mr. Justin Riddoch who was a graduate student at McMaster University and now works at Ontario Power Generation for his technical support.

Appendix

In this study, the triplicate measurements were conducted at each pH. K_d value was determined by average of three measurements. Error of K_d was the maximum deviation between measurement values and average value. Initial concentration of Np(IV) was 1.0×10^{-11} M.

REFERENCES

- [1] Nuclear Waste Management Organization. Postclosure Safety Assessment of a Used Fuel Repository in Crystalline Rock, Nuclear Waste Management Organization Technical Report, 1-46, NWMO TR-2017-02 (2017).
- [2] P. Vilks. Sorption of Selected Radionuclides on Sedimentary Rocks in Saline Conditions—Literature Review, Nuclear Waste Management Organization Technical Report, 1-14, NWMO TR-2011-12 (2011).
- [3] H. Geckeis, J. Lützenkirchen, R. Polly, T. Rabung, and M. Schmidt, “Mineral-Water Interface Reactions of Actinides”, *Chem. Rev.*, 113(2), 1016-1062 (2013).
- [4] M. Zavarin, B.A. Powell, M. Bourbin, P.H. Zhao, and A.B. Kersting, “Np(V) and Pu(V) Ion Exchange and Surface-Mediated Reduction Mechanisms on Montmorillonite”, *Environ. Sci. Technol.*, 46, 2692-2698 (2012).
- [5] N. Chapman, M. Apte, W. Aspinall, K. Berryman, M. Cloos, C. Connor, L. Connor, O. Jaquet, K. Kiyosugi, E. Scourse, S. Sparks, M. Stirling, L. Wallace, and J. Goto. TOPAZ Project: Long-term Tectonic Hazard to Geological Repositories, Nuclear Waste Management Organization of Japan Technical Report, 1-11, NUMO-TR-12-05 (2012).
- [6] National Academy of Science, “A Study of the Isolation for Geologic Disposal of Radioactive Wastes”, *Waste Isolation Systems Panels*, eds., 1st ed., National Academy Press, Washington D.C. (1983).
- [7] S. Tanaka, S. Nagasaki, A. Suzuki, T. Yamaguchi, S. Tsushima, K. Yamaguchi, and Y. Moriyama. The Underground Environment Migration Behaviour of TRU Elements (3), The University of Tokyo Report, 64-65, UTNL-371 (1998).
- [8] G.M.N. Baston, J.A. Berry, M. Brownsword, T.G. Heath, D.J. Ilett, C.J. Tweed, and M. Yui, “The Effect of Temperature on the Sorption of Technetium, Uranium, Neptunium and Curium on Bentonite, Tuff and Grandionite”, *Proc. Mat. Res. Soc.*, 465, 805-812 (1997).
- [9] S. Nagasaki, S. Tanaka, and A. Suzuki, “Sorption of Neptunium on Bentonite and its Migration in Geosphere”, *Colloids Surf. A Physicochem. Eng. Asp.*, 155(2-3), 137-143 (1999).
- [10] N.L. Banik, R. Marsac, J. Lützenkirchen, C.M. Marquardt, K. Dardenne, D. Schild, J. Rothe, A. Diascorn, T. Kupcik, T. Schäfer, and H. Geckeis, “Neptunium Redox Speciation at the Illite Surface”, *Geochim. Cosmochim. Acta*, 152, 39-51 (2015).
- [11] A. Kitamura and T. Tomura, and T. Shibutani. Sorption Behaviour of Neptunium onto Smectite Under Reducing Conditions in Carbonate Media, The Japan Nuclear Cycle Development Institute Report, 659-666, JAERI-CONF-2002-004 (2002).
- [12] T. Ashida, T. Shibutani, H. Sato, Y. Tachi, A. Kitamura

- and K. Kawamura. Nuclide Migration Study in the QUALITY. Data Acquisitions for the Second Progress Report, The Japan Nuclear Cycle Development Institute Report, 1-58, JNC-TN-8400-99-083 (1999).
- [13] P. Bertetti. Determination of Sorption Properties for Sedimentary Rocks Under Saline, Reducing Conditions-Key Radionuclides, Nuclear Waste Management Organization Technical Report, 1-96, NWMO TR-2016-08 (2016).
- [14] S. Nagasaki, J. Riddoch, T. Saito, J. Goguen, A. Walker, and T. Yang, "Sorption Behaviour of Np(IV) on Illite, Shale and MX-80 in High Ionic Strength Solutions", *J. Radioanal. Nucl. Chem.*, 313, 1-11 (2017).
- [15] S. Nagasaki, T. Saito, and T. Yang, "Sorption Behaviour of Np(V) on Illite, Shale and MX-80 in High Ionic Strength Solutions", *J. Radioanal. Nucl. Chem.*, 308, 143-153 (2016).
- [16] M.H. Bradbury and B. Baeyens, "Modelling the Sorption of Mn(II), Co(II), Ni(II), Zn(II), Cd(II), Eu(III), Am(III), Sn(IV), Th(IV), Np(V) and U(VI) on Montmorillonite: Linear Free Energy Relationships and Estimates of Surface Binding Constants for Some Selected Heavy Metals and Actinides", *Geochim. Cosmochim. Acta*, 69(4), 875-892 (2005).
- [17] M.H. Bradbury and B. Baeyens, "Sorption Modelling on Illite. Part II: Actinide Sorption and Linear Free Energy Relationships", *Geochim. Cosmochim. Acta*, 73, 1004-1013 (2009).
- [18] A. Kirishima, O. Tochiyama, K. Tanaka, Y. Niibori, and T. Mitsugashira, "Redox Speciation Method for Neptunium in a Wide Range of Concentrations", *Radiochim. Acta*, 91, 191-196 (2003).
- [19] A. Kirishima. Private Communication (2014).
- [20] S. Nagasaki. "Extraction and Colloidal Geochemistry of Actinides", Ph.D. Dissertation, The University of Tokyo (1993).
- [21] Y. Inoue, O. Tochiyama, and N. Shinohara, "The Effect of Np Concentration on the Preparation of Np(III) by Hydrogen Reduction", *J. Inorg. Nucl. Chem.*, 42(5), 757-759 (1980).
- [22] L. Ciavatta, "The Specific Interaction Theory in the Evaluating Ionic Equilibria", *Ann. Chim. (Rome)*, 70, 551-562 (1980).
- [23] R. Guillaumont, Th. Fanghänel, J. Fuger, I. Grenthe, V. Neck, D.A. Palmer, and M.H. Rand, "Update on the Chemical Thermodynamics of Uranium, Neptunium, Plutonium, Americium and Technetium", in: *Chemical Thermodynamics*, F.J. Mompean, C.D. Orti, K.B. Said, and OECD/NEA Data Bank, eds., Vol. 5, Elsevier, Amsterdam (2003).
- [24] D.L. Parkhurst and C.A.J. Appelo. User's Guide to PHREEQC (Version 2): A Computer Program for Speciation, Batch-Reaction, One-Dimensional Transport and Inverse Geochemical Calculation, U.S. Geological Survey Water-Resources Investigation Report, 1-312, 99-4259 (1999).
- [25] J. Goguen, A. Walker, J. Racette, J. Riddoch, and S. Nagasaki, "Sorption of Pd on Illite, MX-80 Bentonite and Shale in Na-Ca-Cl Solutions", *Nucl. Eng.*, 53(3), 894-900 (2021).