JAEA-Research
2012-044

K_d Setting Approaches for Horonobe Mudstone Systems : Applications of TSMs and Semi-quantitative Estimation Procedures

Michael OCHS, Yukio TACHI, David TRUDEL and Tadahiro SUYAMA

Geological Isolation Research Unit
Geological Isolation Research and Development Directorate

March 2013

Japan Atomic Energy Agency

日本原子力研究開発機構

JAEA-Research

本レポートは独立行政法人日本原子力研究開発機構が不定期に発行する成果報告書です。
本レポートの入手並びに著作権利用に関するお問い合わせは、下記あてにお問い合わせ下さい。
なお、本レポートの全文は日本原子力研究開発機構ホームページ（http://www.jaea.go.jp）
より発信されています。

独立行政法人日本原子力研究開発機構　研究技術情報部　研究技術情報課
〒319-1195　茨城県那珂郡東海村白方白根 2 番地 4
電話 029-282-6387, Fax 029-282-5920, E-mail:ird-support@jaea.go.jp

This report is issued irregularly by Japan Atomic Energy Agency
Inquiries about availability and/or copyright of this report should be addressed to
Intellectual Resources Section, Intellectual Resources Department,
Japan Atomic Energy Agency
2-4 Shirakata Shirane, Tokai-mura, Naka-gun, Ibaraki-ken 319-1195 Japan
Tel +81-29-282-6387, Fax +81-29-282-5920, E-mail:ird-support@jaea.go.jp

JAEA-Research 2012-044

K_d Setting Approaches for Horonobe Mudstone Systems : Applications of TSMs and Semi-quantitative Estimation Procedures

Michael OCHS*, Yukio TACHI, David TRUDEL* and Tadahiro SUYAMA※

Geological Isolation Research Unit
Geological Isolation Research and Development Directorate
Japan Atomic Energy Agency
Tokai-mura, Naka-gun, Ibaraki-ken

（Received December 28, 2012）

Sorption of radionuclides in host rocks is the key processes in the safe geological disposal of radioactive waste, because migration of radionuclides in rocks is expected to be retarded by sorption processes. Sorption K_d values depends critically on relevant geochemical conditions, need to correspond to the specific performance assessment (PA) conditions, including geochemical variability or uncertainty. It is therefore necessary to understand the processes of sorption, and to develop the K_d setting approach including uncertainty assessment for reliable PA calculations. It is not feasible to determine K_d value experimentally for all PA conditions, key challenge is therefore transferring sorption data for experimental conditions to another PA and site-specific conditions.

JAEA has developed the integrated approaches for site-specific K_d setting for PA calculations, including any transferring procedures. This transfer can be done through expert judgment and semi-quantitative way, by considering difference in e.g. surface sites, speciations, competitive reactions, etc. The thermodynamic sorption model (TSM) makes it possible to estimate K_d variations directly, based on mechanistic understanding. The main goal of the development of PA-related integrated approaches for K_d setting is to provide the TSM and the estimation procedure, and their application for site-specific K_d setting for PA calculations. The present report focused on illustrating a range of example calculations regarding the derivation of K_d values and their uncertainties for four key radionuclides, Cs, Ni, Am and Th, and three mineralogical/geochemical conditions of mudstone systems from the Horonobe underground research laboratory. This K_d-setting exercises allowed to estimate the magnitude of sorption under the expected in-situ conditions, and to identify some critical gaps in the existing data and process understanding. Finally, the possibilities of extending such approaches to further rock types including granitic rocks were discussed. These issues are also relevant for the future experimental and modeling researches on sorption process for various radionuclides and rock systems.

Keywords: Geological Disposal, Radionuclide Migration, Sorption, K_d Setting, Uncertainty, Horonobe Mudstone, Thermodynamic Sorption Model, Semi-quantitative Estimation

---

This study was partly funded by the Ministry of Economy, Trade and Industry of Japan.

* BMG ENGINEERING LTD, Switzerland
※Collaborating Engineer

幌延泥岩に対する分配係数設定手法：
熱力学的収着モデルと半定量的推定手法の適用性

日本原子力研究開発機構　地層処分研究開発部門
地層処分基盤研究開発ユニット

Michael OCHS*, 舘 幸男, David TRUDEL*, 陶山 忠宏※

（2012 年 12 月 28 日受理）

放射性廃棄物地層処分の安全評価において，放射性核種の岩石中での収着現象は，その移行遅延を支配する重要な現象である。収着分配係数 K_d は地球化学条件によって大きく変化するため，性能評価においては，地球化学条件の範囲や不確実性を含む具体的な環境条件に対応付けて設定を行う必要がある。このため，収着現象を理解しつつ，信頼性の高い性能評価に資する不確実性を含む分配係数設定手法を構築する必要がある。その際，あらゆる性能評価条件下での実験データを実測することは現実的ではなく，実験条件から性能評価条件へと条件変換を行う手法が重要となる。

この目的のために，日本原子力研究開発機構では，性能評価における具体的な環境条件に対し，条件変換手法を含む分配係数設定手法の開発を進めてきた。この条件変換は，表面サイト，化学種，競合反応といった収着影響因子の差異を，専門家の判断，あるいは，半定量的な推定手法によって補正するものである。また，熱力学的収着モデルが利用可能な場合には，現象理解に基づくモデルを介して，分配係数の変化を直接評価可能である。性能評価のための分配係数設定手法の開発における最終的な目標は，これら熱力学的収着モデルや半定量的推定手法を構築し，具体的な性能評価条件に対する分配係数設定事例を提示することである。本報告では，この具体的な分配係数設定事例の検討として，幌延深地層研究所の泥岩系における複数の鉱物組成と地球化学条件を対象に，Cs, Ni, Am, Th の 4 核種の分配係数設定を試みた。本検討を通じ，実際の環境条件に対する分配係数設定手法を提示することができ，同時に，既存のデータや現象理解に関する重要な課題を抽出することができた。さらに，今回検討した分配係数設定手法を，花崗岩など他の岩種に適用する際の課題についても議論した。これらの課題は，今後の様々な核種と岩種を対象とした実験とモデル化に関する研究を通じて検討される必要がある。

本報告の一部は，経済産業省委託事業「処分システム化学影響評価高度化開発」の成果である。
核燃料サイクル工学研究所（駐在）:〒319-1194　茨城県那珂郡東海村村松 4-33
* BMG ENGINEERING LTD, スイス連邦共和国
※技術開発協力員

Contents

目　　次

1. Introduction

Sorption and diffusion of radionuclides (RNs) in buffer materials (bentonite) and host rocks (rock matrix) are the key processes in the safe geological disposal of radioactive waste, because migration of RNs in these barrier materials is expected to be diffusion-controlled and retarded by sorption processes. Sorption and diffusion of RNs on these barrier materials depends critically on relevant geochemical conditions, especially K_d values are highly conditional parameters (e.g., NEA, 2005). It is therefore necessary to understand the processes of sorption in bentonites and rock systems, and to develop the K_d setting approach including uncertainty assessment, so that reliable parameters can be set under a variety of geochemical conditions relevant to performance assessment (PA).

JAEA has developed the integrated approaches for site-specific K_d setting for PA calculations, as shown in Figure 1.1 (Tachi et al., 2011a), can be made available by three different approaches;

1) Experimental data acquisition for specific/reference conditions

2) Extraction and conversion from existing sorption data through sorption database (SDB)

3) Prediction by mechanistic (thermodynamic) sorption model

Because of the conditional nature of sorption data, K_d values to be used in PA calculations need to correspond to the specific conditions that characterize the respective PA-setting. In addition, geochemical variability or uncertainty, and their effect on K_d, usually have to be considered for reference and alternative scenarios in PA, as discussed in NEA (2005). It is not feasible to determine K_d value experimentally for all conditions to be considered in PA, K_d setting needs to be therefore based on the combination of above three approaches. The sorption database (JAEA-SDB; Tachi et al., 2011a; www//migrationdb.jaea.go.jp), containing large amount of sorption data for approximated, simplified, or generic systems, are used to K_d setting for PA conditions by taking into account any differences in substrates and geochemical conditions. This transfer can be done through expert judgment and semi-quantitative way, by considering difference in e.g. surface sites, speciations, competitive reactions, etc. (e.g., Bradbury and Baeyens, 2003a, 2003b; NEA, 2005; Ochs et al., 2008). The thermodynamic sorption model (TSM) makes it possible to estimate K_d variations directly, based on mechanistic understanding (NEA, 2005; 2012). JAEA has developed the integrated sorption/diffusion (ISD) database in combination with thermodynamic sorption and diffusion model for bentonite systems (e.g., Tachi et al., 2010; Ochs et al., 2011; JAEA, 2012), and for mudstone systems from the Horonobe underground research laboratory (URL) (Tachi et al, 2011b, JAEA, 2012).

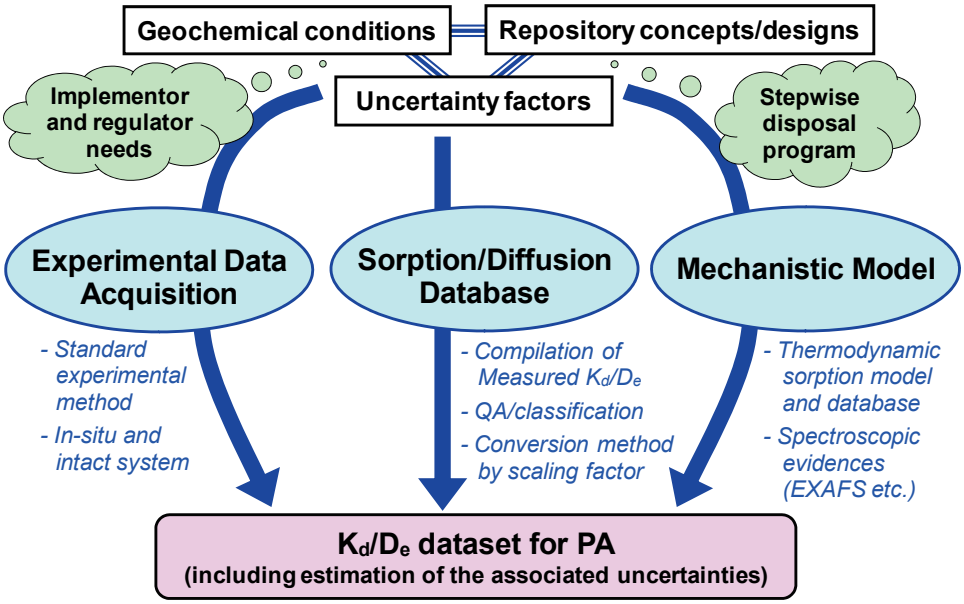

Figure 1.1 Integrated approach for sorption/diffusion parameter setting for PA (Tachi et al., 2011a)

The main goal of the development of integrated K_d setting approaches is to provide the TSMs and estimation procedure, and their application for site-specific K_d setting for PA calculations. The present report focuses on illustrating a range of example calculations regarding the derivation of K_d values and their uncertainties by using TSMs and estimation procedure.

This report includes:

- A summary of the present state of the art of PA-related K_d setting approaches by reviewing the approaches of different waste management organizations (Chapter 2)
- K_d setting approaches including i) the direct application of TSMs and ii) K_d estimation from experimental laboratory data through semi-quantitative estimation procedures, and the setting of geochemical conditions for mudstone systems from the Horonobe URL (Chapter 3)
- K_d setting results for four key radionuclides, Cs, Ni, Am, Th, and three geochemical conditions including the effects of variations in mudstone and groundwater composition (Chapter 4)
- Brief discussions on the extension of K_d setting approaches to further rock systems (Chapter 5).

2. Overview of K_d setting approaches for the performance assessment

To provide a framework of PA-related K_d setting approaches currently in use, a brief summary is provided regarding the methods for K_d setting and uncertainty evaluation taken from international sorption project (NEA, 2005; 2012) and K_d setting reports by various waste management organizations (e.g., Bradbury and Baeyens, 2003a, 2003b; Ochs and Talerico, 2004; Crawford et al., 2006).

2.1 Overview of K_d setting and uncertainty evaluations

As presented in introduction (Figure 1.1), the development of integrated approaches for site-specific K_d setting for PA calculations is the goal of our project. Because of the conditional nature of sorption K_d values, geochemical variability or uncertainty, and their effect on K_d, usually have to be considered for reference and alternative scenarios in PA calculations. It is not feasible to determine K_d value experimentally for all conditions to be considered in K_d setting. A typical challenge is the relatively limited amount of site-specific data can be made available, due to experimental constraint and complexity of conditions. In this setting, key challenge is transferring sorption data from one set of conditions (experimental condition in SDB) to another (PA-related site-specific condition). This must take into account any differences in substrate (mineralogy, site density, etc.) and geochemical conditions (pH, ionic strength, etc.). This transfer can be done through the following methods (as presented in NEA, 2005, and Figure 2.1):

1) qualitative expert judgment (e.g., H12 PA; JNC, 1999)
2) semi-quantitative transfer by scaling method (e.g., Bradbury and Baeyens, 2003a, 2003b; Ochs and Talerico, 2004; Ochs et al., 2008)
3) fully quantitative transfer through the use of TSM (e.g., NEA, 2005; Tachi et al., 2010)

Typically, K_d setting by application of estimation procedures or by modeling is necessary because many sorption values required for PA cannot be measured under the specific conditions that need to be addressed. Typical reasons for this are

- the large numbers of RNs and often also the wide range of conditions that need to be covered in PA exercises;
- the difficulties in reproducing experimentally the specific conditions of interest.

Accordingly, it is widely accepted that K_d values for PA largely have to be estimated on the basis of the relatively large amount of data existing for generic systems. Where possible, these should be supplemented with K_d values measured for site-specific conditions (NEA, 2005, 2012).

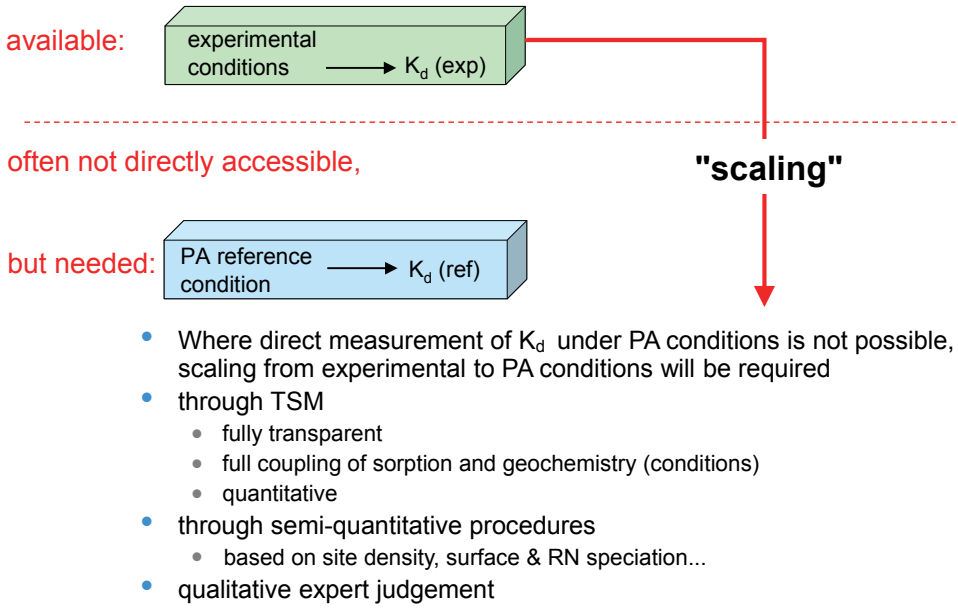

Figure 2.1 PA-related K_d setting approaches based on experimental data and scaling method (from NEA, 2005)

2.2 Steps in K_d setting and sources of uncertainty

K_d values recommended for PA are often associated with relatively large uncertainties. These uncertainties are made up of several contributions. Of these, only some are actually related to sorption, while a significant part of the overall uncertainty is related to uncertainties in the specified geochemical conditions.

Most K_d setting reports use a very similar approach to define K_d values for RN migration in clayey media. Briefly, this includes the following steps:

1. Definition of all geochemical conditions in the respective host formation. All possible variations of conditions are typically considered at this step. Some uncertainties in geochemical parameters (e.g. groundwater pH) are handled by the definition of bounding water compositions (as pH and other parameters cannot be varied independently).

2. In case of clay barriers, porewater compositions are calculated on the basis of the conditions defined in step 1. This has to be done through the use of basic clay-water models involving ion exchange and edge surface protonation/deprotonation. Details of the models (as well as the assignment of porosity to pore types), and therefore of the calculated results, vary from organization to organization.

3. K_d values are derived on the basis of the porewater compositions defined following step 2 above. Most K_d setting reports typically make use of a mix of semi-quantitative estimations (see chapter 3) and the application of TSMs for a few radionuclides (often only those sorbing by ion exchange). It appears that to date, none of the organizations make any adjustments for the use of batch-derived data in predicting sorption for compact clay. I.e., it seems to be largely accepted that the nature of sorption processes as well as the magnitude of sorption in compacted clay are not significantly different from those in disperse systems.

In terms of uncertainties, it appears to be generally realized that the success of such approaches is critically dependent on the degree to which the general geochemical conditions can be constrained. Only in case of well-defined conditions is it possible to apply estimation procedures or TSMs in a straightforward fashion. Therefore, uncertainties of reference conditions should be taken care of by defining a few cases with bounding conditions. Data derivation can then be repeated for these bounding conditions to evaluate the possible effects of geochemical uncertainties.

2.3 Handling of uncertainties in K_d setting

According to our experience, K_d setting and assessment of uncertainties in sorption is normally done using roughly the following steps:

- First, an assessment of the overall data situation and available models is made for each RN that needs to be considered (and for different oxidation states, if relevant).
- From the available datasets and/or TSMs, the most relevant data source is selected, considering quality of the underlying data and closeness to application conditions.
- For alkaline and alkaline-earth elements, often a simple ion exchange model is selected. On the other hand, most other RNs with a more complex sorption behavior are handled by estimation procedures to date, rather than by sorption models. Common arguments for this decision are that the available TSMs are not sufficiently validated with respect to complex materials or that they are not sufficiently calibrated in terms of the influence of the typical porewater constituents (carbonate, silica, Ca, Mg…). Since the estimation procedures include a correction for RN speciation (see chapter 4), this approach is often preferred.
- An important observation can be made with regard to the selection of reference datasets: it appears to be largely accepted to carry out data derivation using single datasets. In case of many of the less well researched radionuclides, this is the only possibility, due to the lack of enough reliable and systematic data. On the other hand, in some cases multiple K_d derivation could be possible using several of the available datasets. Such an approach could be used to assess uncertainties that may result from different experimental protocols used to measure different datasets.
- More typically, a single set of reference data (or a single TSM) is chosen for K_d setting, and arguments are presented to defend the selection.
- Uncertainties of the K_d values derived on the basis of selected reference data are then evaluated in a manner similar to the approach documented in this report (see chapter 3 and 4). In case of TSM application, similar uncertainty factors are used.
- K_d values derived on the basis of generic sorption data can be supplemented with further information to strengthen the case:
    - Ochs and Talerico (2004) compared experimental D_a values with D_a values calculated from the selected K_d values and D_e (typically of HTO). This can be used as a general check of consistency but not directly for defining the uncertainty of K_d.

- Another option is to supplement the derived K_d values with at least a few values measured under specific conditions. This has been done mainly by Nagra. For example, the selection of a K_d for Opalinus Clay is based, where possible, on TSMs developed for smectite and illite as well as sorption data measured in Opalinus Clay samples under relevant conditions that can be used for estimation procedures.

3. Approaches for K_d setting by TSMs and semi-quantitative estimation procedures

K_d setting calculations for mudstone systems from the Horonobe URL have been carried out based on the following approaches. These calculations included the direct application of TSMs to the specified conditions as well as the estimation of K_d values from experimental laboratory data through semi-quantitative estimation procedures. In terms of radionuclides, Cs, Ni, Am, and Th were considered. The effect of variations in mudstone and groundwater composition on sorption was also investigated.

3.1 Overview of TSMs and modeling approaches

Different sorption models have been developed within the project developing the mechanistic sorption model and database for clay systems (Tachi et al., 2010; Ochs et al., 2011; JAEA, 2012). These models were developed based on experimental data from the literature, which had originally been obtained in batch sorption experiments under defined and simplified conditions. Based on the general mineralogy of sedimentary rocks from the Horonobe URL, model development within the JAEA model and database program initially focused on smectite as model sorbent, and was subsequently extended for some RN to include illite (JAEA, 2012). The present chapter discusses the application of some of these sorption models to the prediction of K_d values for mudstone systems of the Horonobe URL, as an illustration of TSM application in a hypothetical safety assessment context.

The modeling approach used for the Horonobe mudstone systems is provided Figure 3.1. The models used for the calculations described in this chapter are briefly summarized below. With exception of the Cs-models taken directly from the literature, all RN-TSMs are based on the underlying clay-water model JAEA11NEM (JAEA, 2012), based on 1-site surface complexation model without EDL correction term and 1-site ion exchange model.

- For Ni, Am(Eu) and Th, the models listed in Table 3.1 were used. These models were developed for smectite. For the present application, they are assumed to be applicable to illite as well, as a first approximation.
- For Cs, adequate models are available in the literature. The models by Wanner et al. (1996) for Cs sorption on smectite and of Bradbury and Baeyens (2000) for Cs sorption on illite were adopted for the present purpose.

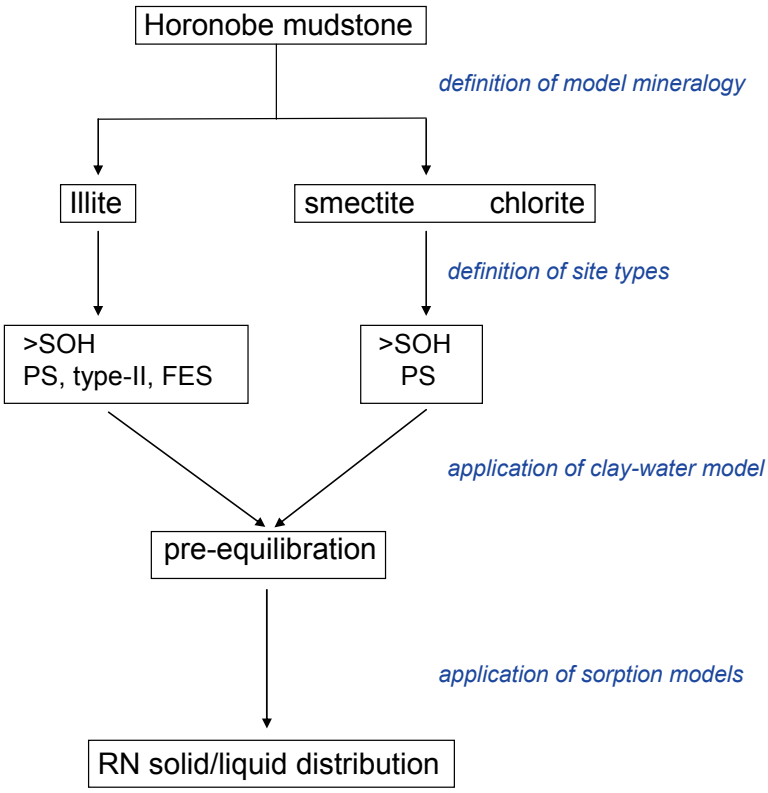

Figure 3.1 Overview of modeling approach for Horonobe mudstone systems. PS are planar (siloxane) ion exchange sites, type-II and FES (frayed edge sites) are illite-specific ion exchange sites, >SOH stands for generic edge surface sites.

Using the different groundwater and simplified rock sample compositions, the first calculations were performed to equilibrate the solid surfaces and aqueous phase at a reasonable solid/liquid ratio (assuming ca. 40 % water-filled porosity and a density of 2.7). The resulting composition of exchangeable ions on the smectite and illite surfaces was then used to calculate the equilibrium distribution of the various radionuclides. All calculations were done using the geochemical code PHREEQC (Parkhurst and Appelo, 1999). Summaries of the model setups for nickel, americium/europium and thorium are shown in Table 3.1, the corresponding summary for the cesium models is given in Table 3.2 : Because the Cs model is accepted form the literature (Bradbury and Baeyens, 2000) as is, the ion exchange constants for some major cations (K^+, NH_4^+) on the planar sites of illite are slightly different from the corresponding constants in the JAEA11NEM model (JAEA, 2012).

Table 3.1 Model summaries for nickel, thorium, and americium/europium. PS are planar sites (siloxane layer), type-II and FES (frayed edge sites) are illite-specific ion exchange sites.

Clay minerals	**Illite**			**Smectite**
Ion exchange				
CEC [meq/100g]	20.05[c]			110
Site type	FES Q	Type II X	PS Y	PS Z
Exchange site capacity [meq/100g]	0.05	4	16	110
(basic clay-water model JAEA11NEM) Site-Na Site-K Site-NH_4 $(Site)_2$-Ca $(Site)_2$-Mg	 20[d] 22.4[c] 23.5[c] - -	 20[d] 22.1[c] 23.04** - -	 20[d] 20.42[a] 20.46[b] 40.69 40.67	 20[d] 20.42[a] 20.46[b] 40.69 40.67
JAEA11NEM_Th $(Site)_4$-Th	 -	 -	 82.92	 82.92
JAEA11NEM_Am(Eu) $(Site)_3$-Am	 -	 -	 62.05	 62.05
JAEA11NEM_Ni $(Site)_2$-Ni	 -	 -	 40.75	 40.75
Surface complexation				
SOH-site density [mol/g] all edge surface sites	4.31E-5			4.31E-5
(basic clay-water model JAEA11NEM) $H^+ + SOH = SOH^{2+}$ $SOH = SO^- + H^+$	 5.75 -8.30			 5.75 -8.30
JAEA11NEM_Th $>SOTh^{3+}$ $>SOTh(OH)_3$ $>SOTh(OH)_4^-$	 7.07 -10.40 -18.59			 7.07 -10.40 -18.59
JAEA11NEM_Am(Eu) $>SOAm^{2+}$ $>SOAmOH^+$ $>SOAm(OH)_2$ $>SOAm(OH)_3^-$ $>SOAmCO_3$ $>SOAm(CO_3)_2^{2-}$	 0.30 -7.62 -16.57 -27.30 6.74 10.42			 0.30 -7.62 -16.57 -27.30 6.74 10.42
JAEA11NEM_Ni $>SONi^+$ $>SONiOH$ $>SONi(OH)_2^-$	 -2.32 -12.16 -21.02			 -2.32 -12.16 -21.02

[a] Oda and Shibata (1999)
[b] Fletcher and Sposito (1989)
[c] Bradbury and Baeyens (2000)
[d] by definition
** interpolated in analogy to the constants for K^+

Table 3.2 Model summary for cesium. PS are planar sites (siloxane layer), FES are frayed edge sites.

Clay minerals	**Illite**			**Smectite**
Ion exchange				
CEC [meq/100g]	20.05[c]			110
Site type	FES Q	Type II X	PS Y	PS Z
Exchange site capacity [meq/100g]	0.05	4	16	110
(basic clay-water model JAEA11NEM) Site-Na Site-K Site-NH_4 Site-Ca Site-Mg	 20[d] 22.4[c] 23.5[c] - -	 20[d] 22.1[c] 22.8** - -	 20[d] 21.1[c] 20.46[b] 40.69 40.67	 20[d] 20.42[a] 20.46[b] 40.69 40.67
Cs-Model (Bradbury and Baeyens, 2000; Wanner et al. 1996) Site-Cs	27[c]	23.6[c]	21.6[c]	21.6[c]
Surface complexation				
SOH-site density [mol/g] all edge surface sites	4.31E-5			4.31E-5
(basic clay-water model JAEA11NEM) $H^+ + SOH = SOH^{2+}$ $SOH = SO^- + H^+$	 5.75 -8.30			 5.75 -8.30

[a] Oda and Shibata (1999)
[b] Fletcher and Sposito (1989
[c] Bradbury and Baeyens (2000)
[d] by definition
** interpolated in analogy to the constants for K^+

3.2 Semi-quantitative estimation approaches

For each RN, the results of the model applications are compared with K_d values estimated through semi-quantitative procedures. For this purpose, the semi-quantitative estimation procedure developed earlier (Ochs et al., 2008; see also Ochs and Talerico, 2004; Bradbury and Baeyens, 2003a, 2003b) has been used. The K_d values for the Horonobe mudstone system were derived using available sorption data of similar water-rock systems taken from the web-version JAEA-SDB (Tachi et al., 2011a). The pre-equilibration step and the resulting occupation of the exchange sites on the illite and smectite components with major cations are discussed in chapter 3.3.

3.2.1 Conversion to application conditions

In general, the approach chosen by Ochs et al. (2008) was followed (see also Ochs and Talerico, 2004; as well as Bradbury and Baeyens, 2003a, 2003b). The scaling factors accounting for differences in sorption capacity and surface as well as solution chemistry in the experimental vs. application conditions are discussed below. Beforehand, the more general issue of using batch data to derive K_d-values for intact rock is discussed in the following section.

i) Use of batch data to derive K_d-values for intact rock

Practically sorption measurements are usually carried out in dilute suspensions. Reliable sorption measurements in intact material are extremely scarce due to experimental difficulties. This situation necessitates the use of sorption data from batch experiments to derive K_d-values valid for intact rock; i.e., the transfer of data from low to very high solid/water ratios.

While K_d is highly dependent on the chemical conditions (taken into account by the scaling factors discussed below), it is in principle independent of the solid/water ratio, and no data conversion is necessary given that the available specific surface area or sorption site density per unit mass as well as the solution composition remain constant. This can be very difficult to assess in a fractured/ heterogeneous rock (e.g., granite), but appears to be nearly established in the case of homogeneous porous media such as bentonite and clay rocks (NEA, 2012).

With regard to the available surface area, Bradbury and Baeyens (1998) measured similar BET values for loose and compacted (2000 kg/m^3) MX-80 and Montigel bentonites. The diffusion data by Kato et al. (1995) also suggest that no reduction of sorption-available surface area takes place upon compaction. Based on such evidence, it was concluded by e.g. Ochs et al. (2001), Bradbury and Baeyens (2003a, 2003b) and Ochs and Talerico (2004) that batch data can be directly applied to compacted systems, provided that any differences in solution composition are accounted for Ochs and Talerico (2004) (see also Ochs et al., 2008) derived K_d-values valid for compacted bentonite for a large number of RN, using scaling factors similar to the ones discussed below. From these K_d-values and D_e for HTO, they calculated the respective D_a-values and compared these to independent experimental D_a-values. This comparison indicated that no conversion factor (CF) is necessary to account for the transfer from batch to compacted systems, as long as the respective chemical conditions are taken into account.

From the information available, it is estimated for the present application that the relevant rock type (mudstone) can be treated in a 1st approximation as homogeneous porous media, and that CF-exp→intact (conversion factor from batch to intact system) is unity. In particular, the results and discussion in Tachi et al. (2011b) support this concept.

ii) Sorption capacity, mineralogy

The working hypothesis for the present K_d-setting is that the sorption capacity of the Horonobe mudstone is determined by the respective content of clay minerals (smectite, chlorite, kaolinite, as well as illite). All available evidence suggests that these clay minerals are able to sorb radionuclides through surface complexation and ion exchange (see e.g., NEA, 2005).

To scale selected experimental sorption data to the present application conditions (i.e., a solid substrate representative of the Horonobe mudstone in equilibrium with HDB-6 groundwater), a conversion or scaling factor (CF-min) based on the relative amount or sorption capacity of clay minerals present in a defined volume of the selected experimental systems vs. in the Horonobe mudstone needed to be defined. Generally, the scaling factor CF-min can be based on:

- the exchange capacity of both solids, CEC (CF-CEC),
- the relative amount of clay minerals (CF-%clay),
- the relative (BET) surface area (CF-BET).

In the case of sorption by clays, such a factor can be based on the ratio of CEC per unit mass for the experimental and application system. CEC value for the Horonobe mudstone was measured as about 20 meq/100 g (Tachi et al., 2011). The CEC was therefore also calculated by considering typical CEC values for pure smectite, chlorite, kaolinite and illite and calculating a mass-weighted mean value for the mineralogical composition of the Horonobe mudstone. The results are given in Tables 3.3 and 3.4 in chapter 3.3.

Scaling on the basis of CEC values was done using the following relation:

$$\text{CF-CEC} = \text{CEC (application)} / \text{CEC (data source)} \quad (3.1)$$

In this relation, CEC is used as a relative measure for total site density, based on the reasonable assumption that the density of surface complexation (edge) sites is proportional to the CEC.

Because of the uncertainty regarding CEC and the assumed relationship between CEC and surface complexation site density, data derivation was additionally based on the ratio of total clay content in both systems (Eq. 3.2). Further, the BET-surface area was also considered (Eq. 3.3) to obtain some information of the sensitivity of the conversion factor for sorption capacity on the actual measure used.

$$\text{CF-\%clay} = \text{\%clay (application)} / \text{\%clay (data source)} \quad (3.2)$$

$$\text{CF-BET} = \text{BET (application)} / \text{BET (data source)} \quad (3.3)$$

iii) pH

The pH is the most critical geochemical parameter for the sorption of most elements as it determines

- the speciation of the sorbing element itself, in particular hydrolysis (relevant for Ni, Am, Th), as well as protonation-deprotonation in case of anions such as Se (see Ochs et al., 2008),
- the protonation-deprotonation of the surface complexation sites,
- the speciation and solubility behavior of major ligands (e.g., carbonate) and major cations (e.g., Ca) that have an important influence on sorption through complexation (e.g., carbonate complexation of Am and Th) and competition (e.g., Ca with Cs, but possibly also carbonate in case of Se).

For this reason, to the degree possible data sources were selected that allowed the selection of sorption data at pH values corresponding to the application conditions (in the data derivation tables in Appendix I, this is indicated by a pH-conversion factor of 1). Where this is not possible, scaling to the appropriate application pH can be done on the basis of additional data for the same RN, such as sorption edges on a montmorillonite. In such cases, scaling to the application pH was done using the conversion factor defined below:

$$\text{CF-pH} = K_d \text{ (pH application)} / K_d \text{ (pH data source)} \quad (3.4)$$

In contrast to Bradbury and Baeyens (2003a, 2003b), CF-pH was interpreted by Ochs and Talerico (2004) as a factor addressing the overall surface/solution speciation. Thus, the conversion factor for additional radionuclide speciation effects defined below (CF-spec) addresses only those effects not included in CF-pH. In particular, CF-spec does not take into account hydrolysis.

iv) Effects on radionuclide surface/solution speciation by dissolved ligands and major cations

As pointed out above, dissolved ligands and major cations can have a significant influence on radionuclide sorption. Therefore, scaling of K_d to application conditions should in principle be carried out in all cases where the source data correspond to a solution composition that is significantly different from that of the reference groundwater. Only thermodynamic sorption models are capable of consistently taking into account all speciation effects. For the present application, it was attempted to account for variable solution compositions in a semi-quantitative, but fully traceable fashion. The following different processes need to be considered:

- RN sorption may decrease due to the formation of aqueous complexes with dissolved ligands (in case of Ni, Am, Th) or with dissolved cations (in case of Se). This is taken into account by the conversion factor CF-spec. For Cs with its simple solution chemistry, this is not a relevant process.
- In case of Cs sorption takes mainly place through weak (electrostatic) interaction (cation or anion exchange on the layer surface, outer-sphere complexation at the edge surface). Here, RN sorption may also decrease due to competition by major cations (Cs). This is taken into account by the conversion factor CF-cmp.

CF-spec:

The formal definition of the factor CF-spec is accepted from Ochs and Talerico (2004).

$$\text{CF-spec} = F_{sorb}\ (\text{application}) \,/\, F_{sorb}\ (\text{data source}) \qquad (3.5a)$$

where

$$F_{sorb} = (RN_{tot} - RN_{cmp}) \,/\, RN_{tot} \qquad (3.5b)$$

with RN_{tot} as the total dissolved concentration of a given radionuclide, and RN_{cmp} as the total concentration of all dissolved RN complexes evaluated as being competitive with regard to sorption.

However, the application of this factor is not straightforward:

- First, the conversion factor CF-pH will already include one or more of the effects of speciation. The following differences in speciation are therefore not included in the factor CF-spec, as the effects would be counted twice:
  - As pointed out above, effects of RN hydrolysis will already be included in CF-pH.
  - Complexation of Am or Th with the carbonate ion (CO_3^{2-}) can differ between two solutions because of i) different total carbonate concentrations, or ii) different pH values leading to different concentrations of CO_3^{2-} even when total carbonate levels are the same. In case ii), the effect will also be included in the conversion factor CF-pH.
- Second, several elements, especially IV- and higher-valent actinides are known or suspected to form mixed hydroxo-carbonato complexes. By analogy, the formation of ternary RN-carbonato surface complexes at the clay edge sites can be assumed. For some cases, the existence of such complexes has been confirmed by spectroscopy (e.g. for U(VI) sorption on montmorillonite by Catalano and Brown, 2005). Therefore, it is highly questionable in these cases whether the presence of carbonate should be counted as a competitive factor. This situation is complicated further by the

uncertainties regarding the existence and importance of various simple and mixed An(IV)-carbonato complexes (see discussions in e.g., Guillaumont et al., 2003; Rand et al., 2009). Therefore, scaling of K_d to account for speciation was carefully evaluated for both Am and Th. As a basis for further evaluation, CF-spec was calculated twice in these cases, taking into account or neglecting competition by complexation involving carbonate ions.

- CF-spec/a: Complexation of a radionuclide with carbonate is viewed as being fully competitive with respect to sorption.
- CF-spec/b: Complexation of a radionuclide with carbonate is viewed as being not competitive with respect to sorption.

CF-cmp:

As pointed out at the beginning of this section, CF-cmp is only relevant in the case of the relatively weakly sorbing Cs. Ni, Am and Th sorb mainly through the formation of strong inner-sphere surface complexes at neutral or slightly alkaline pH and are not notably influenced by competing cations such as Ca. CF-cmp is defined as

$$\text{CF-cmp} = \Sigma_{\text{competing cations/anions}}\ (\text{data source}) \,/\, \Sigma_{\text{competing cations/anions}}\ (\text{application}) \qquad (3.6)$$

v) Summary of scaling factors

Overall scaling of experimental sorption data to the application conditions defined for the Horonobe mudstone is done by combining the individual conversion factors discussed above:

$$\text{CF-overall} = \text{CF-min} \times \text{CF-pH} \times \text{CF-spec} \times \text{CF-cmp} \qquad (3.7)$$

As discussed, some of these may only be important in some cases (such as CF-cmp in the case of Cs). For reasons of transparency, and to illustrate the effects of the various scaling factors, the overall scaling procedures listed below were applied in a step-wise fashion:

$$\text{CF-1} = \text{CF-min} \times \text{CF-pH} \qquad (3.8a)$$

$$\text{CF-2} = \text{CF-min} \times \text{CF-pH} \times \text{CF-spec} \qquad (3.8b)$$

$$\text{CF-3} = \text{CF-min} \times \text{CF-pH} \times \text{CF-spec} \times \text{CF-cmp} \qquad (3.8c)$$

where CF-min is represented by each CF-CEC, CF-%clay, and CF-BET. Also, both CF-spec/a and CF-spec/b were compared where relevant.

3.2.2 Estimation of uncertainties

The treatment of uncertainties follows closely the approach taken by Ochs et al. (2008) (see also Ochs and Talerico, 2004). There is no obvious reason for choosing between quantifying uncertainties as an error on a linear scale (i.e., K_d ± error) vs. a logarithmic scale (i.e., log K_d ± log error). Following the procedure chosen in NEA (2005), the latter type of representation was adopted for the present purpose. On a linear scale, this translates to K_d multiplied/divided by an uncertainty factor UF:

$$\log K_d \text{ (upper, lower limit)} = \log K_d \pm \log \text{uncertainty, or} \quad (3.9a)$$

$$K_d \text{ upper limit} = K_d \times \text{UF and } K_d \text{ lower limit} = K_d / \text{UF} \quad (3.9b)$$

Uncertainties are generally evaluated in a way that makes it likely that the indicated limits encompass all possible values. Where data are more uncertain, this is discussed specifically.

The following uncertainty factors (log errors) are considered:

i) Uncertainty of source data (UF-starting K_d)

For good quality experimental data, an uncertainty of log K_d ± 0.2 log units is proposed based on NEA (2005). This gives an uncertainty factor

→ UF-starting K_d = 1.6

ii) Uncertainty introduced by scaling to application conditions

- *sorption capacity conversion (UF-min):*

  It is estimated that CEC can be measured within an uncertainty of ca. 10 %, and that SOH density can be scaled via CEC again within an uncertainty of ca. 20%. This gives an overall uncertainty of ± 30% or about a factor of 2 between highest and lowest value. This uncertainty is also accepted for sorption capacity conversions based on %clay or BET. Thus, we propose

  → UF-min = 1.4

- *pH conversion (UF-pH):*

  This is only needed where CF-pH ≠ 1, i.e., where scaling to application pH had to be done via additional data and equation (3.4). Because CF-pH contains uncertainties of two K_d values (which are read off at pH-data source and pH-application, respectively), an uncertainty log starting K_d ± 2×0.2 log units is used:

  → UF-pH = 2.5

- *Speciation (UF-spec):*

  Following the evaluation by Hummel and Berner (2002), who propose a factor of 2 between highest and lowest value, a UF-speciation = 1.4 is used. Note, however, that this is the uncertainty associated with the use of a given, self-consistent TDB. If certain species are missing or erroneous

(see e.g. the discussion on mixed actinide OH-CO_3 complexes above), uncertainties could be much higher. Also, this UF does not take into account any inappropriate evaluation of CF-spec.

- *Competition by major cations/anions (UF-cmp):*

  It is assumed that the sum of cation or anion concentration in each system can be known or predicted within 10%, giving an overall uncertainty of about ± 20%.

  → UF-cmp = 1.2

- *Conversion of batch data to conditions in intact rock (UF-exp→intact):*

  Arguments for the applicability of the K_d values derived in this report to intact rock are presented above. Accordingly, CF-exp→intact is unity and no UF is proposed for this conversion per se.

Therefore, an additional uncertainty factor could be introduced to acknowledge uncertainties in the application conditions (i.e., in the mineral and porewater composition). Such an uncertainty factor can only be defined following a detailed assessment of the conditions (and their possible variations) to be considered in K_d-setting. For the present illustrative exercise in K_d-setting, such uncertainties have not been considered.

3.3 Geochemical conditions and pre-equilibration

The calculations documented in this chapter were performed for three alternative conditions. These include two different compositions of Horonobe rock and two types of groundwater (see Tables 3.3, 3.4, 3.5). In total, the following three different cases were calculated:

Reference conditions
Case I: mudstone A (11% smectite, 7% illite, 1% chlorite), in-situ groundwater
Alternative conditions
Case II: mudstone A (11% smectite, 7% illite, 1% chlorite), synthetic groundwater
Case III: mudstone B (16% smectite, 10% illite, 1% chlorite), synthetic groundwater

The groundwater and rock compositions in Case I were used as representative for the HDB-6 borehole from a depth of 450m in previous K_d calculations (Ochs et al., 2008). In terms of sorption-relevant minerals, only smectite, illite and chlorite were considered for all cases, where chlorite was assumed to have the same ion exchange and edge site properties as smectite. Higher clay contents used in Case-III are based on a discussion in Tachi et al. (2011b), whereby clay contents were corrected by considering CEC and specific surface area measured by BET and EGME methods. Two different groundwaters were considered as shown in Table 3.5: The in-situ groundwater composition was estimated for a depth of

450m (JAEA, 2007), it has a relatively low pH. The synthetic groundwater is based on pumping tests, it has a simplified composition with a higher pH of 8.5 (Hama et al., 2007; Tachi et al., 2011).

Using the different groundwater and simplified rock sample compositions, the first calculations were performed to equilibrate the solid surfaces and aqueous phase at a reasonable solid/liquid ratio (assuming ca. 40 % water-filled porosity and a density of 2.7). The resulting composition of exchangeable ions on the smectite and illite surfaces was then used to calculate the equilibrium distribution of the various radionuclides. All calculations were done using the geochemical code PHREEQC. Summaries of the model setups for nickel, americium/europium and thorium are shown in Table 3.1, the corresponding summary for the cesium models is given in Table 3.2: Because the Cs model is accepted form the literature (Bradbury and Baeyens, 2000) as is, the ion exchange constants for some major cations (K^+, NH_4^+) on the planar sites of illite are slightly different from the corresponding constants in the JAEA11NEM model (JAEA, 2012).

Table 3.3 Summary of the properties of the mudstone A Horonobe rock sample considered for the present calculations for cases I and II. The composition was provided by JAEA (2007), CEC and BET values were estimated based on the typical values indicated (cf. Dixon and Weed, 1977; Sposito, 1984, Bradbury and Baeyens, 2000).

	typical values		**Horonobe rock HDB-6, -450m**		
Mineral	CEC [*meq/100g*]	BET [m^2/g]	weight %	calc. CEC [*meq/g*]	calc. BET [m^2/g]
Quartz			13		
Andalusite			2		
Albite			6		
Anorthite			2		
K-feldspar			5		
Illite	20.05	24.2	7	1.40	1.69
Smectite	110	35	11	12.10	3.85
Chlorite	25	0.5	1	0.25	0.01
Limonite			1		
Pyrite			3		
Ilmenite			1		
Halite			2		
Res. SiO_2			42		
Total			96	13.75	5.55

Table 3.4 Summary of the properties of the mudstone B Horonobe rock sample considered for the present calculations for *Case III*. The composition was provided by Tachi et al. (2011), CEC and BET values were estimated based on the typical values indicated (cf. Dixon and Weed, 1977; Sposito, 1984, Bradbury and Baeyens, 2000).

	typical values		**Horonobe rock HDB-6, -450m**		
Mineral	CEC [*meq/100g*]	BET [m^2/g]	weight %	calc. CEC [*meq/g*]	calc. BET [m^2/g]
Quartz			13		
Albite			6		
K-feldspar			5		
Illite	20.05	24.2	10	2.01	2.42
Smectite	110	35	16	17.60	5.60
Pyrite			3		
Andalusite			2		
Anorthite			2		
Limonite			1		
Opal-CT			42		
Total			100	19.61	8.02

Table 3.5 The reference groundwater conditions for the Horonobe mudstone systems. The groundwater A estimated for in-situ conditions in JAEA (2007) was considered for the present calculations for cases I. The groundwater B based on Hama et al.(2007) and Tachi et al.(2011) was considered for cases II and III.

	Groundwater A (Case I)	Groundwater B (Case II and III)
Temp.(^{o}C)	25	25
pH	6.75	8.50
Ionic strength	2.58×10^{-1}	2.57×10^{-1}
Dissolved species	[mol/L]	[mol/L]
Na^{+}	2.28×10^{-1}	2.30×10^{-1}
K^{+}	2.08×10^{-3}	2.10×10^{-3}
Mg^{2+}	5.78×10^{-3}	5.80×10^{-3}
Cl^{-}	2.20×10^{-1}	2.22×10^{-1}
Br^{-}	5.63×10^{-4}	-
Ca^{2+}	2.09×10^{-3}	-
F^{-}	1.05×10^{-5}	-
Sr^{2+}	2.97×10^{-5}	-
Mn^{2+}	3.64×10^{-7}	-
Al^{3+}	3.71×10^{-7}	-
HCO_3^{-}	2.99×10^{-2}	3.00×10^{-2}
CO_3^{2-}	2.04×10^{-5}	-
SO_4^{2-}	4.38×10^{-6}	-
NH_4^{+}	7.76×10^{-3}	7.80×10^{-3}
Fe^{2+}	1.52×10^{-5}	-
H_4SiO_4(aq)	1.14×10^{-3}	-
HPO_4^{2-}	5.06×10^{-7}	-
PO_4^{3-}	7.62×10^{-12}	-

The occupation of the exchange sites on the illite and smectite model components with major cations after the pre-equilibration step is shown as follows:

- corresponding to the *Case I* condition: Table 3.6;
- corresponding to the *Case II* condition: Table 3.7;
- corresponding to the *Case III* condition: Table 3.8.

The pre-equilibration is performed using constants given in Table 3.1. Although the constants for the cesium model are slightly different from the constants for the other models (see Table 3.2), the same pre-equilibration results have been used for all further model applications.

Table 3.6 Composition of ion exchange sites after pre-equilibration of mudstone with Horonobe groundwater *(Case I)*. S/W is 4.12 kg/L.

Exchange site [a]	**Concentration (mol/L)**	**Exchange site** [a]	**Concentration (mol/L)**
CaY_2	1.1E-03	MgY_2	4.1E-04
CaZ_2	1.2E-02	MgZ_2	4.9E-03
HQ	3.5E-11	NaQ	3.2E-05
HX	5.6E-09	NaX	5.1E-03
HY	4.7E-08	NaY	4.3E-02
HZ	5.6E-07	NaZ	5.1E-01
KQ	7.1E-06	NH_4Q	1.1E-04
KX	5.7E-04	NH_4X	5.8E-03
KY	1.0E-04	NH_4Y	1.3E-04
KZ	1.2E-03	NH_4Z	1.5E-03

[a] exchange sites are defined in Table 3.1.

Table 3.7 Composition of ion exchange sites after pre-equilibration of mudstone with Horonobe groundwater *(Case II)*. S/W is 4.12 kg/L.

Exchange site [a]	**Concentration (mol/L)**	**Exchange site** [a]	**Concentration (mol/L)**
CaY_2	3.3E-05	MgY_2	4.1E-04
CaZ_2	3.9E-04	MgZ_2	4.9E-03
HQ	6.4E-13	NaQ	3.2E-05
HX	1.0E-10	NaX	5.1E-03
HY	9.0E-10	NaY	4.5E-02
HZ	1.1E-08	NaZ	5.3E-01
KQ	7.1E-06	NH_4Q	1.0E-04
KX	5.7E-04	NH_4X	5.8E-03
KY	1.0E-04	NH_4Y	1.3E-04
KZ	1.2E-03	NH_4Z	1.6E-03

[a] exchange sites are defined in Table 3.1.

Table 3.8 Composition of ion exchange sites after pre-equilibration of mudstone with Horonobe groundwater *(Case III).* S/W is 4.12 kg/L.

Exchange site [a]	**Concentration (mol/L)**	**Exchange site** [a]	**Concentration (mol/L)**
CaY_2	4.4E-05	MgY_2	4.5E-04
CaZ_2	4.9E-04	MgZ_2	5.0E-03
HQ	1.3E-12	NaQ	6.8E-05
HX	1.8E-10	NaX	9.5E-03
HY	1.3E-09	NaY	6.5E-02
HZ	1.4E-08	NaZ	7.1E-01
KQ	1.0E-05	NH_4Q	1.3E-04
KX	7.2E-04	NH_4X	6.3E-03
KY	1.0E-04	NH_4Y	1.1E-04
KZ	1.1E-03	NH_4Z	1.2E-03

[a] exchange sites are defined in Table 3.1.

4. Results of K_d setting for Horonobe mudstone systems

For each RNs (Cs, Ni, Am and Th), the results for K_d-values for mudstone systems from the Horonobe URL (Horonobe mudstone systems) are firstly presented based on the sorption model, followed by the results based on the semi-quantitative estimation method, and summary including their comparisons.

4.1 Cesium

4.1.1 Application of sorption models

As an alkaline element, Cesium exists only as Cs^+ ion in solution. Sorption takes place by ion exchange; surface complexation is of little or no relevance and is neglected in most TSMs for Cs. It is expected that sorption on Horonobe mudstone is influenced by both illite and smectite, therefore the respective models for these minerals as taken from the literature (see chapters 3.1 and Table 3.2) were combined and used simultaneously in one calculation.

- The sorption model developed by Wanner et al. (1996) was used to calculate the sorption of Cs on smectite. In line with common approaches in the literature and our previous papers (Ochs et al., 2001; Tachi et al., 2009, 2011b), Cs sorption on smectite is taken to occur as ion exchange reaction on only one exchange site (planar site, surface species CsZ).
- To calculate the sorption on illite, the model developed by Bradbury and Baeyens (2000) was used. According to their model, sorption on illite takes place on three different ion exchange sites; the frayed edge sites FES (surface species CsQ), the type-II sites (surface species CsX) and the planar sites (surface species CsY). The highest site capacity is provided by the planar sites (ca. 80% of the total CEC), while the selectivity for Cs increases in the order planar sites < type-II sites < FES.
- As shown in Table 3.2, competition by major cations for the different ion exchange sites needs to be taken into account for both smectite and illite. It should be noted in this context that the system was pre-equilibrated first in the absence of added RNs (as shown in Figure 3.1), so that the major cations present in the Horonobe groundwater could come to equilibrium with the ion exchange sites (Q-, X-, Y- on illite and Z-sites on smectite) on the clay surfaces.

The results of the sorption model application are discussed for each set of conditions (see cases defined in section 3.3) in the following sections.

i) Case I

The amounts of Cs sorbed onto the Q-, X-, Y- and Z-Sites in equilibrium for *Case I* are shown in Table 4.1, which corresponds to an overall K_d value of 0.12 m^3/kg for whole mudstone. The corresponding nominal K_d value for the mass of clay minerals alone (i.e., 19 % of the total solid mudstone mass) would be 0.64 m^3/kg.

Table 4.1 Sorbed amounts of Cs on the illite and smectite ion exchange sites for *Case I*. The S/L ratio is 4.12 kg/L. The indicated percentage is % of total Cs.

Species	**[mol/L]**	**%**
CsQ	7.6E-09	75
CsX	9.2E-10	9.1
CsY	1.3E-10	1.24
CsZ	1.5E-09	14.9

ii) Case II

The amounts of Cs sorbed onto illite and smectite in equilibrium for *Case II* are shown in Table 4.2, which corresponds to an overall K_d value of 0.12 m^3/kg for whole mudstone. The corresponding nominal K_d value for the mass of clay minerals alone (i.e., 19 % of the total solid mudstone mass) would be 0.65 m^3/kg.

Table 4.2 Sorbed amounts of Cs on the illite and smectite ion exchange sites for *Case II*. The S/L ratio is 4.12 kg/L. The indicated percentage is % of total Cs.

Species	**[mol/L]**	**%**
CsQ	7.6E-09	75
CsX	9.1E-10	9.0
CsY	1.3E-10	1.25
CsZ	1.5E-09	14.9

iii) Case III

The amounts of Cs sorbed at equilibrium for case III are shown in Table 4.3, which corresponds to an overall K_d value of 0.25 m^3/kg for whole mudstone. The corresponding nominal K_d value for the mass of clay minerals alone (i.e., 26 % of the total solid mudstone mass) would be 0.95 m^3/kg.

Table 4.3 Sorbed amounts of Cs on the illite and smectite ion exchange sites for *Case III*. The S/L ratio is 4.12 kg/L. The indicated percentage is % of total Cs.

Species	**[mol/L]**	**%**
CsQ	8.2E-09	81
CsX	8.8E-10	8.6
CsY	8.9E-11	0.87
CsZ	9.9E-10	9.7

4.1.2 Semi-quantitative K_d estimation

Figure 4.1 shows a compilation of several datasets for Cs(I) sorption on clay minerals. These sets were chosen initially because they are of good quality and cover a range of relevant conditions, which would allow to approach the Horonobe system from different directions.

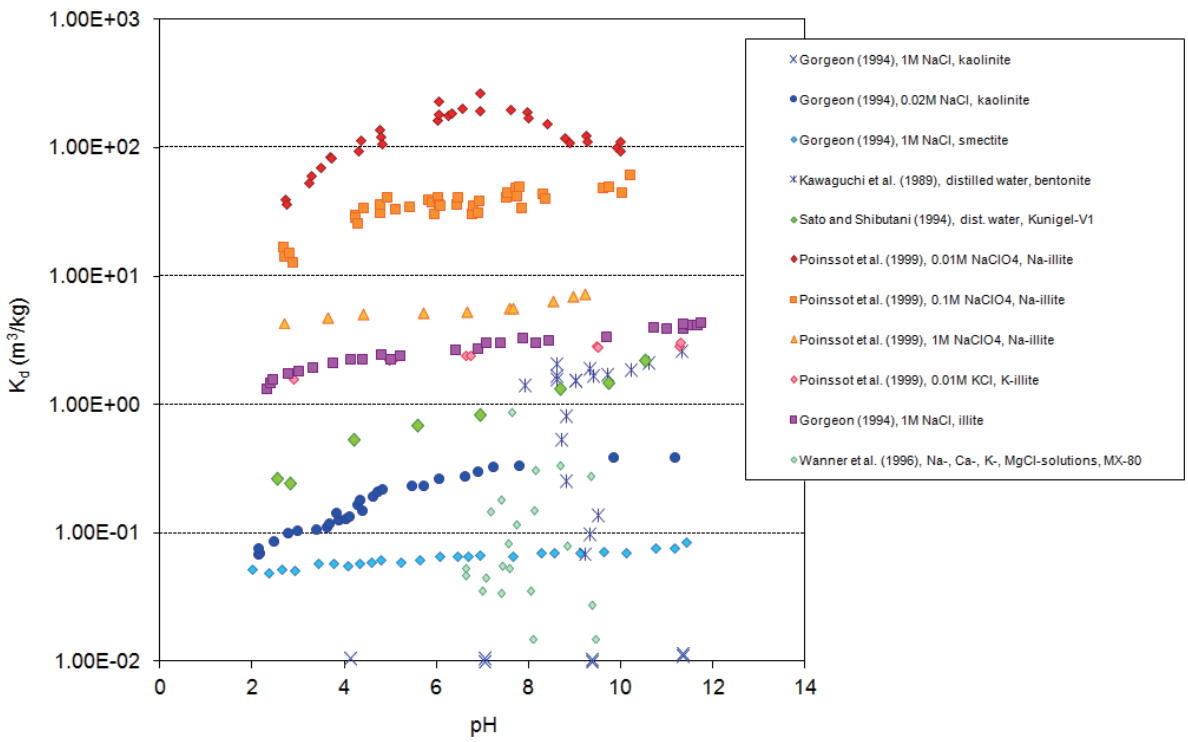

Figure 4.1 Cs(I) sorption data on clay minerals. All data are taken from the JAEA-SDB (Tachi et al., 2011a). Data for smectite and kaolinite are in blue/green colors, data for illite in reddish colors.

As can be expected, the data show that Cs sorption is almost independent of pH, but strongly influenced by the concentration of competing cations in solution. The slight pH dependence may arise from direct competition by H^+ and from the influence of pH on the speciation of competing cations, such as Ca and ammonium. Sorption on illite is higher than sorption on smectite minerals, mainly because of the frayed edge sites on the illite surface.

As in case of our previous report (Ochs et al., 2008), the estimation of K_d is based on the following primary data sources for smectite and illite:

- Cs sorption on Kunigel-V1 by Sato and Shibutani (1994);
- Cs sorption on Na-illite by Gorgeon (1994),
- Cs sorption on Na-illite by Poinssot et al (1999).

These three datasets were selected based on their similarity to the Horonobe mudstone system in terms of solution composition and because they include K_d values that were determined at (or very near) the pH of interest. As shown above, these datasets comprise similar K_d values that are lying in the intermediate range of the distribution.

The results of the estimation for *Case I-III* are shown in Table 4.4; details are given in the summary tables shown in Appendix I. Shown are the K_d values obtained using the two different conversion factors CF-1 and CF-3a (see Eq. (3.8) in chapter 3.2.1):

- The conversion factor CF-1 does not consider competition by major cations present in solution.
- On the other hand, CF-3 includes the competition by all major cations in solution. Two options are considered: with CF-3a, the competitive effect of all cations is taken to be equal; with CF-3b only the cations able to compete with Cs for the type-II sites and FES of illite are considered. CF-3b is not applicable to pure Na-electrolytes and questionable in solutions dominated by Na-cations.

It is admitted that it is difficult to decide which conversion factor should be used. The K_d values estimated on the basis of data for illite (Gorgeon, 1994; Poinssot et al., 1999) are relatively consistent when the conversion factor CF-3 is used, whereas the results using CF-1 are more variable and show a higher dependency with respect to the specific data source used. On the other hand, CF-3 seems to overestimate competition effects (especially by Na) in case of data from Kunigel-V1 (Sato and Shibutani, 1994). Note that in case of the source data by Poinssot et al. (1999), only the data obtained at 0.1 M are considered for Table 4.4. The data at lower ionic strength are less representative and cause much greater deviation between CF-1 and CF-3 (see Appendix I).

Based on the present calculations, the following approach is proposed for selecting recommended values:

- Considering that Cs sorption on Horonobe mudstone should be dominated by illite, based on mineralogy, the studies by Gorgeon (1994) and Poinssot et al. (1999) are considered more relevant for the present purpose.
- In terms of conversion factors, it is estimated that the use of CF-3 leads to smaller mistakes than the use of CF-1 when applied to substrates where Cs sorption can be assumed to be dominated by illite.

It is clearly not straightforward, however, to make appropriate corrections for cation competition, especially in case of Cs sorption on illite. As a result, it is difficult to recommend a best approach for all situations.

Table 4.4 K_d values (best estimates) for Cs for *Case I – III* obtained by semi-quantitative estimation. The results obtained by applying the correction factors CF-1 and CF-3a are reported. The factor CF-3b is not considered as applicable.

Case I

K_d: CF-1 [m^3/kg]	**K_d: CF-3a [m^3/kg]**	**Reference**
0.53	2.05	Gorgeon (1994), Na-illite
8.17	3.15	Poinssot et al. (1999), Na-illite, 0.1M
0.35	0.0014	Sato and Shibutani (1994), Kunigel V1

Case II

K_d: CF-1 [m^3/kg]	**K_d: CF-3a [m^3/kg]**	**Reference**
0.63	2.32	Gorgeon (1994), Na-illite
9.19	3.40	Poinssot et al. (1999), Na-illite, 0.1M
0.54	0.0021	Sato and Shibutani (1994), Kunigel V1

Case III

K_d: CF-1 [m^3/kg]	**K_d: CF-3a [m^3/kg]**	**Reference**
0.86	3.07	Gorgeon (1994), Na-illite
13.06	4.68	Poinssot et al. (1999), Na-illite, 0.1M
0.73	0.0028	Sato and Shibutani (1994), Kunigel V1

4.1.3 Summary for Cs

The following Table 4.5 summarizes the average values obtained with the semi-quantitative estimation procedures and the respective uncertainty expressed as upper/lower limiting values. The selection of data was done following the arguments presented in the previous section. The best estimate K_d values calculated using the smectite/illite ion exchange model are also summarized with the values estimated by the semi-quantitative procedures in Table 4.6 (no overall K_d error was calculated in case of Cs).

For all cases, the resulting K_d values deviate significantly and systematically between model results and semi-quantitative estimation, with the model giving lower values throughout. As already pointed out above, it is not clear which correction factor takes competition by major cations best into account, but all approaches appear to have shortcomings. It is therefore recommended to rely on the model-derived values.

No model uncertainties as defined for the other RN are available for Cs. If uncertainties are needed, it is recommended to calculate an overall TSM error in the same way as for the other RN (i.e., applying the models by Bradbury and Baeyens, 2000, and Wanner et al. 1996 to the corresponding experimental data and calculating the error according to NEA, 2005).

Table 4.5 Average values of recommended K_d for cesium obtained with (a) the semi-quantitative estimation procedure based on CF-3a and illite source data (Gorgeon, 1994; Poinssot et al., 1999).

Cs(I)	recommended K_d (m^3/kg)	upper limit K_d (m^3/kg)	lower limit K_d (m^3/kg)
Case I			
Gorgeon (1994), Na-illite	2.05	11.60	0.31
Poinssot et al. (1999), Na-illite, 0.1M	3.15	5.24	1.18
Average K_d values	**2.6**	**8.4**	**0.7**
Case II			
Gorgeon (1994), Na-illite	2.32	13.12	0.35
Poinssot et al. (1999), Na-illite, 0.1M	3.40	5.56	1.28
Average K_d values	**2.9**	**9.4**	**0.8**
Case III			
Gorgeon (1994), Na-illite	3.07	17.38	0.46
Poinssot et al. (1999), Na-illite, 0.1M	4.68	7.78	1.76
Average K_d values	**3.9**	**12.6**	**1.1**

Table 4.6 K_d values for cesium with their upper and lower estimates. K_d are given in m^3/kg. (no overall K_d error in TSM application was calculated in case of Cs).

Application		Case I	Case II	Case III
TSM	**K_d**	0.12	0.12	0.25
semi-quantitative estimation	**mean K_d**	2.6	2.9	3.9
	mean upper K_d	8.4	9.4	12.6
	mean lower K_d	0.7	0.8	1.1

4.2 Nickel

4.2.1 Application of sorption models

The JAEA11NEM_Ni model (JAEA, 2012) presented in chapter 3.1 is applied for the same sets of Horonobe mudstone conditions. It is based on the experimental sorption edge data by Bradbury and Baeyens (2005) and Tertre et al. (2005), as well as on the underlying clay-water model JAEA11NEM. Thermodynamic data for aqueous Ni species were taken from the JAEA-TDB (version 100331c1; Kitamura et al., 2010). This model considers several surface complexes and one ion exchange species (Table 3.1). Further testing was done to evaluate the model's ability to describe Ni sorption on Ca-montmorillonite (data by Bradbury and Baeyens, 1999).

The JAEA11NEM_Ni1 model for smectite is assumed to be able to describe Ni sorption on both smectite and illite, is then considered applicable to the Horonobe mudstone system. Sorption of Ni is expected to occur mostly by surface complexation at medium to higher pH, whereas sorption on ion

exchange sites is important at medium to lower pH. The same pre-equilibration step as applied in case of Cesium was performed. In this step, the various ion exchange sites of illite and smectite are equilibrated with the solution, so that the composition of all the exchange sites is in equilibrium with the aqueous phase.

i) Case I

The speciation of Ni in the equilibrium solution of the mudstone system considered is summarized in Table 4.7. The Ni-complexes that may be most important with regard to competing with surface sites for the available Ni are the Ni-chloride and the Ni-carbonate complexes. The corresponding concentrations of Ni sorbed in equilibrium on the surface complexation and ion exchange sites are shown in Table 4.8:

The predicted distribution of Ni in the mudstone-groundwater system of *Case I* yields a K_d value of 0.044 m^3/kg for the total mass of mudstone. Considering the overall error in K_d determined for the JAEA11NEM_Ni1 model, the corresponding maximum and minimum K_d values would be 0.083 m^3/kg and 0.023 m^3/kg. The (hypothetical) K_d value calculated for the clay minerals alone would amount to 0.23 m^3/kg. The overall errors in TSM were derived by direct comparison of modeled and experimental K_d values in our previous reports (Tachi et al., 2010; Ochs et al., 2011; JAEA, 2012), these values were directly used in this report.

Table 4.7 Solution speciation of Ni in the Horonobe mudstone system for *Case I*. The most important species are indicated in bold, irrelevant species are omitted. The total dissolved Ni concentration is about 7.2E-10 M (0.55 % of total Ni). The indicated percentage is % of dissolved Ni.

Species	[mol/L]	%	Species	[mol/L]	%	Species	[mol/L]	%
Ni^{2+}	**5.3E-10**	**73.1**	NiS	4.1E-13	0.1	$NiHPO_4$	2.5E-14	0.0
$NiHCO_3^+$	**1.4E-10**	**18.8**	$NiOH^+$	3.4E-13	0.0	$Ni(OH)_2$	4.3E-15	0.0
$NiCl^+$	**4.3E-11**	**6.0**	$NiNH_3^{2+}$	2.0E-13	0.0	$NiHS^+$	3.0E-15	0.0
$NiCO_3$	**1.5E-11**	**2.1**	NiF^+	4.4E-14	0.0			

Table 4.8 Sorbed amounts of Ni on the surface complexation and ion exchange sites assumed to be present in Horonobe mudstone. JAEA11NEM_Ni model, S/L ratio is 4.12 kg/L. The indicated percentage is % of total Ni.

Surface Species	Concentration [mol/L]	%
Y_2Ni	4.5E-10	0.3
Z_2Ni	5.4E-09	4.0
$>SONi^+$	1.3E-07	94.8
>SONiOH	1.0E-10	0.1
$>SONi(OH)_2^-$	7.8E-13	0.0
Ni(aq)	7.2E-10	0.5

ii) Case II

The speciation of Ni in the equilibrium solution of the mudstone system considered is given in Table 4.9. The corresponding concentrations of Ni sorbed at equilibrium on the surface complexation and ion exchange sites are shown in Table 4.10:

The predicted distribution of Ni in the mudstone-groundwater system of *Case II* yields a K_d value of 0.43 m^3/kg for the total mass of mudstone. Considering the overall error in K_d determined for the JAEA11NEM_Ni1 model, the corresponding maximum and minimum K_d values would be 0.82 m^3/kg and 0.23 m^3/kg. The (hypothetical) K_d value calculated for the clay minerals alone would amount to 2.3 m^3/kg.

Table 4.9 Solution speciation of Ni in the Horonobe mudstone system for *Case II*. The most important species are indicated in bold, irrelevant species are omitted. The total dissolved Ni concentration is about 7.3E-11 M (0.06 % of total Ni). The indicated percentage is % of dissolved Ni.

Species	**[mol/L]**	**%**	**Species**	**[mol/L]**	**%**	**Species**	**[mol/L]**	**%**
$NiCO_3$	**2.5E-11**	**34.6**	**$NiCl^+$**	**1.7E-12**	**2.3**	$NiHS^+$	2.5E-15	0.0
Ni^{2+}	**2.1E-11**	**28.9**	$NiOH^+$	7.7E-13	1.0	$Ni(OH)_3^-$	1.7E-15	0.0
NiS	**2.0E-11**	**26.6**	$Ni(OH)_2$	5.4E-13	0.7	$Ni(NH_3)_2^{2+}$	1.2E-15	0.0
$NiHCO_3^+$	**4.1E-12**	**5.5**	$NiNH_3^{2+}$	2.8E-13	0.4			

Table 4.10 Sorbed amounts of Ni on the surface complexation and ion exchange sites assumed to be present in Horonobe mudstone. JAEA11NEM_Ni1 model, S/L ratio is 4.12 kg/L. The indicated percentage is % of total Ni.

Surface Species	**Concentration [mol/L]**	**%**
Y_2Ni	1.9E-11	0.0
Z_2Ni	2.3E-10	0.2
$>SONi^+$	1.2E-07	93.7
>SONiOH	5.6E-09	4.2
$>SONi(OH)_2^-$	2.4E-09	1.8
Ni(aq)	7.3E-11	0.1

iii) Case III

The speciation of Ni in the equilibrium solution of the mudstone system considered is given in Table 4.11. The corresponding concentrations of Ni sorbed in equilibrium on the surface complexation and ion exchange sites are shown in Table 4.12:

The predicted distribution of Ni in the mudstone-groundwater system of *Case III* yields a K_d value of 0.62 m^3/kg for the total mass of mudstone. Considering the overall error in K_d determined for the JAEA11NEM_Ni1 model (JAEA, 2012), the corresponding maximum and minimum K_d values would be 1.18 m^3/kg and 0.33 m^3/kg. The (hypothetical) K_d value calculated for the clay minerals alone (27% of the mudstone) would amount to 2.4 m^3/kg.

Table 4.11 Solution speciation of Ni in the Horonobe mudstone system for *Case III*. The most important species are indicated in bold, irrelevant species are omitted. The total dissolved Ni concentration is about 5.1E-11 M (0.04 % of total Ni). The indicated percentage is % of dissolved Ni.

Species	[mol/L]	%	Species	[mol/L]	%	Species	[mol/L]	%
$NiCO_3$	**1.8E-11**	**35.8**	**$NiCl^+$**	**1.2E-12**	**2.3**	$NiHS^+$	1.6E-15	0.0
Ni^{2+}	**1.5E-11**	**29.4**	$NiOH^+$	5.4E-13	1.1	$Ni(OH)_3^-$	1.2E-15	0.0
NiS	**1.3E-11**	**24.6**	$Ni(OH)_2$	3.8E-13	0.7	$Ni(NH_3)_2^{2+}$	4.5E-16	0.0
$NiHCO_3^+$	**2.9E-12**	**5.7**	$NiNH_3^{2+}$	1.5E-13	0.3			

Table 4.12 Sorbed amounts of Ni on the surface complexation and ion exchange sites assumed to be present in Horonobe mudstone. JAEA11NEM_Ni1 model, S/L ratio is 4.12 kg/L. The indicated percentage is % of total Ni.

Surface Species	Concentration [mol/L]	%
Y_2Ni	1.8E-11	0.0
Z_2Ni	2.0E-10	0.2
$>SONi^+$	1.2E-07	93.7
>SONiOH	5.6E-09	4.2
$>SONi(OH)_2^-$	2.4E-09	1.8
Ni(aq)	5.1E-11	0.0

4.2.2 Semi-quantitative K_d estimation

In Figure 4.2, several relevant experimental datasets for Ni are shown. Sorption of Ni on clay minerals shows a clear tendency; sorption increases to a maximum at about pH 9 and decreases with even higher pH, which is typical for surface complexation and competition by hydrolysis. Based on the differences between data obtained at different ionic strength values, ion exchange appears to be important up to pH ≈ 8. As discussed in Ochs et al. (2011), there are some discrepancies between datasets by Bradbury and Baeyens (2005) on one hand and Tertre et al. (2005) on the other hand that cannot be resolved based on the conditions or the quality of the underlying experiments.

The estimation of a K_d value regarding the sorption of Ni on Horonobe mudstone is based on the following primary data sources:

- Ni sorption on MX-80 converted to the Na-form by Tertre et al. (2005);
- Ni sorption on a Ca-montmorillonite (converted from SWy-1) by Bradbury and Baeyens (1999);
- Ni sorption on Opalinus clay by Lauber et al. (2000).

It was assumed that the sorption of Ni does not differ significantly between smectite and illite, data sources were not therefore primarily chosen to include illite. Rather, high-quality datasets were selected that

- include K_d values that were determined at (or very near) the pH of interest;
- cover a range of solution as well as solid compositions from simple to complex;
- include a range similar to both Tertre et al. (2005) as well as Bradbury and Baeyens (2005) in terms of magnitude of sorption.

In addition, the experimental system with Opalinus clay by Lauber et al. (2000) has some similarity to the Horonobe mudstone system in terms of solution composition and the presence of both smectite and illite. The results of the estimation procedure are shown in Table 4.13 for *Case I-III.* Further details are given in the summary tables in Appendix I.

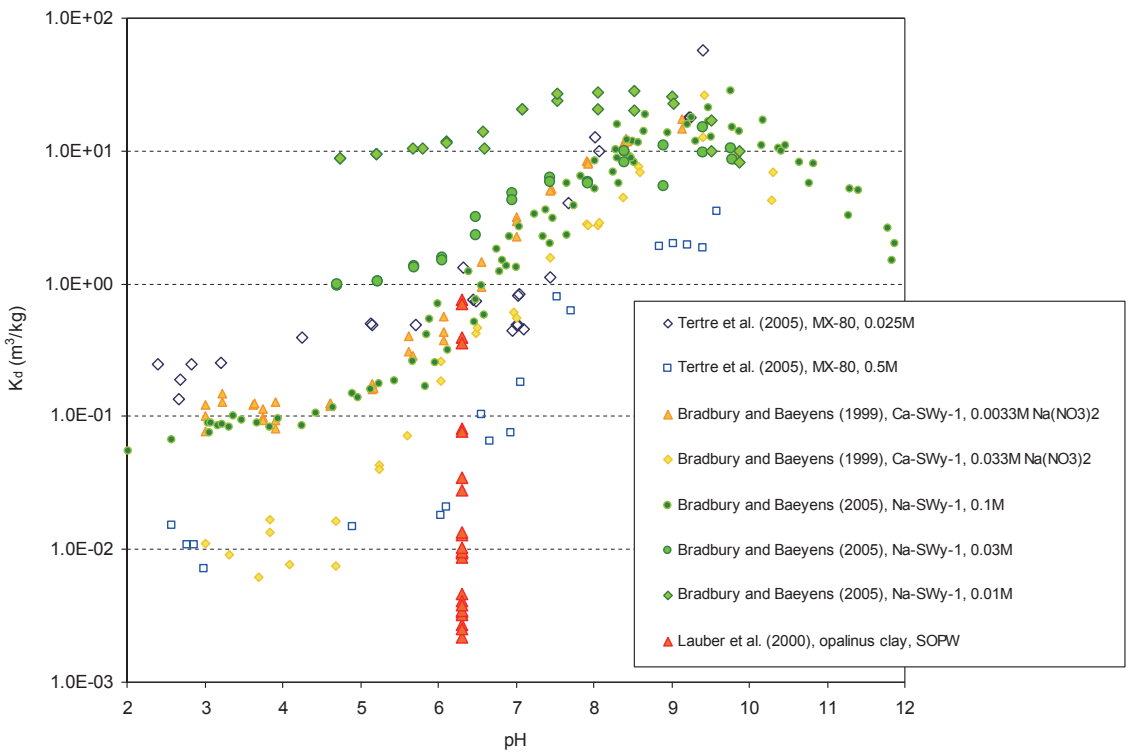

Figure 4.2 Overview of Ni(II) sorption data on smectite and clay rock. All data are taken from the JAEA-SDB (Tachi et al., 2011a).

Table 4.13 K_d values (best estimates) for Cs for *Case I – III* obtained by semi-quantitative estimation on the basis of different data sources.

Case I

K_d: CF-1 [m^3/kg]	**K_d: CF-2a [m^3/kg]**	**Reference**
0.057	0.042	Tertre et al. (2005)
0.15	0.11	Bradbury and Baeyens (1999)
0.14	0.15	Lauber et al. (2000)

Case II

K_d: CF-1 [m^3/kg]	**K_d: CF-2a [m^3/kg]**	**Reference**
0.86	0.26	Tertre et al. (2005)
1.07	0.33	Bradbury and Baeyens (1999)
1.37	0.62	Lauber et al. (2000)

Case III

K_d: CF-1 [m^3/kg]	**K_d: CF-2a [m^3/kg]**	**Reference**
1.22	0.38	Tertre et al. (2005)
1.52	0.47	Bradbury and Baeyens (1999)
1.94	0.89	Lauber et al. (2000)

The K_d values obtained using the two different conversion factors CF-1 and CF-2 are very similar for all cases, for any given reference. Due to the importance of hydrolysis for Ni-speciation in comparison to the effect of other ligands, the solution speciation of Ni is similar for the different experimental systems considered as data source and the Horonobe mudstone system. In all solutions, most of the Ni occurs as the Ni^{2+} ion. Therefore, the conversion factor for speciation effects (CF-spec, which is included in CF-2 but not in CF-1) and thus the difference between CF-1 and CF-2 is only of minor importance. In the solutions of Lauber et al. (2000), the complexed Ni is associated mainly with carbonate, chloride, and sulfate, whereas only hydrolysis is relevant in case of Tertre et al. (2005) and Bradbury and Baeyens (1999).

In the case of CF-Spec/a, the complexation of the radionuclide with carbonate is viewed as fully competitive with respect to sorption. The factor CF-Spec/b is not considered as relevant in case of Ni sorption, since there is no evidence for the existence of ternary carbonate complexes (dissolved or surface-bound) for Nickel.

It can be seen from Table 4.13 that the K_d values derived on the basis of source data from simple systems (Tertre et al., 2005; Bradbury and Baeyens, 1999) and on the basis of the more complicated system by Lauber et al. (2000) all lie within a relatively narrow range.

Table 4.13 further shows that higher K_d values result consistently with all data sources in case of synthetic groundwater (*Case II/III*) than in case of the in-situ groundwater (*Case I*). This is obviously also in agreement with the trend of sorption vs. pH shown in Figure 5.2.

The slightly different amounts of illite and smectite considered in the solid compositions of *Case II* vs. *Case III* also lead to slightly higher K_d values. However, the effect of solution type (especially pH) is more important in comparison. Overall, the agreement of the values derived on the basis of the different data sources is very good for all cases. Accordingly, it is recommended to use the averaged values (see the following section).

4.2.3 Summary for Ni

The following Table 4.14 gives an overview of the average values obtained with the semi-quantitative estimation procedures and the respective uncertainty expressed as upper/lower limiting values. Table 4.15 summarizes these average values as well as the K_d values calculated using the JAEA11NEM_Ni model. Shown are also the corresponding K_d uncertainties in each case.

It can be seen that both approaches (TSM and semi-quantitative estimation) give nearly identical results in terms of recommendable K_d. A comparison of the uncertainties shows that the assumptions made in the semi-quantitative estimation are more conservative.

Because of the similarity of the results, either approach could be used for K_d-setting. Because the coupling of sorption and geochemistry is implemented directly in a TSM, but is handled only by approximative calculations in the estimation procedure, it is generally recommendable to use a TSM (given that a well-tested model is available). For the present case, it is therefore recommended to use best estimate and upper limit as calculated with the TSM. For lower limits, it is recommended to conservatively use the values given by the estimation procedure.

Table 4.14 Average values of recommended K_d for nickel based on CF-2a and obtained with the semi-quantitative estimation procedure. K_d are given in m^3/kg.

Ni(II)	recommended K_d (CF-2a)	upper limit K_d (CF-2a)	lower limit K_d (CF-2a)
Case I			
Tertre (2005), MX-80	0.042	0.32	0.005
Bradbury and Baeyens (1999), Ca-mont.	0.11	0.86	0.014
Lauber (2000), OPA	0.15	1.13	0.019
Average K_d values	**0.10**	**0.77**	**0.013**
Case II			
Tertre (2005), MX-80	0.26	2..04	0.034
Bradbury and Baeyens (1999), Ca-mont.	0.33	2.54	0.042
Lauber (2000), OPA	0.62	4.78	0.079
Average K_d values	**0.40**	**3.12**	**0.052**
Case III			
Tertre (2005), MX-80	0.38	2.95	0.049
Bradbury and Baeyens (1999), Ca-mont.	0.47	3.68	0.061
Lauber (2000), OPA	0.89	6.93	0.115
Average K_d values	**0.58**	**4.52**	**0.075**

Table 4.15 K_d values with their upper and lower estimates. K_d are given in m^3/kg.

Application		Case I	Case II	Case III
TSM	**K_d**	**0.044**	**0.43**	**0.62**
	upper K_d	0.083	0.82	1.18
	lower K_d	0.023	0.23	0.33
semi-quantitative estimation	**mean K_d**	**0.10**	**0.40**	**0.58**
	mean upper K_d	0.77	3.12	4.52
	mean lower K_d	0.013	0.052	0.075

4.3 Americium

4.3.1 Application of sorption models

The JAEA11NEM_Am/Eu model (JAEA, 2012) presented in chapter 3.1 is applied for the same sets of Horonobe mudstone conditions. As basis for model parameterization for Am(III) sorption on montmorillonite, the datasets by Gorgeon (1994) providing the only sorption pH edges can be selected. On the other hand, trivalent lanthanides, especially Eu, are frequently taken as a chemical analogue for the trivalent actinides Am and Cm. For example, Rabung et al. (2005) indicated that the pH dependent Eu and Cm sorption onto the Ca-montmorillonite was mostly consistent. Sorption of Am on montmorillonite is therefore modeled assuming the chemical analogue between Am(III) and Eu(III).

Although some datasets for Eu sorption on montmorillonite can be found, datasets by Marques Fernandes et al. (2008), including carbonate effects can be selected as best choice in combination with datasets by Gorgeon (1994). Thermodynamic data for aqueous Am species were taken from the JAEA-TDB (version 100331c1). This model considers several surface complexes and one ion exchange species (Table 3.1).

i) Case I

The Am speciation in solution in equilibrium with the Horonobe mudstone system (i.e., after sorption equilibrium is established) calculated with the model JAEA11NEM_Am for *Case I* is shown in Table 4.16: The amounts of sorbed Americium on the SOH and Z-Sites in equilibrium are shown in Table 4.17:

Using the JAEA11NEM_Am model gives a K_d value for americium of 0.8 m^3/kg for the total amount of mudstone. Considering the overall error (see 4.2.1) in K_d determined for the JAEA11NEM_Am model, the corresponding maximum and minimum K_d values would be 1.8 m^3/kg and 0.4 m^3/kg. The hypothetical K_d value treating the clay mineral fraction in the mudstone as smectite would be 4.2 m^3/kg.

Table 4.16 Solution speciation of Am in the Horonobe mudstone system for Case I obtained with the model JAEA11NEM_Am. Total dissolved americium in solution is 6.0E-13 M (0.03 % of total Am). The indicated percentage is % of dissolved Am.

Species	**Concentration [mol/L]**	**%**	**Species**	**Concentration [mol/L]**	**%**
$AmCO_3^+$	**2.7E-13**	**45.3**	$Am(OH)_2^+$	1.0E-17	0.0
$AmSiO(OH)_3^{2+}$	**1.5E-13**	**24.8**	$AmCl_2^+$	2.2E-18	0.0
$Am(CO_3)_2^-$	**1.4E-13**	**23.4**	$AmH_2PO_4^{2+}$	4.6E-19	0.0
$AmHCO_3^{2+}$	**3.2E-14**	**5.4**	AmF_2^+	1.6E-20	0.0
Am^{3+}	4.8E-15	0.8	$Am(OH)_3$	3.2E-22	0.0
$Am(CO_3)_3^{3-}$	1.3E-15	0.2	$AmSO_4^+$	2.8E-22	0.0
$AmOH^{2+}$	3.7E-16	0.1	$Am(SO_4)_2^-$	2.3E-31	0.0
$AmCl^{2+}$	3.1E-16	0.1	$AmSCN^{2+}$	2.3E-35	0.0
AmF^{2+}	2.0E-17	0.0			

Table 4.17 Sorbed amounts of americium for Case I as obtained with the JAEA11NEM_Am model. The S/L ratio is 4.12 kg/L. The indicated percentage is % of total Am.

Species	Concentration [mol/L]	%
$>SOAmCO_3$	1.9E-09	92.0
$>SOAm^{2+}$	1.0E-10	5.1
$>SOAm(CO_3)_2^{2-}$	5.8E-11	2.9
$>SOAmOH^{+}$	7.0E-26	0.0
$>SOAm(OH)_2$	4.4E-42	0.0
$>SOAm(OH)_3^{-}$	4.6E-60	0.0
Y_3Am	5.7E-14	0.0
Z_3Am	6.8E-13	0.0
Am(aq)	6.0E-13	0.0

ii) Case II

The Am speciation in solution in equilibrium with the Horonobe mudstone system calculated with the model JAEA11NEM_Am for *Case II* is shown in Table 4.18; the corresponding concentrations of Americium surface species at equilibrium are shown in Table 4.19:

Table 4.18 Solution speciation of Am in the Horonobe mudstone system for *Case II* obtained with the model JAEA11NEM_Am. Total dissolved Am in solution is 1.7E-13 M (0.008 % of total Am). The indicated percentage is % of dissolved Am.

Species	Concentration [mol/L]	%	Species	Concentration [mol/L]	%
$Am(CO_3)_2^{-}$	**1.139E-13**	**67.9**	Am^{3+}	2.23E-18	0.0
$Am(CO_3)_3^{3-}$	**4.461E-14**	**26.6**	$AmCl^{2+}$	1.378E-19	0.0
$AmCO_3^{+}$	5.282E-15	3.1	$Am(OH)_3$	2.623E-20	0.0
$AmSiO(OH)_3^{2+}$	3.877E-15	2.3	$AmCl_2^{+}$	9.44E-22	0.0
$Am(OH)_2^{+}$	1.518E-17	0.0	$AmSO_4^{+}$	1.136E-26	0.0
$AmHCO_3^{2+}$	1.119E-17	0.0	$Am(SO_4)_2^{-}$	8.345E-37	0.0
$AmOH^{2+}$	9.603E-18	0.0	$AmSCN^{2+}$	1.041E-37	0.0

Table 4.19 Sorbed amounts of americium for Case II obtained with the JAEA11NEM_Am model. The S/L ratio is 4.12 kg/L. The indicated percentage is % of total Am.

Species	Concentration [mol/L]	%
$>SOAm(CO_3)_2^{2-}$	1.1E-09	56.5
$>SOAmCO_3$	8.8E-10	43.5
$>SOAm^{2+}$	1.2E-12	0.1
$>SOAmOH^{+}$	4.4E-26	0.0
$>SOAm(OH)_2$	1.6E-40	0.0
$>SOAm(OH)_3^{-}$	9.1E-57	0.0
Y_3Am	2.8E-17	0.0
Z_3Am	3.3E-16	0.0
Am(aq)	1.7E-13	0.0

Using the JAEA11NEM_Am model, the estimated K_d value for americium was 2.9 m^3/kg for the total amount of mudstone. Considering the overall error in K_d, the corresponding maximum and minimum K_d values would be 6.6 m^3/kg and 1.3 m^3/kg. The hypothetical K_d value for the clay mineral fraction in the mudstone alone would be 15.2 m^3/kg.

iii) Case III

The Am speciation in solution in equilibrium with the Horonobe mudstone system calculated with the model JAEA11NEM_Am for *Case III* is shown in Table 4.20: The amount of sorbed Americium on the surface sites in equilibrium is shown in Table 4.21.

Using the JAEA11NEM_Am model, the estimated K_d value for americium was 7.2 m^3/kg for the total amount of mudstone. Considering the overall error in K_d, the corresponding maximum and minimum K_d values would be 16.4 m^3/kg and 3.1 m^3/kg. The hypothetical K_d value considering the clay mineral fraction in the mudstone alone would be 26.5 m^3/kg.

Table 4.20 Solution speciation of Am in the Horonobe mudstone system for Case III obtained with the model JAEA11NEM_Am. Total dissolved Am in solution is 6.8E-14 M (0.003 % of total Am). The indicated percentage is % of dissolved Am.

Species	Concentration [mol/L]	%	Species	Concentration [mol/L]	%
$Am(CO_3)_2^{-}$	**4.6E-14**	**67.5**	Am^{3+}	9.0E-19	0.0
$Am(CO_3)_3^{3-}$	**1.8E-14**	**27.0**	$AmCl^{2+}$	5.5E-20	0.0
$AmCO_3^{+}$	2.1E-15	3.1	$Am(OH)_3$	1.0E-20	0.0
$AmSiO(OH)_3^{2+}$	1.6E-15	2.3	$AmCl_2^{+}$	3.7E-22	0.0
$Am(OH)_2^{+}$	6.0E-18	0.0	$AmSO_4^{+}$	8.2E-27	0.0
$AmHCO_3^{2+}$	4.5E-18	0.0	$Am(SO_4)_2^{-}$	1.1E-36	0.0
$AmOH^{2+}$	3.9E-18	0.0	$AmSCN^{2+}$	3.0E-38	0.0

Table 4.21 Sorbed amounts of americium for Case III obtained with the JAEA11NEM_Am model. The S/L ratio is 4.12 kg/L. The indicated percentage is % of total Am.

Species	Concentration [mol/L]	%
$>SOAm(CO_3)_2^{2-}$	1.1E-09	56.6
$>SOAmCO_3$	8.8E-10	43.4
$>SOAm^{2+}$	1.2E-12	0.1
$>SOAmOH^{+}$	4.4E-26	0.0
$>SOAm(OH)_2$	1.5E-40	0.0
$>SOAm(OH)_3^{-}$	9.0E-57	0.0
Y_3Am	1.2E-17	0.0
Z_3Am	1.3E-16	0.0
Am(aq)	6.8E-14	0.0

4.3.2 Semi-quantitative K_d estimation

Figure 5.3 shows Am(III) and Eu(III) sorption data obtained on different clays and clay minerals. Considering the difference in conditions, the data for both Am and Eu lie within a relatively narrow band, but there may be different trends for smectite vs. illite. Based on the similar sorption behavior of Am and Eu (as already considered for the development of the corresponding sorption model), the sorption data of these elements are also considered as comparable for the present purpose.

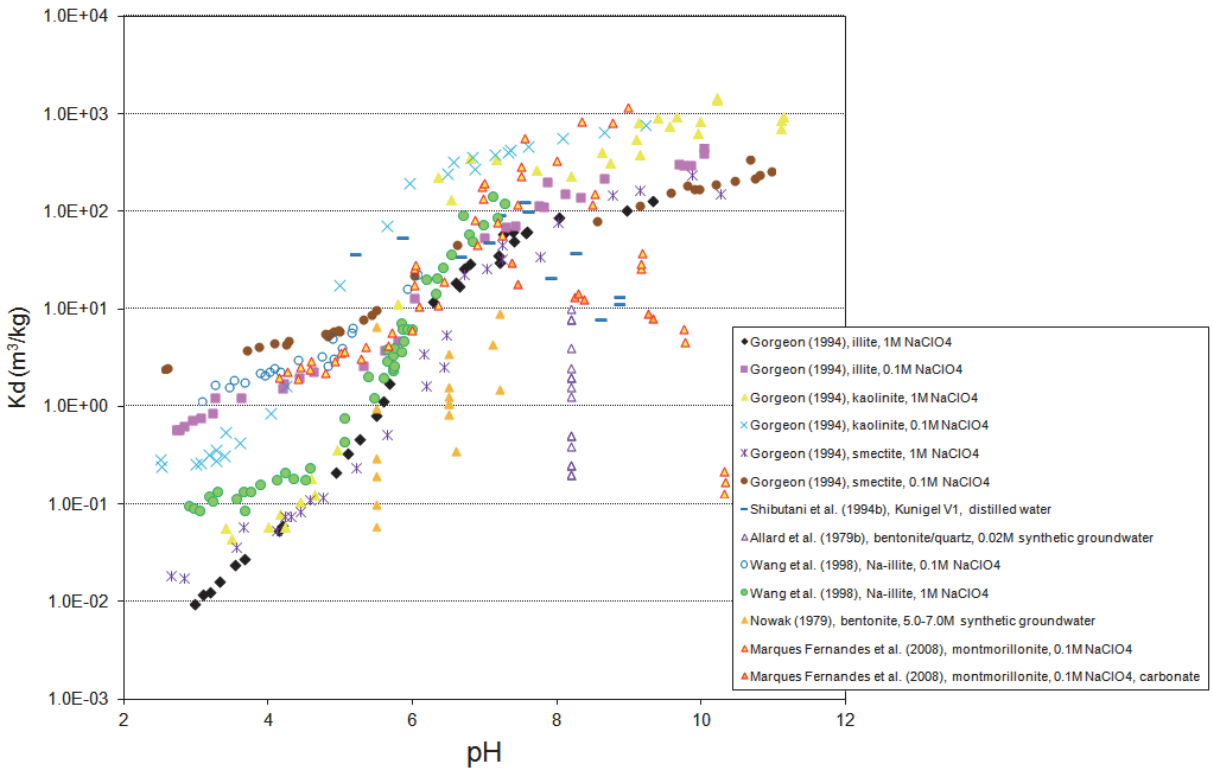

Figure 4.3 Overview of Am(III) and Eu(III) sorption data on clays and clay minerals. Sorption data are taken from the JAEA-SDB (Tachi et al., 2011a)

The K_d-estimation for Americium was conducted using the following datasets:

- Am sorption on Illite by Gorgeon (1994);
- Eu sorption on Opalinus clay by Lauber et al. (2000);
- The data for Am sorption on montmorillonite by Nagasaki et al. (1997) were not considered for the present data derivation, because the transfer of data from the low pH used by Nagasaki et al. (1997) to the Horonobe conditions does not yield reasonable results.

As the sorption behavior of Am may differ between smectite and illite, and because the JAEA11NEM_Am model is based on data for smectite, the above datasets were selected to include pure illite as well as a substrate containing both smectite and illite. The dataset by Lauber et al. (2000) is similar to the Horonobe mudstone system also in terms of solution composition.

The results of the estimation are shown in Table 4.22 for Am and Eu. Details are given in the summary tables in Appendix I. Note that the speciation calculations for Eu were carried out using the thermodynamic data given in Hummel et al. (2002) in lack of corresponding data in the JAEA-TDB.

For each scenario and RN, the data derived using the conversion factors CF-1 and CF-2b agree reasonably well with each other, while the values based on CF-2a are significantly lower. The fact that the CF-2a based values decrease from pH 6.75 to pH 8.5 means that this speciation correction factor is overpredicting the competitive effect of carbonate complexation in solution. This may be expected,

considering that the JAEA11NEM_Am model requires including a ternary Am-carbonato-surface complex.

For *Case I*, the values derived for Am and Eu are in close agreement. However, they deviate strongly under the conditions of *Case II* and *Case III* with the Eu-values increasing to very high numbers. These values result from the extensive pH correction factor needed to transfer the data from pH 6.3 (experimental conditions) to pH 8.5 (groundwater) are considered to be not realistic.

Overall, the values derived for Am using the factor CF-2b are viewed as most realistic. Therefore, no averaging of estimated values is done.

Table 4.22 K_d values for americium and europium obtained by semi-quantitative estimation for *Case I-III*.

Case I

K_d: CF-1 [m3/kg]	K_d: CF-2a [m3/kg]	K_d: CF-2b [m^3/kg]	RN / Reference
5.51	0.05	4.14	Am / Gorgeon (1994)
5.79	0.84	8.63	Eu / Lauber et al. (2000)

Case II

K_d: CF-1 [m3/kg]	K_d: CF-2a [m3/kg]	K_d: CF-2b [m^3/kg]	Reference
20.0	0.002	19.49	Am / Gorgeon (1994)
258.54	6.78	392.43	Eu / Lauber et al. (2000)

Case III

K_d: CF-1 [m3/kg]	K_d: CF-2a [m3/kg]	K_d: CF-2b [m^3/kg]	Reference
28.4	0.003	27.70	Am / Gorgeon (1994)
367.40	8.13	555.23	Eu / Lauber et al. (2000)

4.3.2 Summary for Am

The following Table 4.23 summarizes the K_d values calculated using the JAEA11NEM_Am model as well as the values obtained with the semi-quantitative estimation procedure (based on CF-2b) for Am. The respective K_d uncertainties are also shown. The estimated K_d values using the semi-quantitative estimation procedure are in general slightly higher than the model-derived values, but the agreement is reasonably good overall. This is consistent with the respective treatment of the effect of carbonate: CF-spec/b essentially treats the formation of Am-carbonate complexes as having no net effect on sorption, and the JAEA11NEM_Am model includes a ternary surface Am-carbonate complex.

In cases where TSM application and semi-quantitative estimation arrive at similar results, preference should generally be given to the model-derived data. A TSM allows a direct coupling of geochemistry and sorption reactions, whereas this is handled only approximatively in the estimation approach. Also, the speciation conversion factors are somewhat speculative, as they allow only for full or no competitive effect by the formation of aqueous RN complexes. Therefore, the model-derived values are recommended.

Table 4.23 K_d values for Am with the respective upper and lower estimates. All K_d values are given in m^3/kg.

Application		Case I	Case II	Case III
TSM	**K_d**	0.8	2.9	7.2
	upper K_d	1.8	6.6	16.4
	lower K_d	0.4	1.3	3.1
semi-quantitative estimation	**K_d**	4.14	19.49	27.70
	upper K_d	12.86	60.54	86.05
	lower K_d	1.33	6.27	8.92

4.4 Thorium

4.4.1 Application of sorption models

The JAEA11NEM_Th model (JAEA, 2012) presented in chapter 3.1 was applied to the Horonobe mudstone system in the same way as described for the other RN in the previous sections. As basis for model parameterization for Th sorption on montmorillonite, the datasets by Bradbury and Baeyens (2003a, 2005) providing the only sorption pH edges can be selected. Due to the lack of suitable sorption data of Th in the presence of carbonate, the ternary carbonate surface species cannot be included in the present sorption model. The same aqueous thermodynamic data (JAEA-TDB) were used as in case of the model parameterization. The system was first pre-equilibrated in a first model run without introducing Th, so that the cations in solution could equilibrate with the ion exchange sites on the clay surface. The occupation of the Y-sites of the illite and the Z-sites of the smectite after the pre-equilibration step are given in Table 3.6 for *Case I*, Table 3.7 for *Case II*, and Table 3.8 for *Case III*. After the pre-equilibration step, 1.00E-08 M Th was added to the system. The S/L ratio was 4.12 kg/L. The resulting speciation of dissolved Th for *Cases I* to *III* and the derived K_d values are discussed in the three following short sections.

i) Case I

The speciation of Th in the equilibrium solution of the mudstone system considered is summarized in Table 4.24. The Th-complexes that may be most important with regard to competing with surface sites for the available Th are mixed carbonato/hydroxo-complexes. The corresponding concentrations of Th sorbed at equilibrium on the surface complexation and ion exchange sites are shown in Table 4.25.

The predicted distribution of Th in the mudstone-groundwater system of *Case I* yields a K_d value of 0.10 m^3/kg for the total mass of mudstone. Considering the overall error (see 4.2.1) in K_d determined for the JAEA11NEM_Th model, the corresponding maximum and minimum K_d values would be 0.27 m^3/kg and 0.04 m^3/kg. The (hypothetical) K_d value calculated for the clay minerals alone would amount to 0.53 m^3/kg.

Table 4.24 Solution speciation of Th in the Horonobe mudstone system for *Case I*. The most important species are indicated in bold, irrelevant species are omitted. The equilibrium Th concentration in solution is 2.4E-11 M (0.24 % of total Th). The indicated percentage is % of dissolved Th.

Species	**Concentration [mol/L]**	**%**	**Species**	**Concentration [mol/L]**	**%**
$Th(CO_3)_2(OH)_2^{2-}$	**2.342E-11**	**96.8**	ThF_3^+	2.378E-18	0.0
$Th(CO_3)_4OH^{5-}$	6.849E-13	2.8	$ThOH^{3+}$	2.292E-18	0.0
$Th(CO_3)_5^{6-}$	5.774E-14	0.2	ThF_4	1.028E-18	0.0
$Th(OH)_4$	3.045E-14	0.1	ThF_3^+	7.368E-19	0.0
$Th(OH)_2^{2+}$	5.557E-16	0.0	$Th(OH)_3(H_3SiO_4)_3^{2-}$	2.483E-19	0.0
$ThCO_3(OH)_4^{2-}$	4.459E-17	0.0	$ThCl^{3+}$	1.102E-21	0.0
ThF_2^{2+}	7.066E-18	0.0	Th^{4+}	1.099E-21	0.0

Table 4.25 Sorbed amounts of thorium for *Case I*. S/W is 4.12 kg/L. The indicated percentage is % of total Th.

Species	**[mol/L]**	**%**
$>SOTh(OH)_3$	9.8E-09	96.1
$>SOTh(OH)_4^-$	3.5E-10	3.5
$>SOTh^{3+}$	1.6E-11	0.2
Y_4Th	4.0E-20	0.0
Z_4Th	4.8E-19	0.0
Th(aq)	2.4E-11	0.2

ii) Case II

The speciation of Th in the equilibrium solution of the mudstone system considered is given in Table 4.26. The corresponding concentrations of Th sorbed in equilibrium on the surface complexation and ion exchange sites are shown in Table 4.27. Due to the difference in pH between *Case I* and *Case II / III* and the strong hydrolysis of Th, the Z_4Th species is not relevant anymore for overall sorption at the higher pH of *Case II / III*.

Table 4.26 Solution speciation of Th in the Horonobe mudstone system *(Case II).* The most important species are indicated in bold, irrelevant species are omitted. Total dissolved Th concentration at equilibrium is about 2.0E-11 M (0.20 % of total Th). The indicated percentage is % of dissolved Th.

Species	[mol/L]	%	Species	[mol/L]	%
$Th(CO_3)_2(OH)_2^{2-}$	**1.0E-11**	**51.2**	$Th(OH)_3(H_3SiO_4)_3^{2-}$	6.1E-16	0.0
$Th(CO_3)_4OH^{5-}$	**9.2E-12**	**45.9**	$Th(OH)_2^{2+}$	1.4E-19	0.0
$Th(CO_3)_5^{6-}$	5.7E-13	2.9	$ThOH^{3+}$	1.0E-23	0.0
$Th(OH)_4$	2.4E-14	0.1	Th^{4+}	8.7E-29	0.0
$ThCO_3(OH)_4^{2-}$	1.5E-15	0.0			

Table 4.27 Sorbed amounts of Th on the surface complexation and ion exchange sites assumed to be present in Horonobe mudstone. JAEA11NEM_Th model, S/L ratio is 4.12 kg/L. The indicated percentage is % of total Th.

Surface Species	Concentration [mol/L]	%
$>SOTh(OH)_4^-$	6.8E-09	66.8
$>SOTh(OH)_3$	3.4E-09	33.0
$>SOTh^{3+}$	3.2E-17	0.0
Y_4Th	3.5E-27	0.0
Z_4Th	4.1E-26	0.0
Th(aq)	2.0E-11	0.2

The predicted distribution of Th in the mudstone-groundwater system of *Case II* yields a K_d value of 0.12 m^3/kg for the total mass of mudstone. Considering the overall error in K_d determined for the Th sorption model, the corresponding maximum and minimum K_d values would be 0.33 m^3/kg and 0.05 m^3/kg. The (hypothetical) K_d value calculated for the clay minerals alone would amount to 0.64 m^3/kg.

iii) Case III

The speciation of Th in the equilibrium solution of the mudstone system considered is given in Table 4.28. The corresponding concentrations of Th sorbed in equilibrium on the surface complexation and ion exchange sites are shown in Table 4.29. As expected, the species distribution is similar as in *Case II*.

Table 4.28 Solution speciation of Th in the Horonobe mudstone system *(Case III).* The most important species are indicated in bold, irrelevant species are omitted. Total dissolved Th concentration at equilibrium is about 1.5E-11 M (0.15 % of total Th). The indicated percentage is % of dissolved Th.

Species	[mol/L]	%	Species	[mol/L]	%
$Th(CO_3)_2(OH)_2^{2-}$	**7.5E-12**	**49.8**	$Th(OH)_3(H_3SiO_4)_3^{2-}$	4.3E-16	0.0
$Th(CO_3)_4OH^{5-}$	**7.1E-12**	**47.1**	$Th(OH)_2^{2+}$	9.8E-20	0.0
$Th(CO_3)_5^{6-}$	4.5E-13	3.0	$ThOH^{3+}$	7.2E-24	0.0
$Th(OH)_4$	1.7E-14	0.1	Th^{4+}	6.2E-29	0.0
$ThCO_3(OH)_4^{2-}$	1.1E-15	0.0			

Table 4.29 Sorbed amounts of Th on the surface complexation and ion exchange sites assumed to be present in Horonobe mudstone. JAEA11NEM_Th model, S/L ratio is 4.12 kg/L. The indicated percentage is % of total Th.

Surface Species	Concentration [mol/L]	%
$>SOTh(OH)_4^-$	6.8E-09	66.8
$>SOTh(OH)_3$	3.4E-09	33.0
$>SOTh^{3+}$	3.2E-17	0.0
Y_4Th	3.2E-27	0.0
Z_4Th	3.5E-26	0.0
Th(aq)	1.5E-11	0.1

The predicted distribution of Th in the mudstone-groundwater system of *Case III* yields a K_d value of 0.16 m^3/kg for the total mass of mudstone. Considering the overall error in K_d determined for the JAEA11NEM_Th model, the corresponding maximum and minimum K_d values would be 0.43 m^3/kg and 0.06 m^3/kg. The (hypothetical) K_d value calculated for the clay minerals alone (26% of the mudstone) would amount to 0.62 m^3/kg.

4.4.2 Semi-quantitative K_d estimation

In Figure 4.4 an overview of available Th(IV) sorption data is shown. The cement water used by Baston et al. (1992) in a sorption study on London clay contains a large amount of carbonate in solution. The purified SWy-1 montmorillonite used by Bradbury and Baeyens (2003a, 2005) shows very strong sorption and very high K_d values. The indicated isotherm experiments by Bradbury and Baeyens (2003a, 2003b) were conducted with Opalinus Clay and MX-80 and give K_d values between London Clay and the SWy-1 montmorillonite. On the basis of these data, it appears that the cement water/London Clay system may not be representative for the present purpose.

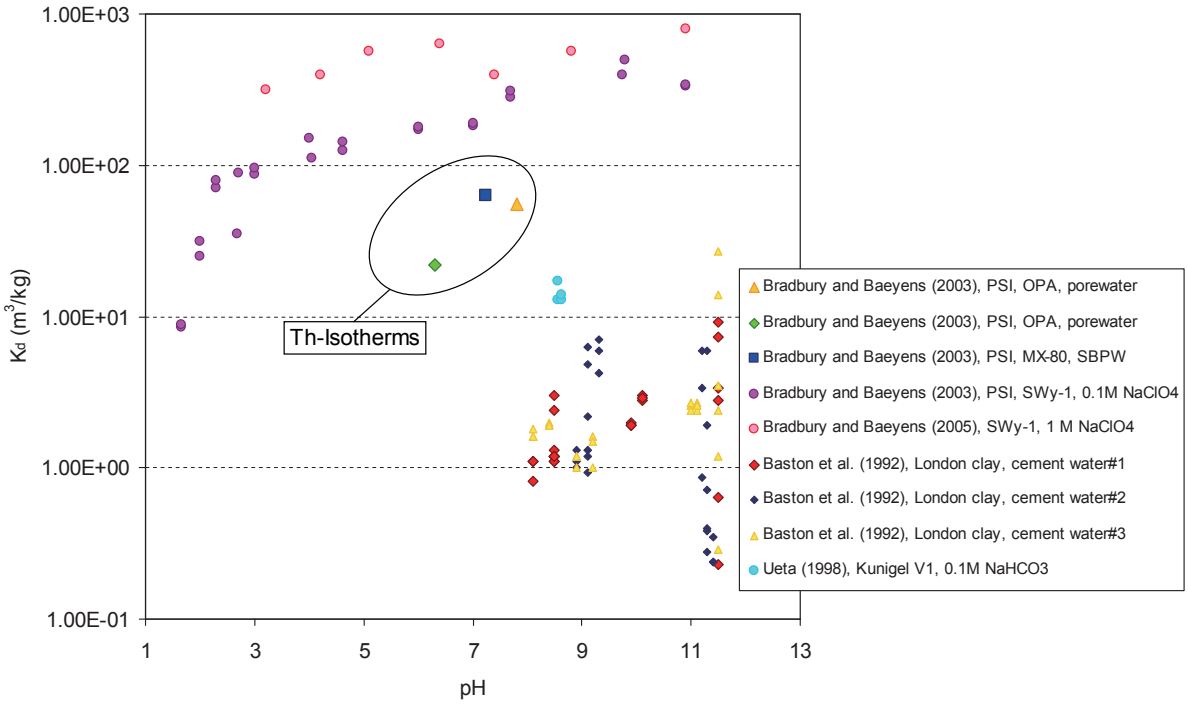

Figure 4.4 Overview of Th(IV) sorption data on clay minerals and clay rock. Sorption data are taken from the JAEA-SDB (Tachi et al., 2011)

Because the sorption of Th is not strongly dependent on pH, the isotherm data by Bradbury and Baeyens (2003a, 2003b) were preferred for K_d-estimation. While these datasets are restricted to a given pH, each value is underpinned by a range of data obtained at different Th concentrations. Moreover, the respective substrates and solutions used in these studies are comparatively similar to the Horonobe system. Accordingly, the semi-quantitative K_d-estimation for Thorium was conducted using the following datasets:

- Th sorption on Opalinus clay by Bradbury and Baeyens (2003b) at pH 6.3 and 7.8;
- Th sorption on MX-80 by Bradbury and Baeyens (2003a).

The results of the K_d estimation for *Case I-III* are summarized in the following Table 4.30. Further details are given in Appendix I.

Table 4.30 Results of the K_d estimation for Th under the conditions of *Case I-III*. Results of the derivation using only CF-1 (not shown) are nearly identical to those using CF-2b.

Case I

K_d: CF-2a [m^3/kg]	K_d: CF-2b [m^3/kg]	Reference
0.02	9.56	Bradbury and Baeyens (2003b), OPA, pH 7.8
0.64	9.51	Bradbury and Baeyens (2003b), OPA, pH 6.3
0.11	14.75	Bradbury and Baeyens (2003a), MX-80

Case II

K_d: CF-2a [m^3/kg]	K_d: CF-2b [m^3/kg]	Reference
0.01	19.11	Bradbury and Baeyens (2003b), OPA, pH 7.8
0.63	19.02	Bradbury and Baeyens (2003b), OPA, pH 6.3
0.10	29.13	Bradbury and Baeyens (2003a), MX-80

Case III

K_d: CF-2a [m^3/kg]	K_d: CF-2b [m^3/kg]	Reference
0.03	26.16	Bradbury and Baeyens (2003b), OPA, pH 7.8
1.25	26.02	Bradbury and Baeyens (2003b), OPA, pH 6.3
0.20	39.86	Bradbury and Baeyens (2003a), MX-80

In both the estimation procedure using CF-2a and the TSM application, the formation of dissolved Th-carbonate complexes is viewed as competitive with regard to sorption. Specifically, the formation of ternary carbonato-hydroxo complexes for Thorium in solution is not balanced by the formation of analogous surface complexes.

The K_d values based on CF-2b are very similar for all cases and for all data sources. Therefore, the average values are recommended.

4.4.3 Summary for Th

The following Table 4.31 gives an overview of the average values obtained with the semi-quantitative estimation procedures and the respective uncertainty expressed as upper/lower limit values. Table 4.32 summarizes the K_d values calculated using the JAEA11NEM_Th model as well as the values obtained with the semi-quantitative estimation procedure. Shown are also the model K_d uncertainties which were calculated using the overall K_d error obtained in the development of the JAEA11NEM_Th model.

Table 4.31 Average values of recommended K_d obtained with the semi-quantitative estimation procedure using CF-2b. K_d values are given in m^3/kg.

Th(IV)	recommended K_d (CF-2b)	upper limit K_d (CF-2b)	lower limit K_d (CF-2b)
Case I			
Bradbury and Baeyens (2003b), OPA, pH 7.8	9.56	74.22	1.23
Bradbury and Baeyens (2003b), OPA, pH 6.3	9.51	73.84	1.22
Bradbury and Baeyens (2003a), MX-80	14.57	113.12	1.88
Average K_d values	**11.2**	**87.1**	**1.4**
Case II			
Bradbury and Baeyens (2003b), OPA, pH 7.8	19.11	147.67	2.45
Bradbury and Baeyens (2003b), OPA, pH 6.3	19.02	73	1.2
Bradbury and Baeyens (2003a), MX-80	29.13	226.23	3.75
Average Kd values	**22.4**	**149.0**	**2.5**
Case III			
Bradbury and Baeyens (2003b), OPA, pH 7.8	26.16	203.13	3.37
Bradbury and Baeyens (2003b), OPA, pH 6.3	26.02	202.08	3.35
Bradbury and Baeyens (2003a), MX-80	39.86	309.59	5.13
Average K_d values	**30.7**	**238.3**	**4.0**

Table 4.32 K_d values derived through sorption modeling and estimation procedures (based on CF-2b) with upper and lower estimates. All K_d values are given in m^3/kg.

Application		Case I	Case II	Case III
TSM	**K_d**	0.10	0.12	0.16
	upper K_d	0.27	0.33	0.43
	lower K_d	0.04	0.05	0.06
semi-quantitative estimation	**mean K_d**	11.2	22.4	30.7
	mean upper K_d	87.1	149.0	238.3
	mean lower K_d	1.4	2.5	4.0

The model calculations predict a surprisingly low sorption of Th. Even lower values result from applying the estimation procedure based on CF-2a. The reason is in both cases the strong competition mainly by mixed Th carbonato-hydroxo complexes in solution. These are predicted to dominate the aqueous Th speciation, and by analogy, the existence of corresponding ternary surface complexes must be assumed. The existence of such surface species is further considered likely in view of the proven existence of such species for +III- and +V-valent actinides. Due to the lack of suitable data for model development, the present sorption model does not include such complexes, however. Similarly, the conversion procedure based on CF-2a considers sorption competition by various aqueous Th-carbonate species that is not balanced by similar surface species. At the other extreme, the conversion procedure based on CF-2b assumes that the effects of aqueous and surface-bound Th-carbonate species balance each other out

completely. While this is obviously a simplication, this latter approach gives much more reasonable results than either CF-2a or the TSM application and is recommended for data setting.

It has to be concluded, therefore, that the present sorption model for Th underestimates K_d in the presence of significant carbonate levels. The calibration of the sorption model to include ternary surface species is recommended but presently not possible due to the lack of systematic sorption data obtained in the presence of carbonate.

5. Outlooks for extension of K_d setting to other rock types

5.1 Extension of K_d setting to granitic rock systems

In comparison to clay systems, the present state of the art in deriving K_d for granitic rocks is clearly less developed, and there is less consensus regarding the most appropriate approaches. In the following, some of the most actively followed approaches are briefly documented.

The general relevance of radionuclide migration in the context of radioactive waste disposal is discussed in chapter 2.1. In chapter 2.2, the general steps required for the derivation of a K_d value for a reference condition (definition of reference in-situ conditions and actual data derivation) and the associated uncertainties are introduced. These principal steps are the same in case of K_d derivation for rocks, but the major difficulties lie with different aspects than in the case of clay systems.

5.1.1 Principal challenges in the definition of K_d values for granitic rocks

From a mineralogical and surface chemical point of view, the estimation of sorption values should not be dramatically different for most types of granitic rocks. The main minerals in these materials are quartz, feldspars, and micas. As pointed out in an earlier report (Lothenbach and Ochs, 1999), the mica minerals can be assumed to offer the largest surface area and the most reactive types of surfaces, based on their mineral structure.

Due to the similarity of mica and clay minerals, principally the same estimation procedures or sorption model types can be used for both minerals. Thus, no additional difficulties in terms of quantifying the actual sorption processes are expected in this sense. It follows that the models used to describe sorption on clays should be easily adapted to address sorption on crushed and dispersed fresh rock material. First modeling trials (see Lothenbach and Ochs, 1999) indicate that this assumption is correct. However, there are significant additional difficulties in the definition of sorption for actual real rock material. These are discussed in NEA (2012) and briefly outlined below:

- First, crystalline rocks consist of primary (rock-forming) minerals which are not stable under ambient conditions. Upon weathering, and depending on the specific environmental conditions, these minerals are transformed into secondary minerals (such as clay-like minerals and metal oxides).
    - Depending on the location of such weathering products (see next point), they may coat the original rock mineral surface (partly or completely), thereby altering the sorption properties.
    - Weathering products are often a complex mixture of minerals that all contribute to sorption and need to be considered. As weathering products are often also difficult to sample and therefore to characterize, their possible presence makes it difficult to estimate the sorption properties of rocks that may have experienced weathering.
- The second major difficulty lies in the transfer of sorption data from crushed material to intact rock. However, this is not related to the actual sorption processes but to physical-hydrological questions. These arise from the presence of two separate sorbing environments in crystalline

rock (rock matrix, fractures) and typically also from the lack of knowledge regarding the amount of flow-wetted surface in fractures.

Because of the ill-defined nature of these challenges, the derivation of sorption values for specific rocks and conditions is very difficult on the basis of generic experimental sorption data.

5.1.2 Examples of K_d setting approaches

In the following, an example of K_d derivation for granitic rocks carried out in the framework of safety assessments is discussed. The K_d setting and uncertainty assessment in granitic rock in support of the SKB Safety Assessment SR-Can is given in Crawford et al. (2006). According to the information given in this report, it represents the first instance within the SKB program that a procedure of applying correction factors is followed to transfer the experimental sorption data to values that are more representative for in situ conditions. In previous reports (e.g., Carbol and Engkvist, 1997), data for PA were apparently directly selected from compilations of experimental data.

Crawford et al. (2006) considered only one type of correction factor: surface area effects (relating to the use of crushed material in laboratory sorption measurements) were reportedly used in a rudimentary and somewhat subjective fashion (a factor of 0.1 was taken to be reasonable as correction for the differences in surface areas of typical crushed rock and intact rock). No other data transformations were carried out to derive data for the specific conditions of interest. Because of the difficulties of defining CEC or surface area of the specific rock surfaces contributing to sorption, only data measured for representative (whole) granitic rock samples were considered. No attempts were made to extrapolate from data for pure mineral phases to data for whole rock.

To take the expected in-situ conditions into account, the compiled experimental data were categorized into several groups according to the water composition (mainly levels of salinity). However, Crawford et al. (2006) concluded that a categorization of experiments is difficult because

- of the general sparsity of good quality data;
- chloride concentration is not a very good correlating parameter for K_d.

In the end, data were selected for two groups of water composition: non-saline and saline water. K_d values were defined by selecting all compiled literature data whose underlying experimental conditions correspond to these two water types (and the desired RN oxidation state, where relevant) and taking average values. Data sources for defining the recommended values included data from primary (original) literature as well as compilations (Shibutani et al. 1999, Stenhouse 1995). In terms of rock types, it was not attempted to strongly delineate different types of crystalline rock. The actual selection of recommendable data was done largely on the basis of expert judgement. More specifically, the numerical median of a subjectively selected group of literature values was calculated.

Uncertainties are discussed following the approach of Ochs and Talerico (2004). These include

- uncertainty in the compiled K_d data itself;
- uncertainty related to the use of generic data in site-specific safety analyses;
- uncertainty in the application conditions themselves.

The actual uncertainties were defined as statistical deviations from the recommended data (i.e.; as confidence intervals of the calculated median value).

5.1.3 Prediction of K_d for granitic environments

In this chapter, a first application of semi-quantitative estimations and TSMs to the evaluation of K_d in granitic environments is presented. As a first case, K_d evaluations of Am for granitic rock were carried out for the Mizunami URL condition (Mizunami granite), which is roughly estimated from MIZ-1 borehole data (Iwatsuki et al., 2005; Yoshida et al., 1998):

- Groundwater properties : Na-Cl type, Ionic strength - 3 mmol/L, Carbonate - 0.08 mmol/L, pH - 8.7
- Rock properties (composition): Quartz - 43.4%, K-feldspar - 16.3%, Plagioclase - 36.1%, Biotite - 4.0%

i) Semi-quantitative K_d estimation for Am

Relatively little is known regarding the mechanisms of Am sorption on granitic rock. When the composition of the Mizunami rock sample is considered, it can be assumed that mica (biotite) will be the only sorption-relevant solid. It had been shown in an earlier report (Lothenbach and Ochs, 1999) that the contribution of quartz and the possible contribution of feldspars can be neglected in comparison. Further, it can be assumed as a first approximation that sorption on mica minerals can be compared to sorption on clays in terms of sorption mechanism.

Figure 5.1 gives an overview of Am sorption on granitic rock and also a comparison to sorption on different clay minerals. Only few systematic and reliable datasets are available for Am sorption on granite. In general, these are compatible with data for smectite over a large pH range. By far the most complete dataset is given by Kitamura et al. (1999). As there is no obvious discrepancy between these data and the values from the other studies shown in Figure 5.1, only the dataset by Kitamura et al. (1999) was selected as primary data source for the estimation of a K_d value regarding the sorption of Am on Mizunami granite. The results of the estimation procedure are shown in Table 5.1 and Appendix I.

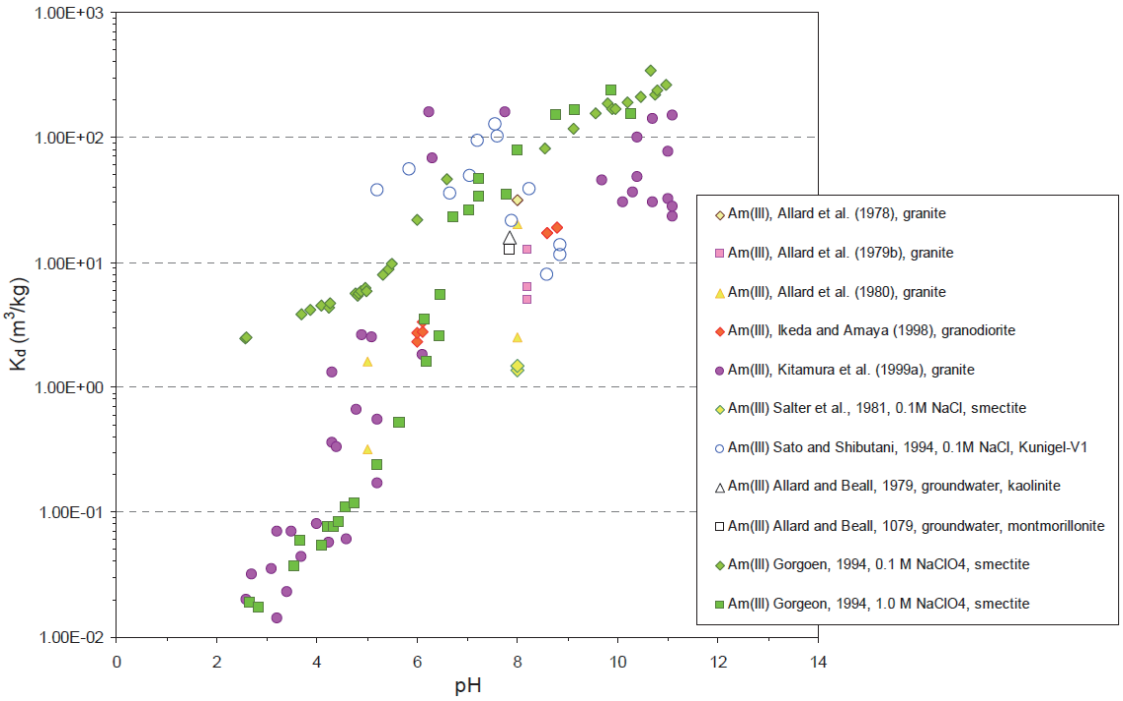

Figure 5.1 Overview of Am(III) sorption data on granite in comparison to sorption on some clays. Sorption data are taken from the JAEA-SDB (Tachi et al., 2011)

Table 5.1 Results of the K_d estimation for Am under the Mizunami condition.

K_d: CF-1 [m^3/kg]	K_d: CF-2a [m^3/kg]	K_d: CF-2b [m^3/kg]	Reference
111	162	264	Kitamura et al. (1999)

Scaling between the different mineralogies was done using the approach based on the BETvalues, because overall CEC and the percentage of mica were not known for both samples. The conversion factor CF-1 calculated using the BET values of the granite samples considered gives a value of 3.18.

The conversion factor CF-2 includes the effect of the solution speciation, which was similar for the two used solutions and resulted in conversion factors relatively close to unity. In the case of the conversion factor CF-2a, the complexation of the radionuclide with carbonate is viewed as fully competitive with respect to sorption. The conversion factor CF-2b excludes the competitive effect of carbonate. The conversion factor CF-2 was calculated using conversion factors of 1.45 (CF-spec/a) and 2.37 (CF-spec/b). Since the solution used by Kitamura et al. (1999) contains no carbonate, CF-2b results in a higher estimated K_d value than CF-2a.

The overall influence of the speciation of Am in the different systems on calculated K_d is relatively small (CF-1 is similar to CF-2). This shows that the conditions of the data source and the application system are relatively well matched, which lends more confidence to the derived values.

ii) TSM application

The JAEA11NEM_Am sorption model (JAEA, 2012; see chapter 3.1) was considered in a first step, using the following approach:

- The JAEA11_Am sorption model was used as is; i.e., without any modification of model parameters.
- To apply the model to granitic rock, a simple approach was followed:
  - mica was considered as the only sorption-relevant mineral in granite, Mizunami granite contains 4 % biotite;
  - mica was considered to contain the same concentration (in moles/g) of SOH edge sites as smectite, and to have 1 % of the CEC of smectite;
  - no correction regarding the accessibility of sorption sites in intact rock were made;
  - the same water composition at pH 8.5 as for the above estimation, and pH was additionally adjusted to lower values;
  - a solid/water ratio of 5 kg/L was considered for the intact rock;

The results are shown in Figure 5.2. It can be seen that this simple approach gives good agreement with values measured on granite samples by various authors. The trend of sorption vs. pH is also following the trend of the data very well. At higher pH, sorption is dominated by surface complexation and there is almost no difference between the two solid/water ratios as there is no competition for these sites by other metal ions. Below pH 6, ion exchange is more important and the competition by major cations strongly influences Am sorption. Since the amount of competing cations is given by the water composition, the extent of competition increases as the amount of available ion exchange sites becomes more limiting (i.e.; competition is important at low solid/water ratio and almost negligible at a very high solid/water ratio).

A critical parameter is the CEC assigned to the mica: if higher values are used, the predictions at low pH and high solid/water ratio start to deviate from the expected trend. Overall, the simple approach seems to be promising and will be investigated further by considering a restricted accessibility in intact rock.

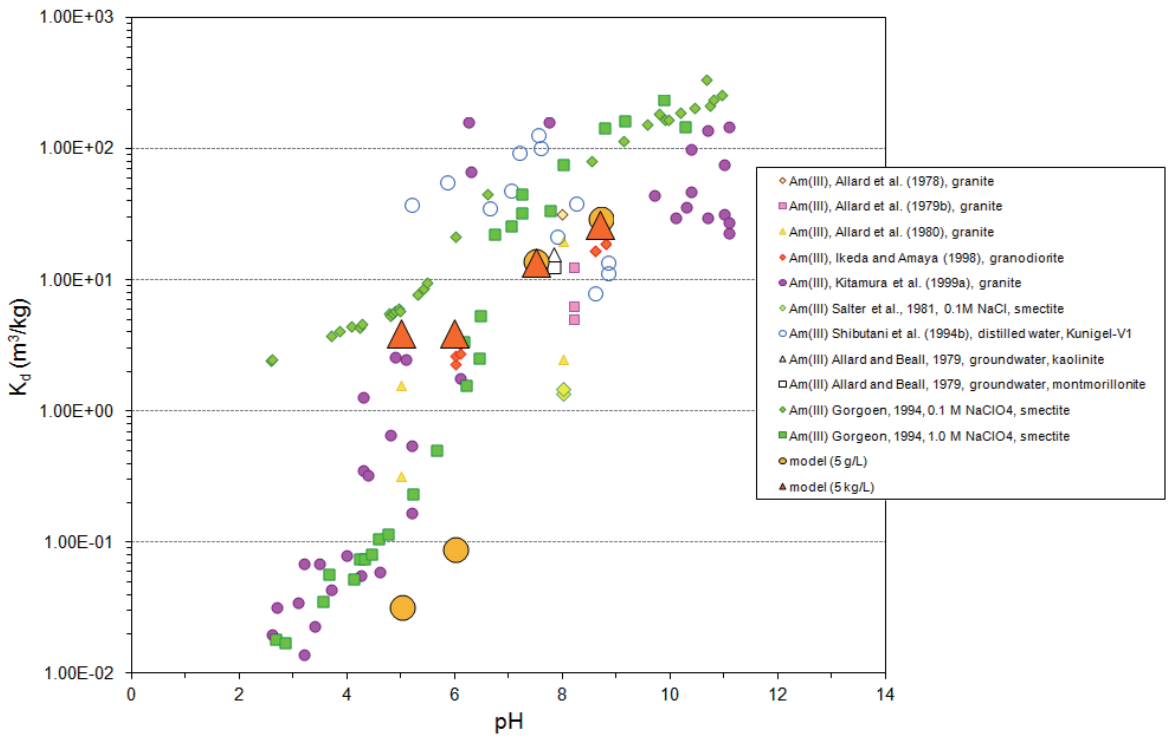

Figure 5.2 Overview of Am(III) sorption data on granite in comparison to sorption on some clays. The results of the model calculations at solid/water ratios of 5 g/L and 5 kg/L are indicated in addition. The calculated value corresponding to the original pH of the groundwater (pH 8.5) is marked by a circle.

5.2 Sorption models for other rock types

In addition to clays (bentonite and clay-type rock formations) and granitic rocks, other rock types may be of relevance as host rocks for the disposal of radioactive waste. Such rock types may include sedimentary and volcanic rock types such as sandstone or tuff. While their specific composition may vary widely, a common feature of most of these rocks is a relatively complex mineralogy. Typically, such rocks contain more than one sorption-relevant mineral phase, and these minerals may range from well-defined crystalline minerals to amorphous phases and coating that are difficult to characterize. Thus, the options available for sorption modeling depend strongly on the specific rock type.

The two principal approaches to sorption modeling, component additivity (CA) and generalized composite (GC) approaches, as well as their application to complex mineral assemblages, have been discussed in NEA (2005, 2012). This discussion is not repeated here. With a view to modeling radionuclide sorption in the context of radioactive waste disposal, the main difference of CA and GC approaches may be summarized as follows:

- A GC model necessarily has to be based on experimental sorption (and other) data obtained for the material in question. I.e., such models can only be developed for materials that are accessible and that can be subjected to sorption experiments. Their purpose is the description or

interpolation of sorption data rather than predictive applications. Because the mineral assemblage is treated as a hypothetical sorbent with the required types of surface groups, the models are not limited to the surface properties of model minerals. Further, such models can be applied to mineral mixtures where the contributions by individual minerals are not additive in a straightforward way. Because of this freedom in the choice of model properties, and because the model is necessarily based on sorption data obtained on the specific material of interest, GC models normally allow a very good description of sorption data.

- A CA model uses surface properties of the individual mineral components of a rock or sediment to approximate the behavior of the whole substrate. Often, the properties of model minerals are assumed to be applicable. Depending on the level of characterization carried out for a given rock and on the level of model calibration, such models may be used for predictive purposes (including scoping to help identify the major types of surfaces responsible for RN sorption) or to demonstrate system understanding. When applications for K_d setting are intended, some system-specific experimental sorption data are typically required to validate the predictive capability of such models.

In comparison to clay-dominated and crystalline rocks, other rock types are much less researched in terms of the sorption properties and modeling options. This is largely due to the fact that such non-clay sedimentary and volcanic rock types are not considered the most suitable host rocks in most disposal programs.

Accordingly, relatively little information is available in the literature with respect to the application of sorption models to such materials. A few illustrative examples are discussed in the following.

Davis et al. (2004) developed a GC model to describe the sorption behavior of U(VI) on a natural surficial (alluvial) sediment contaminated by uranium mining activities (Naturita site, Colorado, USA). The site was geochemically characterized which allowed to restrict the applicability of the model to the observed chemical conditions (i.e.; predictive applications were not of interest). The sediment consisted of weathered rocks and fines, covering a very wide range of grain sizes. The mineralogy was dominated by quartz, with smaller amounts of feldspars, carbonates, magnetite, and clays. The clay minerals included smectite/illite mixed layer clays and chlorite. Using two types of generic surface complexation sites in a surface complexation-non electrostatic model, two U(VI) surface reactions on each site were sufficient to describe U(VI) sorption in the observed range of pH and other conditions.

To aid the interpretation of measured K_d values, Talerico et al. (2003) developed a CA-type sorption model for the sorption of Sr and Ni by sandstone and pumice from the Rokkasho site in Japan. Both rock types contain clay minerals, quartz, other crystalline silicates (e.g. feldspars) as well as about 25 % to 50 % of a non-crystalline (X-ray amorphous) Al-silicate mineral with unknown identity and properties (abbreviated NCM in the above paper).

The complex mineral assemblage of the two rock types was approximated in a component-additivity approach, using simplified model mineralogies. These consisted of the identified minerals with known surface properties and of different model NCM minerals with assumed surface properties:

- The identified minerals in each rock type were represented with defined amounts of montmorillonite and quartz. For both model minerals, surface complexation was considered, using TSMs from the literature. A simple ion exchange model was used additionally for the

montmorillonite component. The same surface properties (site densities) were considered for Sr and Ni.

- The unknown NCM component was modeled in three different ways by assuming the surface properties of i) a amorphous Si-oxide, ii) a mixed Si/Al-oxide with the same properties as the edge surface of montmorillonite, iii) an unknown mineral with the ion exchange properties of montmorillonite (but no surface complexation sites).

The sorption of Ni and Sr on both pumice and sandstone could be consistently explained with the above approach when NCM was considered as an ion exchanger, i.e; using approach iii) above. The other interpretations of the NCM component lead to meaningless results, on the other hand. For both rock types, equilibrium with calcite needed to be considered to explain the competition of ion exchange by major ions (Ca).

6. Summary and conclusions

The integrated approaches for site-specific K_d setting for PA calculations were developed focusing on transferring procedures. These procedures used in this report includes the thermodynamic sorption model (TSM) and the semi-quantitative estimation. It is not feasible to determine K_d value experimentally for all PA conditions, these transferring approaches of sorption data from experimental conditions to another PA and site-specific conditions are critically important.

The present report focused on illustrating a range of example calculations regarding the derivation of K_d values and their uncertainties for four key radionuclides, Cs, Ni, Am and Th, and three geochemical conditions for Horonobe mudstone systems. This K_d-setting exercises allowed to estimate the magnitude of sorption and their uncertainty under the expected in-situ conditions, and to identify some critical gaps in the existing data and process understanding. The comparisons between two transferring procedures indicated that the comparative discussions are effective to confirm the reliability of K_d setting, and the careful selections from two results are needed according to the situation in the existing data and process understanding as the followings:

- Both approaches (TSM and semi-quantitative estimation) gave nearly identical results in terms of recommendable K_d values, when sufficient data and model were existing as in case of Ni.
- It was recommended to rely on the model-derived K_d values in case of Cs, because their TSM were reliable and gave conservative results.
- As in case of Am and Th, K_d values must be carefully selected by accounting data and model situations and conservatism, e.g., underestimated K_d values for Th by TSM due to lack of model parameters of ternary surface complexes.

Further studies are required to test and improve the K_d setting approaches for future site-specific K_d setting for PA calculations as the followings:

- Testing the applicability of K_d setting approaches for wide ranges of conditions and different phases of site investigation, and comparisons with actual experimental data obtained for in-situ conditions in experimental programs.
- The data situation and available models for key radionuclides - solid systems must be improved: e.g., the applicability of TSM of tetravalent actinides for high carbonate conditions can only be resolved by obtaining systematic experimental data on the sorption as a function of carbonate concentration.
- The extension of proposed K_d setting approaches to other rock types (e.g., granitic rocks) must be continuously investigated, so that reliable K_d parameters could be set under a variety of geological setting and geochemical conditions.

References

Allard, B., Kipatsi, H. and Torstenfelt, B. (1978): Sorption of long-lived radionuclides on clays and rocks, part 2, KBS Technical Report 98.

Allard, B. and Beall, G.W. (1979): Sorption of americum on geologic media, Journal of Environmental Science and Health, No.A14, pp.507-518.

Allard, B., Rydberg, J., Kipatsi, H. and Torstenfelt, B. (1979): Disposal of Radioactive Waste in Granitic Bedrock, American Chemical Society, No.4, pp.47-73.

Allard, B., Beall, G.W. and Krajewski, T. (1980): The Sorption of Actinides in Igneous Rocks, Nuclear Technology, vol.49, pp.474-480.

Baston, G.M.N., Berry, J.A., Bond, K.A., Brownsword, M. and Linklater, C.M. (1992): Effects of organic degradation products on the sorption of actinides. Radiochimica Acta 58/59, pp.349-356.

Bradbury, M.H. and Baeyens, B. (1998): N_2-BET surface area measurements on crushed and intact minerals and rocks: A proposal for estimating sorption transfer factors. Nuclear Technology, 122, pp.250–253.

Bradbury, M.H. and Baeyens, B. (1999): Modelling the sorption of Zn and Ni on Ca-montmorillonite. Geochim. Cosmochim. Acta, 63, pp.325-336.

Bradbury, M.H. and Baeyens, B. (2000): A generalised sorption model for the concentration dependent uptake of Cs by argillaceous rocks. J. Contam. Hydrol. 42, pp.141-163.

Bradbury, M.H. and Baeyens, B. (2003a): Near-field sorption data bases for compacted MX-80 bentonite for performance assessment of high-level radioactive waste repository in Opalinus Clay host rock. Technical Report 02-18. Nagra, Wettingen, Switzerland.

Bradbury, M.H. and Baeyens, B. (2003b): Far-field sorption data bases for performance assessment of high-level radioactive waste repository in an undisturbed Opalinus Clay host rock. Technical Report 02-19. Nagra, Wettingen, Switzerland.

Bradbury, M.H. and Baeyens, B. (2005): Modelling the sorption of Mn(II), Co(II), Ni(II), Zn(II), Cd(II), Eu(II), 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, pp.875-892.

Carbol, P. and Engkvist, I. (1997): Compilation of radionuclide sorption coefficients for performance assessment, SKB Technical Report, R-97-13.

Catalano, J.G. and Brown, G.E., Jr. (2005): Uranyl adsorption onto montmorillonite: Evaluation of binding sites and carbonate complexation. Geochim. Cosmochim. Acta 69, pp.2995-3005.

Crawford, J., Neretnieks, I. and Malmström, M. (2006): Data and uncertainty assessment for radionuclide K_d partitioning coefficients in granitic rock for use in SR-Can calculations. SKB report R-06-75, Swedish Nuclear Fuel and Waste Management Co., Stockholm.

Davis, J.A., Curtis, G.P., and Randall, J.D. (2004): Application of surface complexation modelling to describe uranium (VI) adsorption and retardation at the uranium mill tailings site at Naturita, Colorado. U.S. Nuclear Regulatory Commission, Office of Nuclear Regulatory Research, Washington, DC 20555-0001.

Dixon, J.B., Weed, S.B. (1977): Minerals in Soil Environments, Soil Science Society of America.

Fletcher, P. and Sposito, G. (1989): The chemical modelling of clay/electrolyte interactions for montmorillonite, Clay Miner., 24, pp.375-391.

Gorgeon, L. (1994): Contribution à la modelisation physico-chimique de la retention de radioéléments à vie longue par des materiaux argileux. Ph.D. thesis. Université Paris 6.

Guillaumont, R., Fanghänel, T., Fuger, J., Grenthe, I., Neck, V., Palmer, D.A., Rand, M.H. (2003): Update on the Chemical Thermodynamics of Uranium, Neptunium, Plutonium, Americium and Technetium. F. J. Mompean, M. Illemassene, C. Domenchi-Orti, K. Ben Said (eds.), Elsevier, Amsterdam.

Hama, K., Kunimaru, T., Metcalfe, R. and Martin, A.J. (2007): The hydrogeochemstry of argillaceous rock formations at the Horonobe URL site, Japan. Phys. Chem. Earth 32, pp.170–180.

Hummel, W., Berner, U., Curti, E., Pearson, F.J. and Thoenen, T., (2002): Nagra/PSI Chemical Thermodynamic Data Base 01/01", Nagra Technical Report NTB 02-16, Nagra, Wettingen, Switzerland, and Universal Publishers/uPublish.com, Parkland, Florida, USA.

Ikeda, T. and Amaya, T. (1998): Model development of chemical evolution in repository Vol.II Acquisition of nuclide migration data in near-field, PNC Technical Report, PNC TJ 1281 98-003.

Iwatsuki, T., Furue, R., Mie, H., Ioka, S. and Mizuno, T. (2005): Hydrochemical baseline condition of groundwater at the Mizunami underground research laboratory (MIU). Applied Geochemistry, 20(12), pp.2283-2302.

JAEA (2007): Horonobe underground research laboratory project. Synthesis of phase I investigation 2001–2005. Volume “Geological disposal research”. JAEA technical report , JAEA-Research 2007-045.

JAEA (2012): The report on the project for assessment methodology development of chemical effects on geological disposal systems, funded by METI, Japan.

JNC (1999): H12 project to establish technical basis for HLW disposal in Japan. Project overview report. JNC technical report, JNC TN 1400 99-010.

Kato, H., Muroi, M., Yamada, N., Ishida, H. and Sato, H. (1995): Estimation of effective diffusivity in compacted bentonite. In: Scientific Basis for Nuclear Waste Management XVIII, Mater. Res. Soc. Symp. Proc., 353, pp.277–284.

Kawaguchi, M., Kaminoyama, M., Takase, H. and Suzuki, K. (1989): Study of Cation Adsorption Model on Sodium Bentonite, High-Level Radioactive Waste and Spent Fuel Management, 2, pp.313-317.

Kitamura, A., Yamamoto, T., Nishikawa, S. and Moriyama, H. (1999): Sorption Behavior of Am(III) onto Granite, Journal of Radioanalytical and Nuclear Chemistry, vol.239, pp.449-453.

Kitamura, A., Fujiwara, K., Doi, R., Yoshida, Y., Mihara, M., Terashima, M. and Yui, M. (2010): JAEA thermodynamic database for performance assessment of geological disposal of high-level radioactive and TRU wastes. JAEA technical report, JAEA-Data/Code 2009-024.

Lauber, M., Baeyens, B. and Bradbury M.H. (2000): Physico-chemical characterisation and sorption measurements of Cs, Sr, Ni, Eu, Th, Sn and Se on Opalinus Clay from Mont Terri. PSI Bericht Nr. 00-10, Paul Scherrer Institut, Villigen, Switzerland.

Lothenbach, B. and Ochs, M. (1999): Development and application of a simple sorption model for cesium, radium/strontium and americium on granitic rocks. In Ueta et al. (1999) Development of database system for performance assessment (III). JNC TJ8400 99-032.

Marques Fernandes, M., Baeyens, B. and Bradbury, M.H. (2008): The influence of carbonate complexation on lanthanide/actinide sorption on montmorillonite, Radiochim. Acta 96, pp.691-697.

Nagasaki, S., Tanaka, S. and Suzuki, A. (1997): Affinity of finely dispersed montmorillonite colloidal particles for americium and lanthanides. J. Nucl. Materials 244, pp.29-30.

NEA (2005): NEA Sorption Project. Phase II: Interpretation and Prediction of Radionuclide Sorption onto Substrates relevant for Radioactive Waste Disposal using Thermodynamic Sorption Models. By Davis, J., Ochs, M., Olin, M., Payne, T., Tweed, C., OECD-NEA, Paris.

NEA (2012): NEA Sorption Project. Phase III: Thermodynamic sorption modeling in support of radioactive waste disposal safety cases. OECD-NEA, Paris.

Ochs, M., Lothenbach, B., Wanner, H., Sato, H. and Yui, M. (2001): An integrated sorption-diffusion model for the calculation of consistent distribution and diffusion coefficients in compacted bentonite. J. Contam. Hydrol., 47, pp.283-296.

Ochs, M. and Talerico, C. (2004): SR-Can. Data and uncertainty assessment. Migration parameters for the bentonite buffer in t*he* KBS-3 concept. SKB TR-04-18, Svensk Kärnbränslehantering AB.

Ochs, M., Kunze, S., Saito, Y., Kitamura, A., Tachi, Y. and Yui, M. (2008): Application of the sorption database to K_d-setting for Horonobe rocks, JAEA technical report, JAEA-Reasearch 2008-017.

Ochs M., Tachi Y., Ganter C., Suyama T. and Yui M. (2011): Development of mechanistic sorption model and treatment of uncertainties for Ni sorption on montmorillonite / bentonite. JAEA technical report, JAEA-Research 2010-052.

Oda, C. and Shibata, M. (1999): Modeling and experimental studies on bentonite-water interaction, JNC technical report, JNC TN8400 99-032.

Parkhurst D.L. and Appelo C.A.J. (1999): User's guide to PHREEQC (ver.2) – a computer program for speciation, batch-reaction, one-dimensional transport, and inverse geochemical calculations, U.S. Geological Survey, Water-resources investigations report 99-4259.

Poinssot, C., Baeyens, B. and Bradbury, M.H. (1999): Experimental studies of Cs, Sr, Ni, and Eu sorption on Na-illite and the modeling of Cs sorption. PSI Bericht Nr. 99-06, Paul Scherrer Institut, Villigen, Switzerland.

Rabung, Th., Pierret, M.C., Bauer, A., Geckeis, H., Bradbury, M.H. and Baeyens, B. (2005): Sorption of Eu(III)/Cm(III) on Ca-montmorillonite and Na-illite Part 1: Batch sorption and time resolved laser fluorescence spectroscopy experiments. Geochim. Cosmochim. Acta 69, pp.5393–5402.

Rand, M.H., Fuger, J., Grenthe, I., Neck, V. and Rai, D. (2008): Chemical Thermodynamics of Thorium.OECD-NEA, Paris.

Salter, P.F., Ames, L.L. and McGarrah, J. E. (1981): Sorption of selected radionuclides on secondary minerals associated with the Columbia River Basalts, RHO-BWI-LD-43.

Sato, H. and Shibutani, T. (1994). Study on adsorption and diffusion mechanism of nuclides in buffer material and geosphere. PNC technical review 91, pp.71-89.

Shibutani, T., Yui, M. and Yoshikawa, H. (1994): Sorption mechanism of Pu, Am and Se on sodium-bentonite, MRS Symposium Proceedings, vol.333, pp.725-730.

Shibutani, T., Suyama, T. and Shibata, M. (1999): Sorption database for radionuclides on bentonite and rocks. JNC Technical Report, JNC TN8410 99-050.

Sposito G. (1984): The surface chemistry of soils, Oxford University Press, New York.

Stenhouse, M. (1995): Sorption databases for crystalline, marl and bentonite for safety assessment. Nagra Technical Report NTB-93-06, NAGRA, Switzerland.

Tachi, Y., Yotsuji, K., Seida, Y. and Yui, M. (2009): Diffusion of cesium and iodine in compacted sodium montmorillonite under different saline conditions. In: Scientific Basis for Nuclear Waste Management XXXIII, Mater. Res. Soc. Symp. Proc., 1193, pp.545–552.

Tachi, Y., Yotsuji, K., Suyama, T., Ochs, M. and Yui, M. (2010): Development of the mechanistic sorption and diffusion model / database for safety assessment of geological disposal - Prototype model / database for bentonite systems -. JAEA technical report, JAEA-Research 2009-069.

Tachi, Y., Suyama, T., Ochs, M. and Ganter, C. (2011a): Development of JAEA sorption database (JAEA-SDB): Update of data evaluation functions and sorption/QA data. JAEA-Data/Code 2010-03.

Tachi Y., Yotsuji K., Seida Y. and Yui M. (2011b): Diffusion and sorption of Cs^+, I^- and HTO in samples of the argillaceous Wakkanai Formation from the Horonobe URL, Japan: Clay-based modeling. Geochim. Cosmochim. Acta 75, pp.6742–6759.

Talerico, C., Ochs, M., Ueta, S., and Sasaki, N. (2003): Thermodynamic modeling and sensitivity analysis of K_d values for radionuclide migration in sedimentary host rocks. Mat. Res. Soc. Symp. Vol. 807, pp.671-677

Tertre, E., Berger, G., Castet, S., Loubet, M. and Giffaut, E. (2005): Experimental sorption of Ni^{2+}, Cs^+ and Ln^{3+} onto a montmorillonite up to 150 °C. Geochim. Cosmochim. Acta 69, pp.4937-4948.

Ueta, S. (1998): Development of database system for performance assessment (II), PNC technical report, PNC TJ1211 98-001.

Wang, L., Maes, A., De Canniere, P., and Van der Lee, J. (1998): Sorption of europium on illite (Silver Hill Montana). Radiochim. Acta, 82, pp.233-237.

Wanner, H., Albinsson, Y. and Wieland, E. (1996): A thermodynamic surface model for caesium sorption on bentonite, Fresenius J. Anal. Chem., 354, pp.763-769.

Yoshida, M., Zheng, Z., Inoue, Y. and Shintani, K. (1998): Mineralogical tests on the core of the borehole MIU-I, JNC technical report, JNC TJ7440 99-005.

Appendix - I

K_d conversion and RN speciation tables

- Horonobe mudstone (Cs, Ni, Am, Th)
- Mizunami granite (Am)

This is a blank page.

Speciation Case I

Filename Cs_equilibrium_case_I_120511_DAT Gorgeon(1994)_Cs_NAPSI
Preequilibration_case_I_120511_DAT

TDB: JAEA, version 100331c1

Cs(I) speciation (Gorgeon, 1994)

System		Application	Data Source
Substrate		Horonobe mudstone	Na-Illite
Solution		HDB-6	1 M NaCl
pH		6.752	6.75
pCO_2		-	-
CEC		13.75	122.00
BET		5.55	97.00
Cs^{1+} dissolved	(mol/kg)	1.99E-11	2.00E-02
Al^{3+}	(mol/kg)	1.72E-10	
Ca^{2+}	(mol/kg)	1.81E-03	
Fe^{2+}	(mol/kg)	1.41E-05	
Fe^{3+}	(mol/kg)	9.53E-21	
Mg^{2+}	(mol/kg)	1.10E-03	
Mn^{2+}	(mol/kg)	2.52E-07	
Na^{+}	(mol/kg)	2.55E-01	1.00E+00
K^{+}	(mol/kg)	4.19E-04	0.00E+00
Sr^{2+}	(mol/kg)	3.02E-05	
NH_4^{+}	(mol/kg)	1.08E-03	0.00E+00
∑ competitive cations in solution with Na^{+} (C-cmp/a)	(mol/kg)	2.59E-01	1.00E+00
Al^{3+}	(mol/kg)	1.72E-10	
Ca^{2+}	(mol/kg)	1.81E-03	
Fe^{2+}	(mol/kg)	1.41E-05	
Fe^{3+}	(mol/kg)	9.53E-21	
Mg^{2+}	(mol/kg)	1.10E-03	
Mn^{2+}	(mol/kg)	2.52E-07	
Na^{+}	(mol/kg)	2.55E-01	1.00E+00
K^{+}	(mol/kg)	4.19E-04	0.00E+00
Sr^{2+}	(mol/kg)	3.02E-05	
NH_4^{+}	(mol/kg)	1.08E-03	0.00E+00
∑ competitive cations in solution without Na^{+} (C-cmp/b)	(mol/kg)	4.44E-03	0.00E+00

K_d-derivation Cs(I) Gorgeon (1994)

Overview Pred. K_d [m^3/kg]	Mudstone
	HDB-6
Pred. K_d: CF-1 (% clay)	0.53
Pred. K_d: CF-3a (% clay)	2.05

Cs(I) K_d-derivation		Conditions		Conversion (C) / Uncertainty (U)
		Data source[1]	Application	CF
			Mudstone	Mudstone
Solution		1 M NaCl	HDB-6	HDB-6
Substrate		Illite	Mudstone	Mudstone
Experimental K_d	(m^3/kg)	2.80		
Conversion Mineralogy (CF-min)				
CEC[2]	(meq/100g)	21.2	13.75	0.65
BET[2]	(m^2/g)	111.5	5.55	0.05
% clay		100	19.00	0.19
Conversion pH (CF-pH)				
pH		6.75	6.75	1.00
Conversion Speciation (CF-spec)	*irrelevant*			
Conversion Competition (Cationic) (CF-cmp)				
∑ competitive cations in soln. with Na^+ (CF-cmp/a)	(mol/l)	1.00	0.26	3.86
∑ competitive cations in soln. without Na^+ (CF-cmp/b)	*not considered*			

(1) K_d values from data table Gorgeon (1994) given as log Kd
(2) Values of the average composition

Cs(I) K_d-derivation via % clay			Mudstone	CF
CF-1 (% clay)				0.19
Competition				
CF-3 (% clay)				0.73
				UF
Predicted K_d: CF-1 (% clay)	(m^3/kg)		**0.53**	
UF-starting K_d				1.6
UF-pH conversion				2.5
UF-min				1.4
UF-total				5.5
K_d upper limit	(m^3/kg)		2.95	
K_d lower limit	(m^3/kg)		0.10	
Predicted K_d: CF-3 (% clay)	(m^3/kg)		**2.05**	
UF-starting K_d				1.6
UF-pH conversion				2.5
UF-cmp				1.2
UF-min				1.4
UF-total				6.7
K_d upper limit	(m^3/kg)		13.65	
K_d lower limit	(m^3/kg)		0.31	

Speciation Case I

Filename	Cs_equilibrium_case_I_120511_DAT	PSI(1999)_0.1MNaClO4_Cs_NAPSI
	Preequilibration_case_I_120511_DAT	PSI(1999)_0.01MNaClO4_Cs_NAPSI
TDB:	JAEA, version 100331c1	

Cs(I) speciation (Poinssot et al., 1999)

System		Application	Data Source	
Substrate		Horonobe mudstone	Na-Illite	
Solution		HDB-6	0.01 M $NaClO_4$	0.1 M $NaClO_4$
pH		6.752	6.6	6.6
pCO_2		-	-	-
CEC		13.75	122.00	122.00
BET		5.55	97.00	97.00
Cs^{1+} dissolved	(mol/kg)	1.99E-11	1.00E-08	1.00E-08
Al^{3+}	(mol/kg)	1.72E-10		
Ca^{2+}	(mol/kg)	1.81E-03		
Fe^{2+}	(mol/kg)	1.41E-05		
Fe^{3+}	(mol/kg)	9.53E-21		
Mg^{2+}	(mol/kg)	1.10E-03		
Mn^{2+}	(mol/kg)	2.52E-07		
Na^{+}	(mol/kg)	2.55E-01	1.00E-02	1.00E-01
K^{+}	(mol/kg)	4.19E-04	4.70E-06	1.50E-05
Sr^{2+}	(mol/kg)	3.02E-05		
NH_4^{+}	(mol/kg)	1.08E-03	0.00E+00	0.00E+00
∑ competitive cations in solution with Na^{+} (CF-cmp/a)	(mol/kg)	2.59E-01	1.00E-02	1.00E-01
Al^{3+}	(mol/kg)	1.72E-10		
Ca^{2+}	(mol/kg)	1.81E-03		
Fe^{2+}	(mol/kg)	1.41E-05		
Fe^{3+}	(mol/kg)	9.53E-21		
Mg^{2+}	(mol/kg)	1.10E-03		
Mn^{2+}	(mol/kg)	2.52E-07		
Na^{+}	(mol/kg)	2.55E-01	1.00E-02	1.00E-01
K^{+}	(mol/kg)	4.19E-04	4.70E-06	1.50E-05
Sr^{2+}	(mol/kg)	3.02E-05		
NH_4^{+}	(mol/kg)	1.08E-03	0.00E+00	0.00E+00
∑ competitive cations in solution without Na^{+} (CF-cmp/b)	(mol/kg)	4.44E-03	4.70E-06	1.50E-05

K_d-derivation Cs(I) on Illite Poinssot et al. (1999)

Overview Pred. K_d [m^3/kg]	A	B
	Mudstone	Mudstone
	HDB-6	HDB-6
Pred. K_d: CF-1 (% clay)	48.01	8.17
Pred. K_d: CF-3a (% clay)	1.85	3.15

Cs(I) K_d-derivation (Illite)		Conditions			Conversion (C) / Uncertainty (U)	
		Data source[1]		Application[2]	CF for A	CF for B
		A	B	Mudstone	Mudstone	Mudstone
Solution		0.01 M $NaClO_4$	0.1 M $NaClO_4$	HDB-6	HDB-6	HDB-6
Substrate		Na-Illite[3]	Na-Illite[3]	Mudstone	Mudstone	Mudstone
Experimental K_d	(m^3/kg)	235.00	40.00			
Conversion Mineralogy (CF-min)						
CEC[5]	(meq/100g)	122.00	122.00	13.75		
BET[4]	(m^2/g)	97.00	97.00	5.55		
% clay		93.00	93.00	19.00	0.20	0.20
% Illite		93.00	93.00	7.00	0.08	0.08
Conversion pH (CF-pH)	*irrelevant*	6.75	6.75	6.75		
Conversion Speciation (CF-spec)	*irrelevant*					
Conversion Competition (Cationic) (CF-cmp)						
∑ competitive cations in soln. with Na^+ (CF-cmp/a)	(mol/l)	0.01	0.10	0.26	0.04	0.39
∑ competitive cations in soln. without Na^+ (CF-cmp/b)	(mol/l)	4.70E-06	1.50E-05	4.44E-03	1.06E-03	3.37E-03

(1) K_d value from Poinssot et al. (1999),K_d at pH = 6.63; [Cs] = 1.0E-8 M
(2) Values of the average composition
(3) Conditioned Na-Illite; clay mineral composition: 93% illite and 7% kaolinite
(4) BET value of non-treated illite
(5) CEC value of treated Na-illite; pH 7.1

		A	B		
Cs(I) K_d-derivation via % clay		Mudstone	Mudstone	CF for A	CF for B
CF-1 (% clay)				0.20	0.20
Competition					
CF-3 (% clay)				0.01	0.08
				UF	UF
Predicted K_d: CF-1 (% clay)	(m^3/kg)	**48.01**	**8.17**		
UF-starting K_d				1.6	1.6
UF-min				1.4	1.4
UF-total				2.2	2.2
K_d upper limit	(m^3/kg)	106.53	18.13		
K_d lower limit	(m^3/kg)	21.64	3.68		
Predicted K_d: CF-3 (% clay)		**1.85**	**3.15**		
UF-starting K_d				1.6	1.6
UF-cmp				1.2	1.2
UF-min				1.4	1.4
UF-total				2.7	2.7
K_d upper limit		4.93	8.39		
K_d lower limit		0.70	1.18		

Speciation Case I

Filename Cs_equilibrium_case_I_120511_DAT PW_Sato&Shibutani(1994)_NAPSI
Preequilibration_case_I_120511_DAT

TDB: JAEA, version 100331c1

Cs(I) speciation (Sato & Shibutani, 1994)

System		Application	Data Source
Substrate		Horonobe mudstone	Kunigel V1
Solution		HDB-6	Dist. equil. water
pH		6.75	6.75
pCO_2		-	-
CEC		13.75	60.10
BET		5.55	-
Cs^{1+} dissolved	(mol/kg)	1.99E-11	7.52E-05
Al^{3+}	(mol/kg)	1.72E-10	
Ca^{2+}	(mol/kg)	1.81E-03	9.33E-05
Fe^{2+}	(mol/kg)	1.41E-05	-
Fe^{3+}	(mol/kg)	9.53E-21	-
Mg^{2+}	(mol/kg)	1.10E-03	-
Mn^{2+}	(mol/kg)	2.52E-07	-
Na^{+}	(mol/kg)	2.55E-01	9.54E-04
K^{+}	(mol/kg)	4.19E-04	1.68E-05
Sr^{2+}	(mol/kg)	3.02E-05	-
NH_4^{+}	(mol/kg)	1.08E-03	0.00E+00
∑ competitive cations in solution with Na^{+} (C-cmp/a)	(mol/kg)	2.59E-01	1.06E-03
Al^{3+}	(mol/kg)	1.72E-10	
Ca^{2+}	(mol/kg)	1.81E-03	9.33E-05
Fe^{2+}	(mol/kg)	1.41E-05	-
Fe^{3+}	(mol/kg)	9.53E-21	-
Mg^{2+}	(mol/kg)	1.10E-03	-
Mn^{2+}	(mol/kg)	2.52E-07	-
Na^{+}	(mol/kg)	2.55E-01	9.54E-04
K^{+}	(mol/kg)	4.19E-04	1.68E-05
Sr^{2+}	(mol/kg)	3.02E-05	-
NH_4^{+}	(mol/kg)	1.08E-03	0.00E+00
∑ competitive cations in solution without Na^{+} (C-cmp/b)	(mol/kg)	4.44E-03	1.10E-04

K_d-derivation Cs(I) on Kunigel-V1 Sato and Shibutani (1994)

Overview Pred. K_d [m^3/kg]	Mudstone
	HDB-6
Pred. K_d: CF-1 (% clay)	0.35
Pred. K_d: CF-3a (%clay)	1.44E-03

Cs(I) K_d-derivation (Smectite)		Conditions		Conversion (C) / Uncertainty (U)
		Data source[1]	Application[2]	CF
			Mudstone	Mudstone
Solution[5]		Dist. equil. water	HDB-6	HDB-6
Substrate		Kunigel-V1	Mudstone	Mudstone
Experimental K_d	(m^3/kg)	0.85		
Conversion Mineralogy (CF-min)				
CEC[4]	(meq/100g)	60.10	13.75	0.23
BET[6]	(m^2/g)	35.00	5.55	0.16
% clay[3]		46.00	19.00	0.41
Conversion pH (CF-pH)	*irrelevant*	6.75	6.75	
Conversion Speciation (CF-spec)	*irrelevant*			
Conversion Competition (Cationic) (CF-cmp)				
∑ competitive cations in soln. with Na^+ (CF-cmp/a)	(mol/l)	1.06E-03	2.59E-01	4.10E-03
∑ competitive cations in soln. without Na^+ (CF-cmp/b)	*not considered*			

(1) K_d value from JAEA-SDB database, Sato and Shibutani (1994),. [Cs] = 7.52E-5 M
(2) Values of the average composition
(3) clay mineral composition of data source from Sasaki et al. (1995): smectite 46-49%, quartz 29-38% and other trace impurities
(4) CEC value of data source from Sasaki et al. (1995)
(5) [Na^+] calculated with Phreeqci Ion Exchange with Kunigel V1 equilibrated in distilled water (S:W = 2 g/L)
(6) Value from Dixon and Weed (1977)

Cs(I) K_d-derivation via % clay		Mudstone	CF
CF-1 (% clay)			0.41
Competition			
CF-3 (% clay)			1.70E-03
			UF
Predicted Kd: CF-1 (% clay)	(m^3/kg)	**0.35**	
UF-starting K_d			1.6
UF-min			1.4
UF-total			2.2
K_d upper limit	(m^3/kg)	0.78	
K_d lower limit	(m^3/kg)	0.16	
Predicted Kd: CF-3 (% clay)	(m^3/kg)	**1.44E-03**	
UF-starting K_d			1.6
UF-cmp			1.2
UF-min			1.4
UF-total			2.7
K_d upper limit	(m^3/kg)	3.84E-03	
K_d lower limit	(m^3/kg)	5.42E-04	

Speciation Case II

Filename Cs_equilibrium_case_II_120509_DAT Gorgeon(1994)_Cs_NAPSI
Preequilibration_case_II_120509_DAT

TDB: JAEA, version 100331c1

Cs(I) speciation (Gorgeon, 1994)

System		Application	Data Source
Substrate		Horonobe mudstone	Na-Illite
Solution		HDB-6	1 M NaCl
pH		8.5	8.5
pCO_2		-	-
CEC		13.75	122.00
BET		5.55	97.00
Cs^{1+} dissolved	(mol/kg)	1.97E-11	2.00E-02
Al^{3+}	(mol/kg)		
Ca^{2+}	(mol/kg)	4.15E-05	
Fe^{2+}	(mol/kg)	3.22E-07	
Fe^{3+}	(mol/kg)	1.07E-24	
Mg^{2+}	(mol/kg)	1.07E-03	
Mn^{2+}	(mol/kg)		
Na^{+}	(mol/kg)	2.68E-01	1.00E+00
K^{+}	(mol/kg)	4.21E-04	0.00E+00
Sr^{2+}	(mol/kg)		
NH_4^{+}	(mol/kg)	1.06E-03	0.00E+00
∑ competitive cations in solution with Na^{+} (C-cmp/a)	(mol/kg)	2.70E-01	1.00E+00
Al^{3+}	(mol/kg)		
Ca^{2+}	(mol/kg)	4.15E-05	
Fe^{2+}	(mol/kg)	3.22E-07	
Fe^{3+}	(mol/kg)	1.07E-24	
Mg^{2+}	(mol/kg)	1.07E-03	
Mn^{2+}	(mol/kg)		
Na^{+}	(mol/kg)	2.68E-01	1.00E+00
K^{+}	(mol/kg)	4.21E-04	0.00E+00
Sr^{2+}	(mol/kg)		
NH_4^{+}	(mol/kg)	1.06E-03	0.00E+00
∑ competitive cations in solution without Na^{+} (C-cmp/b)	(mol/kg)	2.59E-03	0.00E+00

K_d-derivation Cs(I) Gorgeon (1994)

Overview Pred. K_d [m^3/kg]	Mudstone
	HDB-6
Pred. K_d: CF-1 (% clay)	0.63
Pred. K_d: CF-3a (% clay)	2.32

Cs(I) K_d-derivation		Conditions		Conversion (C) / Uncertainty (U)
		Data source[1]	Application	CF
			Mudstone	Mudstone
Solution		1 M NaCl	HDB-6	HDB-6
Substrate		Illite	Mudstone	Mudstone
Experimental K_d	(m^3/kg)	3.30		
Conversion Mineralogy (CF-min)				
CEC[2]	(meq/100g)	21.2	13.75	0.65
BET[2]	(m^2/g)	111.5	5.55	0.05
% clay		100	19.00	0.19
Conversion pH (CF-pH)				
pH		8.5	8.50	1.00
Conversion Speciation (CF-spec)	*irrelevant*			
Conversion Competition (Cationic) (CF-cmp)				
∑ competitive cations in soln. with Na^+ (CF-cmp/a)	(mol/l)	1.00	0.27	3.70
∑ competitive cations in soln. without Na^+ (CF-cmp/b)	*not considered*			

(1) K_d values from data table Gorgeon (1994) given as log Kd
(2) Values of the average composition

Cs(I) K_d-derivation via % clay			Mudstone	CF
CF-1 (% clay)				0.19
Competition				
CF-3 (% clay)				0.70
				UF
Predicted K_d: CF-1 (% clay)	(m^3/kg)		**0.63**	
UF-starting K_d				1.6
UF-pH conversion				2.5
UF-min				1.4
UF-total				5.5
K_d upper limit	(m^3/kg)		3.48	
K_d lower limit	(m^3/kg)		0.11	
Predicted K_d: CF-3 (% clay)	(m^3/kg)		**2.32**	
UF-starting K_d				1.6
UF-pH conversion				2.5
UF-cmp				1.2
UF-min				1.4
UF-total				6.7
K_d upper limit	(m^3/kg)		15.44	
K_d lower limit	(m^3/kg)		0.35	

Speciation Case II

Filename	Cs_equilibrium_case_II_120509_DAT	PSI(1999)_0.1MNaClO4_Cs_NAPSI
	Preequilibration_case_II_120509_DAT	PSI(1999)_0.01MNaClO4_Cs_NAPSI
TDB:	JAEA, version 100331c1	

Cs(I) speciation (Poinssot et al., 1999)

System		**Application**	**Data Source**	
Substrate		**Horonobe mudstone**	**Na-Illite**	
Solution		**HDB-6**	**0.01 M $NaClO_4$**	**0.1 M $NaClO_4$**
pH		8.5	8.5	8.5
pCO_2		-	-	-
CEC		13.75	122.00	122.00
BET		5.55	97.00	97.00
Cs^{1+} dissolved	(mol/kg)	1.97E-11	1.00E-08	1.00E-08
Al^{3+}	(mol/kg)			
Ca^{2+}	(mol/kg)	4.15E-05		
Fe^{2+}	(mol/kg)	3.22E-07		
Fe^{3+}	(mol/kg)	1.07E-24		
Mg^{2+}	(mol/kg)	1.07E-03		
Mn^{2+}	(mol/kg)			
Na^{+}	(mol/kg)	2.68E-01	1.00E-02	1.00E-01
K^{+}	(mol/kg)	4.21E-04	4.70E-06	1.50E-05
Sr^{2+}	(mol/kg)			
NH_4^{+}	(mol/kg)	1.06E-03	0.00E+00	0.00E+00
∑ competitive cations in solution with Na^{+} (CF-cmp/a)	(mol/kg)	2.70E-01	1.00E-02	1.00E-01
Al^{3+}	(mol/kg)			
Ca^{2+}	(mol/kg)	4.15E-05		
Fe^{2+}	(mol/kg)	3.22E-07		
Fe^{3+}	(mol/kg)	1.07E-24		
Mg^{2+}	(mol/kg)	1.07E-03		
Mn^{2+}	(mol/kg)			
Na^{+}	(mol/kg)	2.68E-01	1.00E-02	1.00E-01
K^{+}	(mol/kg)	4.21E-04	4.70E-06	1.50E-05
Sr^{2+}	(mol/kg)			
NH_4^{+}	(mol/kg)	1.06E-03	0.00E+00	0.00E+00
∑ competitive cations in solution without Na^{+} (CF-cmp/b)	(mol/kg)	2.59E-03	4.70E-06	1.50E-05

K_d-derivation Cs(I) on Illite Poinssot et al., (1999)

Overview Pred. K_d [m^3/kg]	A	B
	Mudstone	Mudstone
	HDB-6	HDB-6
Pred. K_d: CF-1 (% clay)	30.65	9.19
Pred. K_d: CF-3a (% clay)	1.13	3.40

Cs(I) K_d-derivation (Illite)		Conditions			Conversion (C) / Uncertainty (U)	
		Data source[1]		Application[2]	CF for A	CF for B
		A	B	Mudstone	Mudstone	Mudstone
Solution		0.01 M $NaClO_4$	0.1 M $NaClO_4$	HDB-6	HDB-6	HDB-6
Substrate		Na-Illite[3]	Na-Illite[3]	Mudstone	Mudstone	Mudstone
Experimental K_d	(m^3/kg)	150.00	45.00			
Conversion Mineralogy (CF-min)						
CEC[5]	(meq/100g)	122.00	122.00	13.75		
BET[4]	(m^2/g)	97.00	97.00	5.55		
% clay		93.00	93.00	19.00	0.20	0.20
% Illite		93.00	93.00	7.00	0.08	0.08
Conversion pH (CF-pH)	*irrelevant*	8.50	8.50	8.50		
Conversion Speciation (CF-spec)	*irrelevant*					
Conversion Competition (Cationic) (CF-cmp)						
∑ competitive cations in soln. with Na^+ (CF-cmp/a)	(mol/l)	0.01	0.10	0.27	0.04	0.37
∑ competitive cations in soln. without Na^+ (CF-cmp/b)	(mol/l)	4.70E-06	1.50E-05	2.59E-03	1.81E-03	5.79E-03

(1) K_d value from Poinssot et al. (1999),K_d at pH = 6.63; [Cs] = 1.0E-8 M
(2) Values of the average composition
(3) Conditioned Na-Illite; clay mineral composition: 93% illite and 7% kaolinite
(4) BET value of non-treated illite
(5) CEC value of treated Na-illite; pH 7.1

		A	B		
Cs(I) K_d-derivation via % clay		Mudstone	Mudstone	CF for A	CF for B
CF-1 (% clay)				0.20	0.20
Competition					
CF-3 (% clay)				0.01	0.08
				UF	UF
Predicted K_d: CF-1 (% clay)	(m^3/kg)	**30.65**	**9.19**		
UF-starting K_d				1.6	1.6
UF-min				1.4	1.4
UF-total				2.2	2.2
K_d upper limit	(m^3/kg)	68.00	20.40		
K_d lower limit	(m^3/kg)	13.81	4.14		
Predicted K_d: CF-3a (% clay)		**1.13**	**3.40**		
UF-starting K_d				1.6	1.6
UF-cmp				1.2	1.2
UF-min				1.4	1.4
UF-total				2.7	2.7
K_d upper limit		3.02	9.05		
K_d lower limit		0.43	1.28		

Speciation Case II

Filename Cs_equilibrium_case_II_120509_DAT PW_Sato&Shibutani(1994)_NAPSI
Preequilibration_case_II_120509_DAT

TDB: JAEA, version 100331c1

Cs(I) speciation (Sato & Shibutani, 1994)

System		Application	Data Source
Substrate		Horonobe mudstone	Kunigel V1
Solution		HDB-6	Dist. equil. water
pH		8.5	8.5
pCO_2		-	-
CEC		13.75	60.10
BET		5.55	-
Cs^{1+} dissolved	(mol/kg)	1.97E-11	7.52E-05
Al^{3+}	(mol/kg)		
Ca^{2+}	(mol/kg)	4.15E-05	9.33E-05
Fe^{2+}	(mol/kg)	3.22E-07	-
Fe^{3+}	(mol/kg)	1.07E-24	-
Mg^{2+}	(mol/kg)	1.07E-03	-
Mn^{2+}	(mol/kg)		-
Na^{+}	(mol/kg)	2.68E-01	9.54E-04
K^{+}	(mol/kg)	4.21E-04	1.68E-05
Sr^{2+}	(mol/kg)		-
NH_4^{+}	(mol/kg)	1.06E-03	0.00E+00
∑ competitive cations in solution with Na^{+} (C-cmp/a)	(mol/kg)	2.70E-01	1.06E-03
Al^{3+}	(mol/kg)		
Ca^{2+}	(mol/kg)	4.15E-05	9.33E-05
Fe^{2+}	(mol/kg)	3.22E-07	-
Fe^{3+}	(mol/kg)	1.07E-24	-
Mg^{2+}	(mol/kg)	1.07E-03	-
Mn^{2+}	(mol/kg)		-
Na^{+}	(mol/kg)	2.68E-01	9.54E-04
K^{+}	(mol/kg)	4.21E-04	1.68E-05
Sr^{2+}	(mol/kg)		-
NH_4^{+}	(mol/kg)	1.06E-03	0.00E+00
∑ competitive cations in solution without Na^{+} (C-cmp/b)	(mol/kg)	2.59E-03	1.10E-04

K_d-derivation Cs(I) on Kunigel-V1 Sato and Shibutani (1994)

Overview Pred. K_d [m^3/kg]	Mudstone
	HDB-6
Pred. K_d: CF-1 (% clay)	0.54
Pred. K_d: CF-3a (%clay)	2.11E-03

Cs(I) K_d-derivation (Smectite)		Conditions		Conversion (C) / Uncertainty (U)
		Data source[1]	Application[2]	CF
			Mudstone	Mudstone
Solution[5]		Dist. equil. water	HDB-6	HDB-6
Substrate		Kunigel-V1	Mudstone	Mudstone
Experimental K_d	(m^3/kg)	1.30		
Conversion Mineralogy (CF-min)				
CEC[4]	(meq/100g)	60.10	13.75	0.23
BET[6]	(m^2/g)	35.00	5.55	0.16
% clay[3]		46.00	19.00	0.41
Conversion pH (CF-pH)	*irrelevant*	8.50	8.50	
Conversion Speciation (CF-spec)	*irrelevant*			
Conversion Competition (Cationic) (CF-cmp)				
∑ competitive cations in soln. with Na^+ (CF-cmp/a)	(mol/l)	1.06E-03	2.70E-01	3.94E-03
∑ competitive cations in soln. without Na^+ (CF-cmp/b)	*not considered*			

(1) K_d value from JAEA-SDB database, Sato and Shibutani (1994), [Cs] = 7.52E-5 M
(2) Values of the average composition
(3) clay mineral composition of data source from Sasaki et al. (1995): smectite 46-49%, quartz 29-38% and other trace impurities
(4) CEC value of data source from Sasaki et al. (1995)
(5) [Na^+] calculated with Phreeqci Ion Exchange with Kunigel V1 equilibrated in distilled water (S:W = 2 g/l)
(6) Value from Dixon and Weed (1977)

Cs(I) K_d-derivation via % clay		Mudstone	CF
CF-1 (% clay)			0.41
Competition			
CF-3 (% clay)			1.63E-03
			UF
Predicted Kd: CF-1 (% clay)	(m^3/kg)	**0.54**	
UF-starting K_d			1.6
UF-min			1.4
UF-total			2.2
K_d upper limit	(m^3/kg)	1.19	
K_d lower limit	(m^3/kg)	0.24	
Predicted Kd: CF-3 (% clay)	(m^3/kg)	**2.11E-03**	
UF-starting K_d			1.6
UF-cmp			1.2
UF-min			1.4
UF-total			2.7
K_d upper limit	(m^3/kg)	5.63E-03	
K_d lower limit	(m^3/kg)	7.94E-04	

Speciation Case III

Filename Cs_equilibrium_case_III_120109_DAT Gorgeon(1994)_Cs_NAPSI
Preequilibration_case_III_120109_DAT

TDB: JAEA, version 100331c1

Cs(I) speciation (Gorgeon, 1994)

System		Application	Data Source
Substrate		Horonobe mudstone	Na-Illite
Solution		HDB-6	1 M NaCl
pH		8.5	8.5
pCO_2		-	-
CEC		13.75	122.00
BET		5.55	97.00
Cs^{1+} dissolved	(mol/kg)	9.83E-12	2.00E-02
Al^{3+}	(mol/kg)		
Ca^{2+}	(mol/kg)	4.17E-05	
Fe^{2+}	(mol/kg)	3.24E-07	
Fe^{3+}	(mol/kg)	1.08E-24	
Mg^{2+}	(mol/kg)	8.60E-04	
Mn^{2+}	(mol/kg)		
Na^{+}	(mol/kg)	2.77E-01	1.00E+00
K^{+}	(mol/kg)	3.08E-04	0.00E+00
Sr^{2+}	(mol/kg)		
NH_4^{+}	(mol/kg)	6.68E-04	0.00E+00
∑ competitive cations in solution with Na^{+} (C-cmp/a)	(mol/kg)	2.79E-01	1.00E+00
Al^{3+}	(mol/kg)		
Ca^{2+}	(mol/kg)	4.17E-05	
Fe^{2+}	(mol/kg)	3.24E-07	
Fe^{3+}	(mol/kg)	1.08E-24	
Mg^{2+}	(mol/kg)	8.60E-04	
Mn^{2+}	(mol/kg)		
Na^{+}	(mol/kg)	2.77E-01	1.00E+00
K^{+}	(mol/kg)	3.08E-04	0.00E+00
Sr^{2+}	(mol/kg)		
NH_4^{+}	(mol/kg)	6.68E-04	0.00E+00
∑ competitive cations in solution without Na^{+} (C-cmp/b)	(mol/kg)	1.88E-03	0.00E+00

K_d-derivation Cs(I) Gorgeon (1994)

Overview Pred. K_d [m^3/kg]	Mudstone
	HDB-6
Pred. K_d: CF-1 (% clay)	0.86
Pred. K_d: CF-3a (% clay)	3.07

Cs(I) K_d-derivation		Conditions		Conversion (C) / Uncertainty (U)
		Data source[1]	Application	CF
			Mudstone	Mudstone
Solution		1 M NaCl	HDB-6	HDB-6
Substrate		Illite	Mudstone	Mudstone
Experimental K_d	(m^3/kg)	3.30		
Conversion Mineralogy (CF-min)				
CEC[2]	(meq/100g)	21.2	19.61	0.93
BET[2]	(m^2/g)	111.5	5.55	0.05
% clay		100	26.00	0.26
Conversion pH (CF-pH)				
pH		8.5	8.50	1.00
Conversion Speciation (CF-spec)	*irrelevant*			
Conversion Competition (Cationic) (CF-cmp)				
∑ competitive cations in soln. with Na^+ (CF-cmp/a)	(mol/l)	1.00	0.28	3.58
∑ competitive cations in soln. without Na^+ (CF-cmp/b)	*not considered*			

(1) K_d values from data table Gorgeon (1994) given as log Kd
(2) Values of the avarage composition (Table Mineralogy 1)

Cs(I) K_d-derivation via % clay			Mudstone	CF
CF-1 (% clay)				0.26
Competition				
CF-3 (% clay)				0.93
				UF
Predicted K_d: CF-1 (% clay)	(m^3/kg)		**0.86**	
UF-starting K_d				1.6
UF-pH conversion				2.5
UF-min				1.4
UF-total				5.5
K_d upper limit	(m^3/kg)		4.76	
K_d lower limit	(m^3/kg)		0.15	
Predicted K_d: CF-3 (% clay)	(m^3/kg)		**3.07**	
UF-starting K_d				1.6
UF-pH conversion				2.5
UF-cmp				1.2
UF-min				1.4
UF-total				6.7
K_d upper limit	(m^3/kg)		20.46	
K_d lower limit	(m^3/kg)		0.46	

Speciation Case III

Filename	Cs_equilibrium_case_III_120109_DAT	PSI(1999)_0.1MNaClO4_Cs_NAPSI
	Preequilibration_case_III_120109_DAT	PSI(1999)_0.01MNaClO4_Cs_NAPSI
TDB:	JAEA, version 100331c1	

Cs(I) speciation (Poinssot et al., 1999)

System		Application	Data Source	
Substrate		Horonobe mudstone	Na-Illite	
Solution		HDB-6	0.01 M $NaClO_4$	0.1 M $NaClO_4$
pH		8.5	8.5	8.5
pCO_2		-	-	-
CEC		13.75	122.00	122.00
BET		5.55	97.00	97.00
Cs^{1+} dissolved	(mol/kg)	9.83E-12	1.00E-08	1.00E-08
Al^{3+}	(mol/kg)			
Ca^{2+}	(mol/kg)	4.17E-05		
Fe^{2+}	(mol/kg)	3.24E-07		
Fe^{3+}	(mol/kg)	1.08E-24		
Mg^{2+}	(mol/kg)	8.60E-04		
Mn^{2+}	(mol/kg)			
Na^{+}	(mol/kg)	2.77E-01	1.00E-02	1.00E-01
K^{+}	(mol/kg)	3.08E-04	4.70E-06	1.50E-05
Sr^{2+}	(mol/kg)			
NH_4^{+}	(mol/kg)	6.68E-04	0.00E+00	0.00E+00
∑ competitive cations in solution with Na^{+} (CF-cmp/a)	(mol/kg)	2.79E-01	1.00E-02	1.00E-01
Al^{3+}	(mol/kg)			
Ca^{2+}	(mol/kg)	4.17E-05		
Fe^{2+}	(mol/kg)	3.24E-07		
Fe^{3+}	(mol/kg)	1.08E-24		
Mg^{2+}	(mol/kg)	8.60E-04		
Mn^{2+}	(mol/kg)			
Na^{+}	(mol/kg)	2.77E-01	1.00E-02	1.00E-01
K^{+}	(mol/kg)	3.08E-04	4.70E-06	1.50E-05
Sr^{2+}	(mol/kg)			
NH_4^{+}	(mol/kg)	6.68E-04	0.00E+00	0.00E+00
∑ competitive cations in solution without Na^{+} (CF-cmp/b)	(mol/kg)	1.88E-03	4.70E-06	1.50E-05

K_d-derivation Cs(I) on Illite Poinssot et al., (1999)

Overview Pred. K_d [m^3/kg]	A	B
	Mudstone	Mudstone
	HDB-6	HDB-6
Pred. K_d: CF-1 (% clay)	43.55	13.06
Pred. K_d: CF-3a (% clay)	1.56	4.68

Cs(I) K_d-derivation (Illite)		Conditions			Conversion (C) / Uncertainty (U)	
		Data source[1]		Application[2]	CF for A	CF for B
		A	B	Mudstone	Mudstone	Mudstone
Solution		0.01 M $NaClO_4$	0.1 M $NaClO_4$	HDB-6	HDB-6	HDB-6
Substrate		Na-Illite[3]	Na-Illite[3]	Mudstone	Mudstone	Mudstone
Experimental K_d	(m^3/kg)	150.00	45.00			
Conversion Mineralogy (CF-min)						
CEC[5]	(meq/100g)	122.00	122.00	19.61		
BET[4]	(m^2/g)	97.00	97.00	5.55		
% clay		93.00	93.00	27.00	0.29	0.29
% Illite		93.00	93.00	7.00	0.08	0.08
Conversion pH (CF-pH)	*irrelevant*	8.50	8.50	8.50		
Conversion Speciation (CF-spec)	*irrelevant*					
Conversion Competition (Cationic) (CF-cmp)						
∑ competitive cations in soln. with Na^+ (CF-cmp/a)	(mol/l)	0.01	0.10	0.28	0.04	0.36
∑ competitive cations in soln. without Na^+ (CF-cmp/b)	(mol/l)	4.70E-06	1.50E-05	1.88E-03	2.50E-03	7.99E-03

(1) K_d value from Cs(I) sorption datapoint from Poinssot et al. (1999), PSI Report; Kd value from graph at pH = 6.63; [Cs] = 1.0E-8 M
(2) Values of the avarage composition (Table Mineralogy 1)
(3) Conditioned Na-Illite; clay mineral composition: 93% illite and 7% kaolinite
(4) BET value of non-treated illite
(5) CEC value of treated Na-illite; pH 7.1

		A	B		
Cs(I) K_d-derivation via % clay		Mudstone	Mudstone	CF for A	CF for B
CF-1 (% clay)				0.29	0.29
Competition					
CF-3 (% clay)				0.01	0.10
				UF	UF
Predicted K_d: CF-1 (% clay)	(m^3/kg)	**43.55**	**13.06**		
UF-starting K_d				1.6	1.6
UF-min				1.4	1.4
UF-total				2.2	2.2
K_d upper limit	(m^3/kg)	96.63	28.99		
K_d lower limit	(m^3/kg)	19.63	5.89		
Predicted K_d: CF-3 (% clay)		**1.56**	**4.68**		
UF-starting K_d				1.6	1.6
UF-cmp				1.2	1.2
UF-min				1.4	1.4
UF-total				2.7	2.7
K_d upper limit		4.16	12.46		
K_d lower limit		0.59	1.76		

Speciation Case III

Filename Cs_equilibrium_case_III_120109_DAT PW_Sato&Shibutani(1994)_NAPSI
Preequilibration_case_III_120109_DAT

TDB: JAEA, version 100331c1

Cs(I) speciation (Sato & Shibutani, 1994)

System		Application	Data Source
Substrate		Horonobe mudstone	Kunigel V1
Solution		HDB-6	Dist. equil. water
pH		8.5	8.5
pCO_2		-	-
CEC		13.75	60.10
BET		5.55	-
Cs^{1+} dissolved	(mol/kg)	9.83E-12	7.52E-05
Al^{3+}	(mol/kg)		
Ca^{2+}	(mol/kg)	4.17E-05	9.33E-05
Fe^{2+}	(mol/kg)	3.24E-07	-
Fe^{3+}	(mol/kg)	1.08E-24	-
Mg^{2+}	(mol/kg)	8.60E-04	-
Mn^{2+}	(mol/kg)		-
Na^{+}	(mol/kg)	2.77E-01	9.54E-04
K^{+}	(mol/kg)	3.08E-04	1.68E-05
Sr^{2+}	(mol/kg)		-
NH_4^{+}	(mol/kg)	6.68E-04	0.00E+00
∑ competitive cations in solution with Na^{+} (C-cmp/a)	(mol/kg)	2.79E-01	1.06E-03
Al^{3+}	(mol/kg)		
Ca^{2+}	(mol/kg)	4.17E-05	9.33E-05
Fe^{2+}	(mol/kg)	3.24E-07	-
Fe^{3+}	(mol/kg)	1.08E-24	-
Mg^{2+}	(mol/kg)	8.60E-04	-
Mn^{2+}	(mol/kg)		-
Na^{+}	(mol/kg)	2.77E-01	9.54E-04
K^{+}	(mol/kg)	3.08E-04	1.68E-05
Sr^{2+}	(mol/kg)		-
NH_4^{+}	(mol/kg)	6.68E-04	0.00E+00
∑ competitive cations in solution without Na^{+} (C-cmp/b)	(mol/kg)	1.88E-03	1.10E-04

K_d-derivation Cs(I) on Kunigel-V1 Sato and Shibutani (1994)

Overview Pred. K_d [m^3/kg]	Mudstone
	HDB-6
Pred. K_d: CF-1 (% clay)	0.73
Pred. K_d: CF-3a (%clay)	2.80E-03

Cs(I) K_d-derivation (Smectite)		Conditions		Conversion (C) / Uncertainty (U)
		Data source[1]	Application[2]	CF
			Mudstone	Mudstone
Solution[5]		Dist. equil. water	HDB-6	HDB-6
Substrate		Kunigel-V1	Mudstone	Mudstone
Experimental K_d	(m^3/kg)	1.30		
Conversion Mineralogy (CF-min)				
CEC[4]	(meq/100g)	60.10	19.61	0.33
BET[6]	(m^2/g)	35.00	5.55	0.16
% clay[3]		46.00	26.00	0.57
Conversion pH (CF-pH)	*irrelevant*	8.5	8.5	
Conversion Speciation (CF-spec)	*irrelevant*			
Conversion Competition (Cationic) (CF-cmp)				
∑ competitive cations in soln. with Na^+ (CF-cmp/a)	(mol/l)	1.06E-03	2.79E-01	3.81E-03
∑ competitive cations in soln. without Na^+ (CF-cmp/b)	*not considered*			

(1) K_d value from SDB database, Sato and Shibutani (1994), [Cs] = 7.52E-5 M
(2) Values of the avarage composition (Table Mineralogy 1)
(3) clay mineral composition of data source from Sasaki et al. (1995): smectite 46-49%, quartz 29-38% and other trace impurities
(4) CEC value of data source from Sasaki et al. (1995)
(5) [Na^+] calculated with Phreeqci Ion Exchange with Kunigel V1 equilibrated in distilled water (S:W = 2 g/l)
(6)Value from 'Minerals in Soil Environments; J.B. Dixon and S.B. Weed; Soil Science Society of America'

Cs(I) K_d-derivation via % clay		Mudstone	CF
CF-1 (% clay)			0.57
Competition			
CF-3 (% clay)			2.16E-03
			UF
Predicted Kd: CF-1 (% clay)	(m^3/kg)	**0.73**	
UF-starting K_d			1.6
UF-min			1.4
UF-total			2.2
K_d upper limit	(m^3/kg)	1.63	
K_d lower limit	(m^3/kg)	0.33	
Predicted Kd: CF-3 (% clay)	(m^3/kg)	**2.80E-03**	
UF-starting K_d			1.6
UF-cmp			1.2
UF-min			1.4
UF-total			2.7
K_d upper limit	(m^3/kg)	7.46E-03	
K_d lower limit	(m^3/kg)	1.05E-03	

Speciation Case I

Filename Ni1_equilibrium_case_I_120605_DAT Tertre 2005_solution speciation
Preequilibration_case_I_120511_DAT

TDB: JAEA, version 100331c1

Ni(II) speciation (Tertre et al., 2005)

System		Application	Data Source
Substrate		Horonobe mudstone	MX-80
Solution		HDB-6	0.025M $NaClO_4$
pH		6.752	6.75
pCO_2		-	-3.97
Ni(II) dissolved	(mol/kg)	7.21E-10	only hydrolysis relevant
Ni^{2+}	(mol/kg)	5.27E-10	
$NiOH^+$	(mol/kg)	3.40E-13	
$Ni(OH)_2$	(mol/kg)	4.28E-15	
$NiHCO_3^+$	(mol/kg)	1.35E-10	
$NiCO_3$	(mol/kg)	1.51E-11	
$NiCl^+$	(mol/kg)	4.30E-11	
$NiNH_3^{2+}$	(mol/kg)	2.00E-13	
NiF^+	(mol/kg)	4.35E-14	
$NiHPO_4$	(mol/kg)	2.50E-14	
NiS	(mol/kg)	4.07E-13	
$NiHS^+$	(mol/kg)	2.98E-15	
∑ competitive Nicomplexes (Ni-cmp) with Ni-CO_3	(mol/kg)	1.94E-10	0.00E+00
Ni tot-(Ni-cmp)	(mol/kg)	5.27E-10	1.00E+00
(Ni tot- (Ni-cmp)) / Ni tot (CF-spec/a)		**7.31E-01**	**1.00E+00**
$NiHCO_3^+$	(mol/kg)	1.35E-10	
$NiCO_3$	(mol/kg)	1.51E-11	
$NiCl^+$	(mol/kg)	4.30E-11	
$NiNH_3^{2+}$	(mol/kg)	2.00E-13	
NiF^+	(mol/kg)	4.35E-14	
$NiHPO_4$	(mol/kg)	2.50E-14	
NiS	(mol/kg)	4.07E-13	
$NiHS^+$	(mol/kg)	2.98E-15	
∑ competitive Ni complexes (Ni-cmp) without Ni-CO_3	(mol/kg)	4.37E-11	0.00E+00
Ni tot-(Ni-cmp) without Ni-CO_3	(mol/kg)	6.77E-10	1.00E+00
(Ni tot- (Ni-cmp)) / Ni tot (CF-spec/b)		**9.39E-01**	**1.00E+00**

K_d-Derivation Ni(II) on MX-80, pH 6.75, Tertre et al. (2005)

Overview Pred. K_d [m^3/kg]	Mudstone
	HDB-6
Pred. K_d: CF-1 (% clay)	0.057
Pred. K_d: CF-2a (% clay)	0.042

Ni(II) K_d-derivation		Conditions		Conversion (C) / Uncertainty (U)
		Data source[1]	Application	CF
Solution		0.025M $NaClO_4$	HDB-6	HDB-6
Substrate		MX-80	Mudstone	Mudstone
Experimental K_d	(m^3/kg)	0.300		
Conversion Mineralogy (CF-min)				
%clay		100	19	0.19
Conversion Speciation (CF-spec)				
CF-spec/a		1.00	0.73	0.73
CF-spec/b		1.00	0.94	0.94
Conversion Competition (Anionic or Cationic) (CF-cmp)	*irrelevant*			

(1) K_d values from Ni(II) sorption edge

Ni(II) K_d-derivation via % clay				CF
CF-1 (%clay)				0.19
Speciation				
CF-2a (% clay)				0.14
CF-2b (% clay)				0.18
				UF
Predicted K_d: CF-1 (% clay)	(m^3/kg)		**0.057**	
UF-starting K_d				1.6
UF-pH conversion				2.5
UF-min				1.4
UF-total				5.5
K_d upper limit	(m^3/kg)		0.32	
K_d lower limit	(m^3/kg)		0.01	
Predicted K_d: CF-2a (% clay)	(m^3/kg)		**0.042**	
UF-starting K_d				1.6
UF-pH conversion				2.5
UF-min				1.4
UF-speciation				1.4
UF-total				7.8
K_d upper limit	(m^3/kg)		0.32	
K_d lower limit	(m^3/kg)		0.0054	
Predicted K_d: CF-2b (% clay)	(m^3/kg)		**0.054**	
UF-starting K_d				1.6
UF-pH conversion				2.5
UF-min				1.4
UF-speciation				1.4
UF-total				7.8
K_d upper limit	(m^3/kg)		0.42	
K_d lower limit	(m^3/kg)		0.01	

Speciation Case I

Filename Ni1_equilibrium_case_I_120605_DAT B&B 1999_solution speciation
Preequilibration_case_I_120511_DAT

TDB: JAEA, version 100331c1

Ni(II) speciation (Bradbury&Baeyens, 1999)

System		Application	Data Source
Substrate		**Horonobe mudstone**	**Ca-Montmorillonite**
Solution		**HDB-6**	**$Ca(NO_3)_2$ solution**
Ni(II) added	(mol/kg)	1.30E-07	2.70E-08
pH		6.75	6.75
pCO_2		-	-
Ni(II) dissolved	(mol/kg)	7.21E-10	only hydrolysis considered [1)]
Ni^{2+}	(mol/kg)	5.27E-10	
$NiOH^+$	(mol/kg)	3.40E-13	
$Ni(OH)_2$	(mol/kg)	4.28E-15	
$NiHCO_3^+$	(mol/kg)	1.35E-10	
$NiCO_3$	(mol/kg)	1.51E-11	
$NiCl^+$	(mol/kg)	4.30E-11	
$NiNH_3^{2+}$	(mol/kg)	2.00E-13	
NiF^+	(mol/kg)	4.35E-14	
$NiHPO_4$	(mol/kg)	2.50E-14	
NiS	(mol/kg)	4.07E-13	
$NiHS^+$	(mol/kg)	2.98E-15	
∑ competitive Nicomplexes (Ni-cmp) with Ni-CO_3	(mol/kg)	1.94E-10	0.00E+00
Ni tot-(Ni-cmp)	(mol/kg)	5.27E-10	2.70E-08
(Ni tot- (Ni-cmp)) / Ni tot (CF-spec/a)		**7.31E-01**	**1.00E+00**
$NiHCO_3^+$	(mol/kg)	1.35E-10	
$NiCO_3$	(mol/kg)	1.51E-11	
$NiCl^+$	(mol/kg)	4.30E-11	
$NiNH_3^{2+}$	(mol/kg)	2.00E-13	
NiF^+	(mol/kg)	4.35E-14	
$NiHPO_4$	(mol/kg)	2.50E-14	
NiS	(mol/kg)	4.07E-13	
$NiHS^+$	(mol/kg)	2.98E-15	
∑ competitive Ni complexes (Ni-cmp) without Ni-CO_3	(mol/kg)	4.37E-11	0.00E+00
Ni tot-(Ni-cmp) without Ni-CO_3	(mol/kg)	6.77E-10	2.70E-08
(Ni tot- (Ni-cmp)) / Ni tot (CF-spec/b)		**9.39E-01**	**1.00E+00**

[1)] Ni-NO3 complexes are not relevant (ca. 1E-20 M) according to the JAEA-TDB

K_d-Derivation Ni(II) on Ca-Montmorillonite, pH 6.75, Bradbury&Baeyens (1999)

Overview Pred. K_d [m^3/kg]	Horonobe
	HDB-6
Pred. K_d: CF-1 (% clay)	0.15
Pred. K_d: CF-2a (% clay)	0.11

Ni(II) K_d-derivation		Conditions		Conversion (C) / Uncertainty (U)
		Data source[1]	Application	CF
Solution		0.033M $Ca(NO_3)_2$	HDB-6	HDB-6
Substrate		Ca-Montmorillonite	Mudstone	Mudstone
Experimental K_d	(m^3/kg)	0.8		
Conversion Mineralogy (CF-min)				
%clay		100	19	0.19
Conversion Speciation (CF-spec)				
CF-spec/a		1.00	0.73	0.73
CF-spec/b		1.00	0.94	0.94
Conversion Competition (Anionic or Cationic) (CF-cmp)	*irrelevant*			

(1) K_d values from Ni(II) sorption edge

Ni(II) K_d-derivation via % clay				CF
CF-1 (%clay)				0.19
Speciation				
CF-2a (% clay)				0.14
CF-2b (% clay)				0.18
				UF
Predicted K_d: CF-1 (% clay)	(m^3/kg)		**0.15**	
UF-starting K_d				1.6
UF-pH conversion				2.5
UF-min				1.4
UF-total				5.5
K_d upper limit	(m^3/kg)		0.84	
K_d lower limit	(m^3/kg)		0.03	
Predicted K_d: CF-2a (% clay)	(m^3/kg)		**0.11**	
UF-starting K_d				1.6
UF-pH conversion				2.5
UF-min				1.4
UF-speciation				1.4
UF-total				7.8
K_d upper limit	(m^3/kg)		0.86	
K_d lower limit	(m^3/kg)		0.014	
Predicted K_d: CF-2b (% clay)	(m^3/kg)		**0.14**	
UF-starting K_d				1.6
UF-pH conversion				2.5
UF-min				1.4
UF-speciation				1.4
UF-total				7.8
K_d upper limit	(m^3/kg)		1.10	
K_d lower limit	(m^3/kg)		0.02	

Speciation Case I

Filename Ni1_equilibrium_case_I_120605_DAT N1_Lauber_2000_sol_spec_pH6.3_120619_DAT
Preequilibration_case_I_120511_DAT

TDB: JAEA, version 100331c1

Ni speciation (Lauber et al., (2000)

System		Application	Data Source
Substrate		Horonobe Mudstone	OPA
Solution		HDB-6	synthetic OPA porewater
pH		6.752	6.30
pCO_2		-	-
Ni(II) dissolved	(mol/kg)	7.21E-10	3.76E-07
Ni^{2+}	(mol/kg)	5.27E-10	2.56E-07
$NiOH^+$	(mol/kg)	3.40E-13	6.56E-11
$Ni(OH)_2$	(mol/kg)	4.28E-15	3.28E-13
$NiHCO_3^+$	(mol/kg)	1.35E-10	2.00E-08
$NiCO_3$	(mol/kg)	1.51E-11	8.84E-10
$NiCl^+$	(mol/kg)	4.30E-11	1.76E-08
$NiNH_3^{2+}$	(mol/kg)	2.00E-13	
NiF^+	(mol/kg)	4.35E-14	2.17E-10
$NiHPO_4$	(mol/kg)	2.50E-14	
NiS	(mol/kg)	4.07E-13	1.95E-12
$NiHS^+$	(mol/kg)	2.98E-15	3.59E-14
$NiSO_4$	(mol/kg)		7.68E-08
$Ni(SO_4)_2^{2-}$	(mol/kg)		4.88E-09
∑ competitive Ni complexes (Ni-cmp) with $Ni\text{-}CO_3$	(mol/kg)	1.94E-10	1.20E-07
Ni tot-(Ni-cmp)	(mol/kg)	5.27E-10	2.56E-07
(Ni tot- (Ni-cmp)) / Ni tot (CF-spec/a)		**7.31E-01**	**6.80E-01**
$NiHCO_3^+$	(mol/kg)	1.35E-10	2.00E-08
$NiCO_3$	(mol/kg)	1.51E-11	8.84E-10
$NiCl^+$	(mol/kg)	4.30E-11	1.76E-08
$NiNH_3^{2+}$	(mol/kg)	2.00E-13	
NiF^+	(mol/kg)	4.35E-14	2.17E-10
$NiHPO_4$	(mol/kg)	2.50E-14	
NiS	(mol/kg)	4.07E-13	1.95E-12
$NiHS^+$	(mol/kg)	2.98E-15	3.59E-14
$NiSO_4$	(mol/kg)		7.68E-08
$Ni(SO_4)_2^{2-}$	(mol/kg)		4.88E-09
∑ competitive Ni complexes (Ni-cmp) without $Ni\text{-}CO_3$	(mol/kg)	4.37E-11	9.95E-08
Ni tot-(Ni-cmp) without $Ni\text{-}CO_3$	(mol/kg)	6.77E-10	2.77E-07
(Ni tot- (Ni-cmp)) / Ni tot (CF-spec/b)		**9.39E-01**	**7.35E-01**

K_d-Derivation Ni(II) on opalinus clay, pH 6.3, Lauber 2000

Overview Pred. K_d [m^3/kg]	Mudstone
	HDB-6
Pred. K_d: CF-1 (% clay)	0.14
Pred. K_d: CF-2a (% clay)	0.15

Ni(II) K_d-derivation		Conditions				Conversion (C) / Uncertainty (U)
		Data source[1]	Additional information[3]		Application[2]	CF
Solution		synthetic porewater	0.1M $NaClO_4$		HDB-6	HDB-6
Substrate		OPA	SWy-1	SWy-1	Mudstone	Mudstone
Experimental K_d	(m^3/kg)	0.3				
Conversion Mineralogy (CF-min)						
%clay		70			19	0.27
Conversion pH (CF-pH)						
pH		6.3	6.3	6.75	6.752	
Kd for 0.1 M NaClO4 (pH conversion)[3]		0.3	0.8	1.3	0.10	1.58
Conversion Speciation (CF-spec)						
CF-spec/a		0.68			0.73	1.08
CF-spec/b		0.74			0.94	1.28
Conversion Competition (Anionic or Cationic) (CF-cmp)	*irrelevant*					

(1) K_d values from Lauber et al. (2000)
(2) Values of the avarage composition (Table Mineralogy 1)
(3) K_d values from Ni sorption edge from Bradbury and Baeyens (2003)

Ni(II) K_d-derivation via % clay						CF
CF-1 (%clay)						0.43
Speciation						
CF-2a (% clay)						0.46
CF-2b (% clay)						0.55
						UF
Predicted K_d: CF-1 (% clay)	(m^3/kg)				**0.14**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-total						5.5
K_d upper limit	(m^3/kg)				0.75	
K_d lower limit	(m^3/kg)				0.02	
Predicted K_d: CF-2a (% clay)	(m^3/kg)				**0.15**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-speciation						1.4
UF-total						7.8
K_d upper limit	(m^3/kg)				1.13	
K_d lower limit	(m^3/kg)				0.019	
Predicted K_d: CF-2b (% clay)	(m^3/kg)				**0.17**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-speciation						1.4
UF-total						7.8
K_d upper limit	(m^3/kg)				1.34	
K_d lower limit	(m^3/kg)				0.02	

Speciation Case II

Filename Ni1_equilibrium_case_II_120605_DAT Tertre 2005_solution speciation
Preequilibration_case_II_120509_DAT

TDB: JAEA,version 100331c1

Ni(II) speciation (Tertre et al., 2005)

System		Application	Data Source
Substrate		Horonobe mudstone	MX-80
Solution		HDB-6	0.025M $NaClO_4$
pH		8.5	8.50
pCO_2		-	-3.97
Ni(II) dissolved	(mol/kg)	7.34E-11	only hydrolysis relevant
Ni^{2+}	(mol/kg)	2.12E-11	
$NiOH^+$	(mol/kg)	7.68E-13	
$Ni(OH)_2$	(mol/kg)	5.42E-13	
Ni_2OH^{3+}	(mol/kg)		
$Ni(OH)_3^-$	(mol/kg)	1.65E-15	
$NiHCO_3^+$	(mol/kg)	4.07E-12	
$NiCO_3$	(mol/kg)	2.54E-11	
$NiCl^+$	(mol/kg)	1.67E-12	
NiS	(mol/kg)	1.95E-11	
$NiHS^+$	(mol/kg)	2.55E-15	
$NiNH_3^{2+}$	(mol/kg)	2.85E-13	
$Ni(NH_3)_2^{2+}$	(mol/kg)	1.18E-15	
∑ competitive Ni complexes (Ni-cmp) with Ni-CO_3	(mol/kg)	5.092E-11	0.00E+00
Ni tot-(Ni-cmp)	(mol/kg)	2.25E-11	1.70E-06
(Ni tot- (Ni-cmp)) / Ni tot (CF-spec/a)		**3.07E-01**	**1.00E+00**
$NiHCO_3^+$	(mol/kg)	4.07E-12	
$NiCO_3$	(mol/kg)	2.54E-11	
$NiCl^+$	(mol/kg)	1.67E-12	
NiS	(mol/kg)	1.95E-11	
$NiHS^+$	(mol/kg)	2.55E-15	
$NiNH_3^{2+}$	(mol/kg)	2.85E-13	
$Ni(NH_3)_2^{2+}$	(mol/kg)	1.18E-15	
∑ competitive Ni complexes (Ni-cmp) without Ni-CO_3	(mol/kg)	2.15E-11	0.00E+00
Ni tot-(Ni-cmp) without Ni-CO_3	(mol/kg)	5.20E-11	1.70E-06
(Ni tot- (Ni-cmp)) / Ni tot (CF-spec/b)		**7.08E-01**	**1.00E+00**

K_d-Derivation Ni(II) on MX-80, pH 6.75, Tertre et al. (2005)

Overview Pred. K_d [m^3/kg]	Mudstone
	HDB-6
Pred. K_d: CF-1 (% clay)	0.86
Pred. K_d: CF-2a (% clay)	0.26

Ni(II) K_d-derivation		Conditions		Conversion (C) / Uncertainty (U)
		Data source[1]	Application	CF
Solution		0.025M $NaClO_4$	HDB-6	HDB-6
Substrate		MX-80	Mudstone	Mudstone
Experimental K_d	(m^3/kg)	4.500		
Conversion Mineralogy (CF-min)				
%clay		100	19	0.19
Conversion Speciation (CF-spec)				
CF-spec/a		1.00	0.31	0.31
CF-spec/b		1.00	0.71	0.71
Conversion Competition (Anionic or Cationic) (CF-cmp)	*irrelevant*			

(1) K_d values from Ni(II) sorption edge

Ni(II) K_d-derivation via % clay				CF
CF-1 (%clay)				0.19
Speciation				
CF-2a (% clay)				0.06
CF-2b (% clay)				0.13
				UF
Predicted K_d: CF-1 (% clay)	(m^3/kg)		**0.86**	
UF-starting K_d				1.6
UF-pH conversion				2.5
UF-min				1.4
UF-total				5.5
K_d upper limit	(m^3/kg)		4.74	
K_d lower limit	(m^3/kg)		0.15	
Predicted K_d: CF-2a (% clay)	(m^3/kg)		**0.26**	
UF-starting K_d				1.6
UF-pH conversion				2.5
UF-min				1.4
UF-speciation				1.4
UF-total				7.8
K_d upper limit	(m^3/kg)		2.04	
K_d lower limit	(m^3/kg)		0.034	
Predicted K_d: CF-2b (% clay)	(m^3/kg)		**0.60**	
UF-starting K_d				1.6
UF-pH conversion				2.5
UF-min				1.4
UF-speciation				1.4
UF-total				7.8
K_d upper limit	(m^3/kg)		4.70	
K_d lower limit	(m^3/kg)		0.08	

Speciation Case II

Filename Ni1_equilibrium_case_II_120605_DAT B&B 1999_solution speciation
Preequilibration_case_II_120509_DAT

TDB: JAEA,version 100331c1

Ni(II) speciation (Bradbury&Baeyens, 1999)

System		Application	Data Source
Substrate		Horonobe mudstone	Ca-Montmorillonite
Solution		HDB-6	$Ca(NO_3)_2$ solution
$Ni(II)_{initial}$	(mol/kg)	1.30E-07	2.70E-08
pH		8.5	8.5
pCO_2		-	-
Ni(II) dissolved	(mol/kg)	7.34E-11	only hydrolysis considered [1)]
Ni^{2+}	(mol/kg)	2.12E-11	
$NiOH^+$	(mol/kg)	7.68E-13	
$Ni(OH)_2$	(mol/kg)	5.42E-13	
$Ni(OH)_3^-$	(mol/kg)	1.65E-15	
$NiHCO_3^+$	(mol/kg)	4.07E-12	
$NiCO_3$	(mol/kg)	2.54E-11	
$NiCl^+$	(mol/kg)	1.67E-12	
NiS	(mol/kg)	1.95E-11	
$NiHS^+$	(mol/kg)	2.55E-15	
$NiNH_3^{2+}$	(mol/kg)	2.85E-13	
$Ni(NH_3)_2^{2+}$	(mol/kg)	1.18E-15	
∑ competitive Ni complexes (Ni-cmp) with $Ni\text{-}CO_3$	(mol/kg)	5.09E-11	0.00E+00
Ni tot-Ni-cmp)	(mol/kg)	2.25E-11	2.70E-08
(Ni tot- (Ni-cmp)) / Ni tot (CF-spec/a)		**3.07E-01**	**1.00E+00**
$NiHCO_3^+$	(mol/kg)	4.07E-12	
$NiCO_3$	(mol/kg)	2.54E-11	
$NiCl^+$	(mol/kg)	1.67E-12	
NiS	(mol/kg)	1.95E-11	
$NiHS^+$	(mol/kg)	2.55E-15	
$NiNH_3^{2+}$	(mol/kg)	2.85E-13	
$Ni(NH_3)_2^{2+}$	(mol/kg)	1.18E-15	
Ni complexes (Ni-cmp) without $Ni\text{-}CO_3$	(mol/kg)	2.15E-11	0.00E+00
Ni tot-(Ni-cmp) without $Ni\text{-}CO_3$	(mol/kg)	5.20E-11	2.70E-08
(Ni tot- (Ni-cmp)) / Ni tot (CF-spec/b)		**7.075E-01**	**1.000E+00**

[1)] Ni-NO3 complexes are not relevant (ca. 1E-20 M) according to the JAEA-TDB

K_d-Derivation Ni(II) on Ca-Montmorillonite, pH 6.75, Bradbury&Baeyens (1999)

Overview Pred. K_d [m^3/kg]	Horonobe
	HDB-6
Pred. K_d: CF-1 (% clay)	1.07
Pred. K_d: CF-2a (% clay)	0.33

Ni(II) K_d-derivation		Conditions		Conversion (C) / Uncertainty (U)
		Data source[1]	Application	CF
Solution		0.033M $Ca(NO_3)_2$	HDB-6	HDB-6
Substrate		Ca-Montmorillonite	Mudstone	Mudstone
Experimental K_d	(m^3/kg)	5.6		
Conversion Mineralogy (CF-min)				
%clay		100	19	0.19
Conversion Speciation (CF-spec)				
CF-spec/a		1.00	0.31	0.31
CF-spec/b		1.00	0.71	0.71
Conversion Competition (Anionic or Cationic) (CF-cmp)	*irrelevant*			

(1) K_d from Ni(II) sorption edge

Ni(II) K_d-derivation via % clay				CF
CF-1 (%clay)				0.19
Speciation				
CF-2a (% clay)				0.06
CF-2b (% clay)				0.13
				UF
Predicted K_d: CF-1 (% clay)	(m^3/kg)		**1.07**	
UF-starting K_d				1.6
UF-pH conversion				2.5
UF-min				1.4
UF-total				5.5
K_d upper limit	(m^3/kg)		5.93	
K_d lower limit	(m^3/kg)		0.19	
Predicted K_d: CF-2a (% clay)	(m^3/kg)		**0.33**	
UF-starting K_d				1.6
UF-pH conversion				2.5
UF-min				1.4
UF-speciation				1.4
UF-total				7.8
K_d upper limit	(m^3/kg)		2.54	
K_d lower limit	(m^3/kg)		0.042	
Predicted K_d: CF-2b (% clay)	(m^3/kg)		**0.76**	
UF-starting K_d				1.6
UF-pH conversion				2.5
UF-min				1.4
UF-speciation				1.4
UF-total				7.8
K_d upper limit	(m^3/kg)		5.87	
K_d lower limit	(m^3/kg)		0.10	

Speciation Case II

Filename Ni1_equilibrium_case_II_120605_DAT N1_Lauber_2000_sol_spec_pH6.3_120619_DAT
Preequilibration_case_II_120509_DAT

TDB: JAEA,version 100331c1

Ni speciation (Lauber et al., 2000)

System		Application	Data Source
Substrate		Horonobe Mudstone	OPA
Solution		HDB-6	synthetic OPA porewater
pH		8.500	6.30
pCO_2		-	-
Ni(II) dissolved	(mol/kg)	7.34E-11	3.76E-07
Ni^{2+}	(mol/kg)	2.12E-11	2.56E-07
$NiOH^+$	(mol/kg)	7.68E-13	6.56E-11
$Ni(OH)_2$	(mol/kg)	5.42E-13	3.28E-13
$Ni(OH)_3^-$	(mol/kg)	1.65E-15	
$NiHCO_3^+$	(mol/kg)	4.07E-12	2.00E-08
$NiCO_3$	(mol/kg)	2.54E-11	8.84E-10
$NiCl^+$	(mol/kg)	1.67E-12	1.76E-08
NiF^+			2.17E-10
NiS	(mol/kg)	1.95E-11	1.95E-12
$NiHS^+$	(mol/kg)	2.55E-15	3.59E-14
$NiNH_3^{2+}$	(mol/kg)	2.85E-13	
$Ni(NH_3)_2^{2+}$	(mol/kg)	1.18E-15	
$NiSO_4$	(mol/kg)		7.68E-08
$Ni(SO_4)_2^{2-}$	(mol/kg)		4.88E-09
∑ competitive Ni complexes (Ni-cmp) with Ni-CO_3	(mol/kg)	5.09E-11	1.20E-07
Ni tot-(Ni-cmp)	(mol/kg)	2.25E-11	2.56E-07
(Ni tot- (Ni-cmp)) / Ni tot (CF-spec/a)		**3.07E-01**	**6.80E-01**
$NiHCO_3^+$	(mol/kg)	4.07E-12	2.00E-08
$NiCO_3$	(mol/kg)	2.54E-11	8.84E-10
$NiCl^+$	(mol/kg)	1.67E-12	1.76E-08
NiF^+			2.17E-10
NiS	(mol/kg)	1.95E-11	1.95E-12
$NiHS^+$	(mol/kg)	2.55E-15	3.59E-14
$NiNH_3^{2+}$	(mol/kg)	2.85E-13	
$Ni(NH_3)_2^{2+}$	(mol/kg)	1.18E-15	
$NiSO_4$	(mol/kg)		7.68E-08
$Ni(SO_4)_2^{2-}$	(mol/kg)		4.88E-09
∑ competitive Ni complexes (Ni-cmp) without Ni-CO_3	(mol/kg)	2.15E-11	9.95E-08
Ni tot-(Ni-cmp) without Ni-CO_3	(mol/kg)	5.20E-11	2.77E-07
(Ni tot- (Ni-cmp)) / Ni tot (CF-spec/b)		**7.08E-01**	**7.35E-01**

K_d-Derivation Ni(II) on opalinus clay, pH 6.3, Lauber 2000

Overview Pred. K_d [m^3/kg]	Mudstone
	HDB-6
Pred. K_d: CF-1 (% clay)	1.37
Pred. K_d: CF-2a (% clay)	0.62

Ni(II) K_d-derivation		Conditions				Conversion (C) / Uncertainty (U)
		Data source[1]	Additional information[3]		Application[2]	CF
Solution		synthetic porewater	0.1M $NaClO_4$		HDB-6	HDB-6
Substrate		OPA	SWy-1	SWy-1	Mudstone	Mudstone
Experimental K_d	(m^3/kg)	0.3				
Conversion Mineralogy (CF-min)						
%clay		70			19	0.27
Conversion pH (CF-pH)						
pH		6.3	6.3	8.5	8.5	
Kd for 0.1 M NaClO4 (pH conversion)[3]		0.3	0.8	12.6		15.92
Conversion Speciation (CF-spec)						
CF-spec/a		0.68			0.31	0.45
CF-spec/b		0.74			0.71	0.96
Conversion Competition (Anionic or Cationic) (CF-cmp)	*irrelevant*					

(1) K_d value from sorption isotherm
(2) Values of the avarage composition
(3) K_d values from Ni sorption edge in Bradbury and Baeyens (2003)

Ni(II) K_d-derivation via % clay						CF
CF-1 (%clay)						4.32
Speciation						
CF-2a (% clay)						1.95
CF-2b (% clay)						4.16
						UF
Predicted K_d: CF-1 (% clay)	(m^3/kg)				**1.37**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-total						5.5
K_d upper limit	(m^3/kg)				7.58	
K_d lower limit	(m^3/kg)				0.25	
Predicted K_d: CF-2a (% clay)	(m^3/kg)				**0.62**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-speciation						1.4
UF-total						7.8
K_d upper limit	(m^3/kg)				4.78	
K_d lower limit	(m^3/kg)				0.079	
Predicted K_d: CF-2b (% clay)	(m^3/kg)				**1.31**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-speciation						1.4
UF-total						7.8
K_d upper limit	(m^3/kg)				10.20	
K_d lower limit	(m^3/kg)				0.17	

Speciation Case III

Filename Ni1_equilibrium_case_III_120605_DAT Tertre 2005_solution speciation
Preequilibration_case_III_120509_DAT

TDB: JAEA, version 100331c1

Ni(II) speciation (Tertre et al., 2005)

System		Application	Data Source
Substrate		Horonobe mudstone	MX-80
Solution		HDB-6	0.025M $NaClO_4$
pH		8.5	8.50
pCO_2		-	-3.97
Ni(II) dissolved	(mol/kg)	5.09E-11	only hydrolysis relevant
Ni^{2+}	(mol/kg)	1.50E-11	
$NiOH^+$	(mol/kg)	5.41E-13	
$Ni(OH)_2$	(mol/kg)	3.81E-13	
Ni_2OH^{3+}	(mol/kg)		
$Ni(OH)_3^-$	(mol/kg)	1.16E-15	
$NiHCO_3^+$	(mol/kg)	2.92E-12	
$NiCO_3$	(mol/kg)	1.82E-11	
$NiCl^+$	(mol/kg)	1.17E-12	
NiS	(mol/kg)	1.25E-11	
$NiHS^+$	(mol/kg)	1.64E-15	
$NiNH_3^{2+}$	(mol/kg)	1.49E-13	
$Ni(NH_3)_2^{2+}$	(mol/kg)	4.54E-16	
∑ competitive Ni complexes (Ni-cmp) with Ni-CO_3	(mol/kg)	3.497E-11	0.00E+00
Ni tot-(Ni-cmp)	(mol/kg)	1.59E-11	1.70E-06
(Ni tot- (Ni-cmp)) / Ni tot (CF-spec/a)		**3.12E-01**	**1.00E+00**
$NiHCO_3^+$	(mol/kg)	2.92E-12	
$NiCO_3$	(mol/kg)	1.82E-11	
$NiCl^+$	(mol/kg)	1.17E-12	
NiS	(mol/kg)	1.25E-11	
$NiHS^+$	(mol/kg)	1.64E-15	
$NiNH_3^{2+}$	(mol/kg)	1.49E-13	
$Ni(NH_3)_2^{2+}$	(mol/kg)	4.54E-16	
∑ competitive Ni complexes (Ni-cmp) without Ni-CO_3	(mol/kg)	1.39E-11	0.00E+00
Ni tot-(Ni-cmp) without Ni-CO_3	(mol/kg)	3.70E-11	1.70E-06
(Ni tot- (Ni-cmp)) / Ni tot (CF-spec/b)		**7.28E-01**	**1.00E+00**

K_d-Derivation Ni(II) on MX-80, pH 6.75, Tertre et al. (2005)

Overview Pred. K_d [m^3/kg]	Mudstone
	HDB-6
Pred. K_d: CF-1 (% clay)	1.22
Pred. K_d: CF-2a (% clay)	0.38

Ni(II) K_d-derivation		Conditions		Conversion (C) / Uncertainty (U)
		Data source[1]	Application	CF
Solution		0.025M $NaClO_4$	HDB-6	HDB-6
Substrate		MX-80	Mudstone	Mudstone
Experimental K_d	(m^3/kg)	4.500		
Conversion Mineralogy (CF-min)				
%clay		100	27	0.27
Conversion Speciation (CF-spec)				
CF-spec/a		1.00	0.31	0.31
CF-spec/b		1.00	0.73	0.73
Conversion Competition (Anionic or Cationic) (CF-cmp)	*irrelevant*			

(1) K_d values from Ni(II) sorption edge

Ni(II) K_d-derivation via % clay				CF
CF-1 (%clay)				0.27
Speciation				
CF-2a (% clay)				0.08
CF-2b (% clay)				0.20
				UF
Predicted K_d: CF-1 (% clay)	(m^3/kg)		**1.22**	
UF-starting K_d				1.6
UF-pH conversion				2.5
UF-min				1.4
UF-total				5.5
K_d upper limit	(m^3/kg)		6.74	
K_d lower limit	(m^3/kg)		0.22	
Predicted K_d: CF-2a (% clay)	(m^3/kg)		**0.38**	
UF-starting K_d				1.6
UF-pH conversion				2.5
UF-min				1.4
UF-speciation				1.4
UF-total				7.8
K_d upper limit	(m^3/kg)		2.95	
K_d lower limit	(m^3/kg)		0.049	
Predicted K_d: CF-2b (% clay)	(m^3/kg)		**0.88**	
UF-starting K_d				1.6
UF-pH conversion				2.5
UF-min				1.4
UF-speciation				1.4
UF-total				7.8
K_d upper limit	(m^3/kg)		6.87	
K_d lower limit	(m^3/kg)		0.11	

Speciation Case III

Filename Ni1_equilibrium_case_III_120605_DAT B&B 1999_solution speciation
Preequilibration_case_III_120509_DAT

TDB: JAEA,version 100331c1

Ni(II) speciation (Bradbury&Baeyens, 1999)

System		Application	Data Source
Substrate		Horonobe mudstone	Ca-Montmorillonite
Solution		HDB-6	$Ca(NO_3)_2$ solution
Ni(II) added	(mol/kg)	1.30E-07	2.70E-08
pH		8.5	8.5
pCO_2		-	-
Ni(II) dissolved	(mol/kg)	5.09E-11	only hydrolysis considered [1)]
Ni^{2+}	(mol/kg)	1.50E-11	
$NiOH^+$	(mol/kg)	5.41E-13	
$Ni(OH)_2$	(mol/kg)	3.81E-13	
$Ni(OH)_3^-$	(mol/kg)	1.16E-15	
$NiHCO_3^+$	(mol/kg)	2.92E-12	
$NiCO_3$	(mol/kg)	1.82E-11	
$NiCl^+$	(mol/kg)	1.17E-12	
NiS	(mol/kg)	1.25E-11	
$NiHS^+$	(mol/kg)	1.64E-15	
$NiNH_3^{2+}$	(mol/kg)	1.49E-13	
$Ni(NH_3)_2^{2+}$	(mol/kg)	4.54E-16	
∑ competitive Ni complexes (Ni-cmp) with Ni-CO_3	(mol/kg)	3.50E-11	0.00E+00
Ni tot-Ni-cmp)	(mol/kg)	1.59E-11	2.70E-08
(Ni tot- (Ni-cmp)) / Ni tot (CF-spec/a)		**3.12E-01**	**1.00E+00**
$NiHCO_3^+$	(mol/kg)	2.92E-12	
$NiCO_3$	(mol/kg)	1.82E-11	
$NiCl^+$	(mol/kg)	1.17E-12	
NiS	(mol/kg)	1.25E-11	
$NiHS^+$	(mol/kg)	1.64E-15	
$NiNH_3^{2+}$	(mol/kg)	1.49E-13	
$Ni(NH_3)_2^{2+}$	(mol/kg)	4.54E-16	
Ni complexes (Ni-cmp) without Ni-CO_3	(mol/kg)	1.39E-11	0.00E+00
Ni tot-(Ni-cmp) without Ni-CO_3	(mol/kg)	3.70E-11	2.70E-08
(Ni tot- (Ni-cmp)) / Ni tot (CF-spec/b)		**7.276E-01**	**1.000E+00**

[1)] Ni-NO_3 complexes are not relevant (ca. 1E-20 M) according to the JAEA-TDB

K_d-Derivation Ni(II) on Ca-Montmorillonite, pH 6.75, Bradbury&Baeyens (1999)

Overview Pred. K_d [m^3/kg]	Horonobe
	HDB-6
Pred. K_d: CF-1 (% clay)	1.52
Pred. K_d: CF-2a (% clay)	0.47

Ni(II) K_d-derivation		Conditions		Conversion (C) / Uncertainty (U)
		Data source[1]	Application	CF
Solution		0.033M $Ca(NO_3)_2$	HDB-6	HDB-6
Substrate		Ca-Montmorillonite	Mudstone	Mudstone
Experimental K_d	(m^3/kg)	5.6		
Conversion Mineralogy (CF-min)				
%clay		100	27	0.27
Conversion Speciation (CF-spec)				
CF-spec/a		1.00	0.31	0.31
CF-spec/b		1.00	0.73	0.73
Conversion Competition (Anionic or Cationic) (CF-cmp)	*irrelevant*			

(1) K_d values from Ni(II) sorption edge

Ni(II) K_d-derivation via % clay				CF
CF-1 (%clay)				0.27
Speciation				
CF-2a (% clay)				0.08
CF-2b (% clay)				0.20
				UF
Predicted K_d: CF-1 (% clay)	(m^3/kg)		**1.52**	
UF-starting K_d				1.6
UF-pH conversion				2.5
UF-min				1.4
UF-total				5.5
K_d upper limit	(m^3/kg)		8.42	
K_d lower limit	(m^3/kg)		0.27	
Predicted K_d: CF-2a (% clay)	(m^3/kg)		**0.47**	
UF-starting K_d				1.6
UF-pH conversion				2.5
UF-min				1.4
UF-speciation				1.4
UF-total				7.8
K_d upper limit	(m^3/kg)		3.68	
K_d lower limit	(m^3/kg)		0.061	
Predicted K_d: CF-2b (% clay)	(m^3/kg)		**1.10**	
UF-starting K_d				1.6
UF-pH conversion				2.5
UF-min				1.4
UF-speciation				1.4
UF-total				7.8
K_d upper limit	(m^3/kg)		8.58	
K_d lower limit	(m^3/kg)		0.14	

Speciation Case III

Filename Ni1_equilibrium_case_III_120605_DAT N1_Lauber_2000_sol_spec_pH6.3_120619_DAT
Preequilibration_case_III_120509_DAT

TDB: JAEA,version 100331c1

Ni speciation (Lauber et al., 2000)

System		Application	Data Source
Substrate		Horonobe Mudstone	OPA
Solution		HDB-6	synthetic OPA porewater
pH		8.5	6.30
pCO_2		-	-
Ni(II) dissolved	(mol/kg)	5.09E-11	3.76E-07
Ni^{2+}	(mol/kg)	1.50E-11	2.56E-07
$NiOH^+$	(mol/kg)	5.41E-13	6.56E-11
$Ni(OH)_2$	(mol/kg)	3.81E-13	3.28E-13
$Ni(OH)_3^-$	(mol/kg)	1.16E-15	
$NiHCO_3^+$	(mol/kg)	2.92E-12	2.00E-08
$NiCO_3$	(mol/kg)	1.82E-11	8.84E-10
$NiCl^+$	(mol/kg)	1.17E-12	1.76E-08
NiS	(mol/kg)	1.25E-11	1.95E-12
$NiHS^+$	(mol/kg)	1.64E-15	3.59E-14
$NiNH_3^{2+}$	(mol/kg)	1.49E-13	
$Ni(NH_3)_2^{2+}$	(mol/kg)	4.54E-16	
$NiSO_4$	(mol/kg)		7.68E-08
$Ni(SO_4)_2^{2-}$	(mol/kg)		4.88E-09
NiF^+	(mol/kg)		2.17E-10
∑ competitive Ni-complexes (Ni-cmp) with $Ni\text{-}CO_3$	(mol/kg)	3.50E-11	1.20E-07
Ni tot-(Ni-cmp)	(mol/kg)	1.59E-11	2.56E-07
(Ni tot- (Ni-cmp)) / Ni tot (CF-spec/a)		**3.12E-01**	**6.80E-01**
$NiHCO_3^+$	(mol/kg)	2.92E-12	2.00E-08
$NiCO_3$	(mol/kg)	1.82E-11	8.84E-10
$NiCl^+$	(mol/kg)	1.17E-12	1.76E-08
NiS	(mol/kg)	1.25E-11	1.95E-12
$NiHS^+$	(mol/kg)	1.64E-15	3.59E-14
$NiNH_3^{2+}$	(mol/kg)	1.49E-13	
$Ni(NH_3)_2^{2+}$	(mol/kg)	4.54E-16	
$NiSO_4$	(mol/kg)		7.68E-08
$Ni(SO_4)_2^{2-}$	(mol/kg)		4.88E-09
NiF^+	(mol/kg)		2.17E-10
∑ competitive Ni complexes (Ni-cmp) without $Ni\text{-}CO_3$	(mol/kg)	1.39E-11	9.95E-08
Ni tot-(Ni-cmp) without $Ni\text{-}CO_3$	(mol/kg)	3.70E-11	2.77E-07
(Ni tot- (Ni-cmp)) / Ni tot (CF-spec/b)		**7.28E-01**	**7.35E-01**

K_d-Derivation Ni(II) on opalinus clay, pH 6.3, Lauber 2000

Overview Pred. K_d [m^3/kg]	Mudstone
	HDB-6
Pred. K_d: CF-1 (% clay)	1.87
Pred. K_d: CF-2a (% clay)	0.86

Ni(II) K_d-derivation		Conditions				Conversion (C) / Uncertainty (U)
		Data source[1]	Additional information[3]		Application[2]	CF
Solution		synthetic porewater	0.1M $NaClO_4$		HDB-6	HDB-6
Substrate		OPA	SWy-1	SWy-1	Mudstone	Mudstone
Experimental K_d	(m^3/kg)	0.3				
Conversion Mineralogy (CF-min)						
%clay		70			26	0.37
Conversion pH (CF-pH)						
pH		6.3	6.3	8.5	8.5	
Kd for 0.1 M NaClO4 (pH conversion)[3]		0.3	0.8	12.6	0.10	15.92
Conversion Speciation (CF-spec)						
CF-spec/a		0.68			0.31	0.46
CF-spec/b		0.74			0.73	0.99
Conversion Competition (Anionic or Cationic) (CF-cmp)	*irrelevant*					

(1) K_d values from sorption isotherm
(2) Values of the avarage composition
(3) K_d values from Ni sorption edge in Bradbury and Baeyens (2003)

Ni(II) K_d-derivation via % clay						CF
CF-1 (%clay)						5.91
Speciation						
CF-2a (% clay)						2.72
CF-2b (% clay)						5.85
						UF
Predicted K_d: CF-1 (% clay)	(m^3/kg)				**1.87**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-total						5.5
K_d upper limit	(m^3/kg)				10.37	
K_d lower limit	(m^3/kg)				0.34	
Predicted K_d: CF-2a (% clay)	(m^3/kg)				**0.86**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-speciation						1.4
UF-total						7.8
K_d upper limit	(m^3/kg)				6.67	
K_d lower limit	(m^3/kg)				0.111	
Predicted K_d: CF-2b (% clay)	(m^3/kg)				**1.85**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-speciation						1.4
UF-total						7.8
K_d upper limit	(m^3/kg)				14.36	
K_d lower limit	(m^3/kg)				0.24	

Speciation Case I

Filename	EuAm1_equilibrium_case_I_120606_DAT Preequilibration_case_I_120511_DAT	Gorgeon 1994_solution speciation
TDB	JAEA,version 100331c1	

Am(III) speciation (Gorgeon, 1994)

System		Application	Data Source
Substrate		Horonobe mudstone	Illite
Solution		HDB-6	1M $NaClO_4$
pH		6.752	6.75
pCO_2		-	-
Am(III) dissolved	(mol/kg)	6.04E-13	only hydrolysis considered
Am^{3+}	(mol/kg)	4.84E-15	
$AmOH^{2+}$	(mol/kg)	3.71E-16	
$Am(OH)_2^+$	(mol/kg)	1.05E-17	
$Am(OH)_3$	(mol/kg)	3.22E-22	
$AmCO_3^+$	(mol/kg)	2.73E-13	
$Am(CO_3)_2^-$	(mol/kg)	1.41E-13	
$Am(CO_3)_3^{3-}$	(mol/kg)	1.33E-15	
$AmHCO_3^{2+}$	(mol/kg)	3.25E-14	
$AmCl^{2+}$	(mol/kg)	3.10E-16	
$AmCl_2^+$	(mol/kg)	2.20E-18	
$AmSCN^{2+}$	(mol/kg)	2.26E-35	
AmF^{2+}	(mol/kg)	2.03E-17	
AmF_2^+	(mol/kg)	1.56E-20	
$AmSO_4^+$	(mol/kg)	2.78E-22	
$Am(SO_4)_2^-$	(mol/kg)	2.31E-31	
$AmSiO(OH)_3^{2+}$	(mol/kg)	1.50E-13	
$AmH_2PO_4^{2+}$	(mol/kg)	4.63E-19	
∑ competitive Am complexes (Am-cmp) with Am-CO_3	(mol/kg)	5.98E-13	0.00E+00
Am tot-(Am-cmp)	(mol/kg)	5.25E-15	3.30E-10
(Am tot- (Am-cmp)) / Am tot (CF-spec/a)		**8.71E-03**	**1.00E+00**
$AmCO_3^+$	(mol/kg)	2.73E-13	
$Am(CO_3)_2^-$	(mol/kg)	1.41E-13	
$Am(CO_3)_3^{3-}$	(mol/kg)	1.33E-15	
$AmHCO_3^{2+}$	(mol/kg)	3.25E-14	
$AmCl^{2+}$	(mol/kg)	3.10E-16	
$AmCl_2^+$	(mol/kg)	2.20E-18	
$AmSCN^{2+}$	(mol/kg)	2.26E-35	
AmF^{2+}	(mol/kg)	2.03E-17	
AmF_2^+	(mol/kg)	1.56E-20	
$AmSO_4^+$	(mol/kg)	2.78E-22	
$Am(SO_4)_2^-$	(mol/kg)	2.31E-31	
$AmSiO(OH)_3^{2+}$	(mol/kg)	1.50E-13	
$AmH_2PO_4^{2+}$	(mol/kg)	4.63E-19	
Σ competitive Am complexes (Am-cmp) without Am-CO_3	(mol/kg)	1.50E-13	0.00E+00
Am tot-(Am-cmp) without Am-CO_3	(mol/kg)	4.53E-13	3.30E-10
(Am tot- (Am-cmp)) / Am tot (CF-spec/b)		**7.51E-01**	**1.00E+00**

K_d-Derivation Am(III) on Illite, pH 6.78, Gorgeon (1994)

Overview Pred. K_d [m^3/kg]	Mudstone
	HDB-6
Pred. K_d: CF-1 (% clay)	5.51
Pred. K_d: CF-2a (% clay)	0.05
Pred. K_d: CF-2b (% clay)	4.14

Am(III) K_d-derivation		Conditions		Conversion (C) / Uncertainty (U)
		Data source[1]	Application[2]	CF
Solution		1M $NaClO_4$	HDB-6	HDB-6
Substrate		Illite	Mudstone	Mudstone
Experimental K_d	(m^3/kg)	29.0		
Conversion Mineralogy (CF-min)				
%clay		100	19	0.19
Conversion pH (CF-pH)				
pH		6.78	6.752	
pH conversion	*irrelevant*			1.00
Conversion Speciation (CF-spec)				
CF-spec/a		1.00	0.01	0.01
CF-spec/b		1.00	0.75	0.75
Conversion Competition (Anionic or Cationic) (CF-cmp)	*irrelevant*			

(1) K_d values from sorption edge
(2) Values of the avarage composition

Am(III) K_d-derivation via % clay				CF
CF-1 (% clay)				0.19
Speciation				
CF-2a (% clay)				0.002
CF-2b (% clay)				0.14
				UF
Predicted K_d: CF-1 (% clay)	(m^3/kg)		**5.51**	
UF-starting K_d				1.6
UF-min				1.4
UF-total				2.2
K_d upper limit	(m^3/kg)		12.23	
K_d lower limit	(m^3/kg)		2.48	
Predicted K_d: CF-2a (% clay)	(m^3/kg)		**0.05**	
UF-starting K_d				1.6
UF-speciation				1.4
UF-min				1.4
UF-total				3.1
K_d upper limit	(m^3/kg)		0.15	
K_d lower limit	(m^3/kg)		0.02	
Predicted K_d: CF-2b (% clay)	(m^3/kg)		**4.14**	
UF-starting K_d				1.6
UF-speciation				1.4
UF-min				1.4
UF-total				3.1
K_d upper limit	(m^3/kg)		12.86	
K_d lower limit	(m^3/kg)		1.33	

Speciation Case I

Filename Eu2_equilibrium_case_I_120106_DAT Lauber 2000_solution speciation_Eu
Preequilibration_case_I_120511_DAT

TDB: Hummel et al. (2002)

Eu speciation (Lauber et al., 2000)

System		Application	Data Source
Substrate		Horonobe	OPA
Solution		HDB-6	Synthetic OPA porewater
pH		6.752	6.30
pCO2		-	-
Eu dissolved	(mol/kg)	4.49E-18	3.76E-07
Eu^{3+}	(mol/kg)	2.39E-20	1.41E-08
$EuOH^{2+}$	(mol/kg)	1.50E-21	3.14E-10
$Eu(OH)_2^+$	(mol/kg)	9.96E-23	7.12E-12
$Eu(OH)_3$	(mol/kg)	9.23E-25	2.29E-14
$Eu(OH)_4^-$	(mol/kg)	2.48E-30	2.22E-20
$EuCO_3^+$	(mol/kg)	3.23E-18	1.79E-07
$Eu(CO_3)_2^-$	(mol/kg)	2.07E-19	1.11E-09
$EuSO_4^+$	(mol/kg)	1.02E-21	9.10E-08
$Eu(SO_4)_2^-$	(mol/kg)	1.63E-24	2.28E-08
$EuCl^{2+}$	(mol/kg)	2.31E-20	1.18E-08
$EuCl_2^+$	(mol/kg)	3.10E-21	1.32E-09
EuF^{2+}	(mol/kg)	5.15E-22	3.28E-09
EuF_2^+	(mol/kg)	6.15E-25	4.08E-11
$EuSiO(OH)_3^{2+}$	(mol/kg)	9.75E-19	5.16E-08
$Eu(SiO(OH)_3)_2^+$	(mol/kg)	2.77E-20	1.27E-10
∑ competitive Eu complexes (Eu-cmp) with Eu-CO_3	(mol/kg)	4.47E-18	3.62E-07
Eu tot-(Eu-cmp)	(mol/kg)	2.50E-20	1.44E-08
(Eu tot- (Eu-cmp)) / Eu tot (CF-spec/a)		**5.56E-03**	**3.83E-02**
$EuCO_3^+$	(mol/kg)	3.23E-18	1.79E-07
$Eu(CO_3)_2^-$	(mol/kg)	2.07E-19	1.11E-09
$EuSO_4^+$	(mol/kg)	1.98E-21	9.10E-08
$Eu(SO_4)_2^-$	(mol/kg)	6.18E-24	2.28E-08
$EuCl^{2+}$	(mol/kg)	2.32E-20	1.18E-08
$EuCl_2^+$	(mol/kg)	3.11E-21	1.32E-09
EuF^{2+}	(mol/kg)	5.17E-22	3.28E-09
EuF_2^+	(mol/kg)	6.17E-25	4.08E-11
$EuSiO(OH)_3^{2+}$	(mol/kg)	9.78E-19	5.16E-08
$Eu(SiO(OH)_3)_2^+$	(mol/kg)	2.78E-20	1.27E-10
∑ competitive Eu complexes (Eu-cmp) without Eu-CO_3	(mol/kg)	1.03E-18	1.82E-07
Eu tot-(Eu-cmp) without Eu-CO_3	(mol/kg)	3.46E-18	1.94E-07
(Eu tot- (Eu-cmp)) / Eu tot (CF-spec/b)		**7.70E-01**	**5.16E-01**

K_d-Derivation Eu(III) on Opalinus Clay, pH 6.3, Lauber et al. (2000)

Overview Pred. K_d [m^3/kg]	Mudstone
	HDB-6
Pred. K_d: CF-1 (% clay)	5.79
Pred. K_d: CF-2a (% clay)	0.84
Pred. K_d: CF-2b (% clay)	8.63

Eu(III) K_d-derivation		Conditions				Conversion (C) / Uncertainty (U)
		Data source	Additional information[1]		Application[2]	CF
Solution		synthetic porewater			HDB-6	HDB-6
Substrate		OPA	Ca-SWy-1	Ca-SWy-1	Mudstone	Mudstone
Experimental K_d	(m^3/kg)	6.0				
Conversion Mineralogy (CF-min)						
%clay		70			19	0.27
Conversion pH (CF-pH)						
pH		6.3	6.3	6.75	6.752	
Kd (pH conversion)[1]	(m^3/kg)		1.6	5.6		3.55
Conversion Speciation (CF-spec)						
CF-spec/a		0.04			0.01	0.15
CF-spec/b		0.52			0.77	1.49
Conversion Competition (Anionic or Cationic) (CF-cmp)	*irrelevant*					

(1) sorption edge for EU on Ca-SWy-1 (Bradbury and Baeyens, 2002)
(2) Values of the avarage composition

Eu(III) K_d-derivation via % clay						CF
CF-1 (%clay)						0.96
Speciation						
CF-2a (% clay)						0.14
CF-2b (% clay)						1.44
						UF
Predicted K_d: CF-1 (% clay)	(m^3/kg)				**5.79**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-total						5.5
K_d upper limit	(m^3/kg)				32.11	
K_d lower limit	(m^3/kg)				1.04	
Predicted K_d: CF-2a (% clay)	(m^3/kg)				**0.84**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-speciation						1.4
UF-total						7.8
K_d upper limit	(m^3/kg)				6.53	
K_d lower limit	(m^3/kg)				0.11	
Predicted K_d: CF-2b (% clay)	(m^3/kg)				**8.63**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-speciation						1.4
UF-total						7.8
K_d upper limit	(m^3/kg)				67.02	
K_d lower limit	(m^3/kg)				1.11	

Speciation Case II

Filename	EuAm1_equilibrium_case_II_120606_DAT Preequilibration_case_II_120509_DAT	Gorgeon 1994_solution speciation
TDB	JAEA, version 100331c1	

Am(III) speciation (Gorgeon, 1994)

System		Application	Data Source
Substrate		Horonobe mudstone	Illite
Solution		HDB-6	1M $NaClO_4$
pH		8.500	8.50
pCO_2		-	-
Am(III) dissolved	(mol/kg)	1.68E-13	only hydrolysis considered
Am^{3+}	(mol/kg)	2.23E-18	
$AmOH^{2+}$	(mol/kg)	9.60E-18	
$Am(OH)_2^+$	(mol/kg)	1.52E-17	
$Am(OH)_3$	(mol/kg)	2.62E-20	
$AmCO_3^+$	(mol/kg)	5.28E-15	
$Am(CO_3)_2^-$	(mol/kg)	1.14E-13	
$Am(CO_3)_3^{3-}$	(mol/kg)	4.46E-14	
$AmHCO_3^{2+}$	(mol/kg)	1.12E-17	
$AmCl^{2+}$	(mol/kg)	1.38E-19	
$AmCl_2^+$	(mol/kg)	9.44E-22	
$AmSCN^{2+}$	(mol/kg)	1.041E-37	
AmF^{2+}	(mol/kg)		
AmF_2^+	(mol/kg)		
$AmSO_4^+$	(mol/kg)	1.14E-26	
$Am(SO_4)_2^-$	(mol/kg)	8.35E-37	
$AmSiO(OH)_3^{2+}$	(mol/kg)	3.88E-15	
$AmH_2PO_4^{2+}$	(mol/kg)		
∑ competitive Am complexes (Am-cmp) with Am-CO_3	(mol/kg)	1.68E-13	0.00E+00
Am tot-(Am-cmp)	(mol/kg)	1.97E-17	3.30E-10
(Am tot- (Am-cmp)) / Am tot (CF-spec/a)		**1.17E-04**	**1.00E+00**
$AmCO_3^+$	(mol/kg)	5.28E-15	
$Am(CO_3)_2^-$	(mol/kg)	1.14E-13	
$Am(CO_3)_3^{3-}$	(mol/kg)	4.46E-14	
$AmHCO_3^{2+}$	(mol/kg)	1.12E-17	
$AmCl^{2+}$	(mol/kg)	1.38E-19	
$AmCl_2^+$	(mol/kg)	9.44E-22	
$AmSCN^{2+}$	(mol/kg)	1.041E-37	
AmF^{2+}	(mol/kg)		
AmF_2^+	(mol/kg)		
$AmSO_4^+$	(mol/kg)	1.14E-26	
$Am(SO_4)_2^-$	(mol/kg)	8.35E-37	
$AmSiO(OH)_3^{2+}$	(mol/kg)	3.88E-15	
$AmH_2PO_4^{2+}$	(mol/kg)		
∑ competitive Am complexes (Am-cmp) without Am-CO_3	(mol/kg)	3.88E-15	0.00E+00
Am tot-(Am-cmp) without Am-CO_3	(mol/kg)	1.64E-13	3.30E-10
(Am tot- (Am-cmp)) / Am tot (CF-spec/b)		**9.77E-01**	**1.00E+00**

K_d-Derivation Am(III) on Illite, pH 6.78, Gorgeon (1994)

Overview Pred. K_d [m^3/kg]	Mudstone
	HDB-6
Pred. K_d: CF-1 (% clay)	19.95
Pred. K_d: CF-2a (% clay)	0.002
Pred. K_d: CF-2b (% clay)	19.49

Am(III) K_d-derivation		Conditions		Conversion (C) / Uncertainty (U)
		Data source[1]	Application[2]	CF
Solution		1M $NaClO_4$	HDB-6	HDB-6
Substrate		Illite	Mudstone	Mudstone
Experimental K_d	(m^3/kg)	105.0		
Conversion Mineralogy (CF-min)				
%clay		100	19	0.19
Conversion pH (CF-pH)				
pH		6.78	8.5	
pH conversion	*irrelevant*			1.00
Conversion Speciation (CF-spec)				
CF-spec/a		1.00	0.00	0.00
CF-spec/b		1.00	0.98	0.98
Conversion Competition (Anionic or Cationic) (CF-cmp)	*irrelevant*			

(1) K_d values from sorption edge
(2) Values of the avarage composition

Am(III) K_d-derivation via % clay				CF
CF-1 (% clay)				0.19
Speciation				
CF-2a (% clay)				0.000
CF-2b (% clay)				0.19
				UF
Predicted K_d: CF-1 (% clay)	(m^3/kg)		**19.95**	
UF-starting K_d				1.6
UF-min				1.4
UF-total				2.2
K_d upper limit	(m^3/kg)		44.27	
K_d lower limit	(m^3/kg)		8.99	
Predicted K_d: CF-2a (% clay)	(m^3/kg)		**0.00**	
UF-starting K_d				1.6
UF-speciation				1.4
UF-min				1.4
UF-total				3.1
K_d upper limit	(m^3/kg)		0.01	
K_d lower limit	(m^3/kg)		0.00	
Predicted K_d: CF-2b (% clay)	(m^3/kg)		**19.49**	
UF-starting K_d				1.6
UF-speciation				1.4
UF-min				1.4
UF-total				3.1
K_d upper limit	(m^3/kg)		60.54	
K_d lower limit	(m^3/kg)		6.27	

Speciation Case II

Filename Eu2_equilibrium_case_II_120106_DAT Lauber 2000_solution speciation_Eu
Preequilibration_case_II_120509_DAT

TDB: Hummel et al. (2002)

Eu speciation (Lauber et al., 2000)

System		Application	Data Source
Substrate		Horonobe	OPA
Solution		HDB-6	Synthetic OPA porewater
pH		8.500	6.30
pCO2		-	-
Eu dissolved	(mol/kg)	9.87E-16	3.76E-07
Eu^{3+}	(mol/kg)	3.67E-20	1.41E-08
$EuOH^{2+}$	(mol/kg)	1.29E-19	3.14E-10
$Eu(OH)_2^+$	(mol/kg)	4.68E-19	7.12E-12
$Eu(OH)_3$	(mol/kg)	2.40E-19	2.29E-14
$Eu(OH)_4^-$	(mol/kg)	3.65E-23	2.22E-20
$EuCO_3^+$	(mol/kg)	2.07E-16	1.79E-07
$Eu(CO_3)_2^-$	(mol/kg)	5.66E-16	1.11E-09
$EuSO_4^+$	(mol/kg)	1.27E-28	9.10E-08
$Eu(SO_4)_2^-$	(mol/kg)	1.69E-38	2.28E-08
$EuCl^{2+}$	(mol/kg)	3.46E-20	1.18E-08
$EuCl_2^+$	(mol/kg)	4.44E-21	1.32E-09
EuF^{2+}	(mol/kg)		3.28E-09
EuF_2^+	(mol/kg)		4.08E-11
$EuSiO(OH)_3^{2+}$	(mol/kg)	8.36E-17	5.16E-08
$Eu(SiO(OH)_3)_2^+$	(mol/kg)	1.30E-16	1.27E-10
∑ competitive Eu complexes (Eu-cmp) with Eu-CO_3	(mol/kg)	9.86E-16	3.62E-07
Eu tot-(Eu-cmp)	(mol/kg)	9.91E-19	1.44E-08
(Eu tot- (Eu-cmp)) / Eu tot (CF-spec/a)		**1.00E-03**	**3.83E-02**
$EuCO_3^+$	(mol/kg)	2.07E-16	1.79E-07
$Eu(CO_3)_2^-$	(mol/kg)	5.66E-16	1.11E-09
$EuSO_4^+$	(mol/kg)	1.27E-28	9.10E-08
$Eu(SO_4)_2^-$	(mol/kg)	1.69E-38	2.28E-08
$EuCl^{2+}$	(mol/kg)	3.46E-20	1.18E-08
$EuCl_2^+$	(mol/kg)	4.44E-21	1.32E-09
EuF^{2+}	(mol/kg)		3.28E-09
EuF_2^+	(mol/kg)		4.08E-11
$EuSiO(OH)_3^{2+}$	(mol/kg)	8.36E-17	5.16E-08
$Eu(SiO(OH)_3)_2^+$	(mol/kg)	1.30E-16	1.27E-10
∑ competitive Eu complexes (Eu-cmp) without Eu-CO_3	(mol/kg)	2.14E-16	1.82E-07
Eu tot-(Eu-cmp) without Eu-CO_3	(mol/kg)	7.73E-16	1.94E-07
(Eu tot- (Eu-cmp)) / Eu tot (CF-spec/b)		**7.84E-01**	**5.16E-01**

K_d-Derivation Eu(III) on Opalinus Clay, pH 6.3, Lauber et al. (2000)

Overview Pred. K_d [m^3/kg]	Mudstone
	HDB-6
Pred. K_d: CF-1 (% clay)	258.54
Pred. K_d: CF-2a (% clay)	6.78
Pred. K_d: CF-2b (% clay)	392.43

Eu(III) K_d-derivation		Conditions				Conversion (C) / Uncertainty (U)
		Data source[1]	Additional information[1]		Application[2]	CF
Solution		synthetic porewater			HDB-6	HDB-6
Substrate		OPA	Ca-SWy-1	Ca-SWy-1	Mudstone	Mudstone
Experimental K_d	(m^3/kg)	6.0				
Conversion Mineralogy (CF-min)						
%clay		70			19	0.27
Conversion pH (CF-pH)						
pH		6.3	6.3	8.5	8.5	
Kd (pH conversion)[1]	(m^3/kg)		1.6	251.2		158.49
Conversion Speciation (CF-spec)						
CF-spec/a		0.04			1.00E-03	2.62E-02
CF-spec/b		0.52			0.78	1.52
Conversion Competition (Anionic or Cationic) (CF-cmp)	*irrelevant*					

(1) K_d values from sorption edge for Eu on Ca-Swy-1(Bradbury and Baeyens, 2002)
(2) Values of the avarage composition

Eu(III) K_d-derivation via % clay						CF
CF-1 (%clay)						43.02
Speciation						
CF-2a (% clay)						1.13
CF-2b (% clay)						65.30
						UF
Predicted K_d: CF-1 (% clay)	(m^3/kg)				**258.54**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-total						5.5
K_d upper limit	(m^3/kg)				1434.16	
K_d lower limit	(m^3/kg)				46.61	
Predicted K_d: CF-2a (% clay)	(m^3/kg)				**6.78**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-speciation						1.4
UF-total						7.8
K_d upper limit	(m^3/kg)				52.62	
K_d lower limit	(m^3/kg)				0.87	
Predicted K_d: CF-2b (% clay)	(m^3/kg)				**392.43**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-speciation						1.4
UF-total						7.8
K_d upper limit	(m^3/kg)				3047.63	
K_d lower limit	(m^3/kg)				50.53	

Speciation Case III

Filename	EuAm1_equilibrium_situation_case_III_120607_DAT Preequilibration_case_III_120509_DAT	Gorgeon 1994_solution speciation
TDB	JAEA, version 100331c1	

Am(III) speciation (Gorgeon, 1994)

System		Application	Data Source
Substrate		**Horonobe mudstone**	**Illite**
Solution		**HDB-6**	**1M $NaClO_4$**
pH		8.500	8.50
pCO_2		-	-
Am(III) dissolved	(mol/kg)	6.78E-14	only hydrolysis considered
Am^{3+}	(mol/kg)	9.05E-19	
$AmOH^{2+}$	(mol/kg)	3.85E-18	
$Am(OH)_2^+$	(mol/kg)	6.05E-18	
$Am(OH)_3$	(mol/kg)	1.04E-20	
$AmCO_3^+$	(mol/kg)	2.12E-15	
$Am(CO_3)_2^-$	(mol/kg)	4.58E-14	
$Am(CO_3)_3^{3-}$	(mol/kg)	1.83E-14	
$AmHCO_3^{2+}$	(mol/kg)	4.51E-18	
$AmCl^{2+}$	(mol/kg)	5.52E-20	
$AmCl_2^+$	(mol/kg)	3.75E-22	
$AmSCN^{2+}$	(mol/kg)	3.00E-38	
AmF^{2+}	(mol/kg)		
AmF_2^+	(mol/kg)		
$AmSO_4^+$	(mol/kg)	8.20E-27	
$Am(SO_4)_2^-$	(mol/kg)	1.09E-36	
$AmSiO(OH)_3^{2+}$	(mol/kg)	1.55E-15	
$AmH_2PO_4^{2+}$	(mol/kg)		
∑ competitive Am complexes (Am-cmp) with Am-CO_3	(mol/kg)	6.78E-14	0.000E+00
Am tot-(Am-cmp)	(mol/kg)	6.43E-18	3.30E-10
(Am tot- (Am-cmp)) / Am tot (CF-spec/a)		**9.48E-05**	**1.00E+00**
$AmCO_3^+$	(mol/kg)	2.12E-15	
$Am(CO_3)_2^-$	(mol/kg)	4.58E-14	
$Am(CO_3)_3^{3-}$	(mol/kg)	1.83E-14	
$AmHCO_3^{2+}$	(mol/kg)	4.51E-18	
$AmCl^{2+}$		5.52E-20	
$AmCl_2^+$		3.75E-22	
$AmSCN^{2+}$	(mol/kg)	3.00E-38	
AmF^{2+}	(mol/kg)		
AmF_2^+	(mol/kg)		
$AmSO_4^+$	(mol/kg)	8.20E-27	
$Am(SO_4)_2^-$	(mol/kg)	1.09E-36	
$AmSiO(OH)_3^{2+}$	(mol/kg)	1.55E-15	
$AmH_2PO_4^{2+}$	(mol/kg)		
∑ competitive Am complexes (Am-cmp) without Am-CO_3	(mol/kg)	1.55E-15	0.00E+00
Am tot-(Am-cmp) without Am-CO_3	(mol/kg)	6.63E-14	3.30E-10
(Am tot- (Am-cmp)) / Am tot (CF-spec/b)		**9.77E-01**	**1.00E+00**

K_d-Derivation Am(III) on Illite, pH 6.78, Gorgeon (1994)

Overview Pred. K_d [m^3/kg]	Mudstone
	HDB-6
Pred. K_d: CF-1 (% clay)	28.35
Pred. K_d: CF-2a (% clay)	0.003
Pred. K_d: CF-2b (% clay)	27.70

Am(III) K_d-derivation		Conditions		Conversion (C) / Uncertainty (U)
		Data source[1]	Application[2]	CF
Solution		1M $NaClO_4$	HDB-6	HDB-6
Substrate		Illite	Mudstone	Mudstone
Experimental K_d	(m^3/kg)	105.0		
Conversion Mineralogy (CF-min)				
%clay		100	27	0.27
Conversion pH (CF-pH)				
pH		6.78	8.5	
pH conversion	*irrelevant*			1.00
Conversion Speciation (CF-spec)				
CF-spec/a		1.00	0.00	0.00
CF-spec/b		1.00	0.98	0.98
Conversion Competition (Anionic or Cationic) (CF-cmp)	*irrelevant*			

(1) K_d values from data table Gorgeon (1994) given as log Kd
(2) Values of the avarage composition (Table Mineralogy 1)

Am(III) K_d-derivation via % clay				CF
CF-1 (% clay)				0.27
Speciation				
CF-2a (% clay)				0.000
CF-2b (% clay)				0.26
				UF
Predicted K_d: CF-1 (% clay)	(m^3/kg)		**28.35**	
UF-starting K_d				1.6
UF-min				1.4
UF-total				2.2
K_d upper limit	(m^3/kg)		62.90	
K_d lower limit	(m^3/kg)		12.78	
Predicted K_d: CF-2a (% clay)	(m^3/kg)		**0.00**	
UF-starting K_d				1.6
UF-speciation				1.4
UF-min				1.4
UF-total				3.1
K_d upper limit	(m^3/kg)		0.01	
K_d lower limit	(m^3/kg)		0.00	
Predicted K_d: CF-2b (% clay)	(m^3/kg)		**27.70**	
UF-starting K_d				1.6
UF-speciation				1.4
UF-min				1.4
UF-total				3.1
K_d upper limit	(m^3/kg)		86.05	
K_d lower limit	(m^3/kg)		8.92	

Speciation Case III

Filename Eu2_equilibrium_case_III_120109_DAT Lauber 2000_solution speciation_Eu
Preequilibration_case_III_120509_DAT

TDB: Hummel et al. (2002)

Eu speciation (Lauber et al., 2000)

System		Application	Data Source
Substrate		Horonobe	OPA
Solution		HDB-6	Synthetic OPA porewater
pH		8.500	6.30
pCO2		-	-
Eu dissolved	(mol/kg)	8.19E-16	3.76E-07
Eu^{3+}	(mol/kg)	3.12E-20	1.41E-08
$EuOH^{2+}$	(mol/kg)	1.10E-19	3.14E-10
$Eu(OH)_2^+$	(mol/kg)	3.93E-19	7.12E-12
$Eu(OH)_3$	(mol/kg)	2.00E-19	2.29E-14
$Eu(OH)_4^-$	(mol/kg)	3.06E-23	2.22E-20
$EuCO_3^+$	(mol/kg)	1.72E-16	1.79E-07
$Eu(CO_3)_2^-$	(mol/kg)	4.66E-16	1.11E-09
$EuSO_4^+$	(mol/kg)	1.81E-28	9.10E-08
$Eu(SO_4)_2^-$	(mol/kg)	4.12E-38	2.28E-08
$EuCl^{2+}$	(mol/kg)	2.93E-20	1.18E-08
$EuCl_2^+$	(mol/kg)	3.70E-21	1.32E-09
EuF^{2+}	(mol/kg)		3.28E-09
EuF_2^+	(mol/kg)		4.08E-11
$EuSiO(OH)_3^{2+}$	(mol/kg)	7.10E-17	5.16E-08
$Eu(SiO(OH)_3)_2^+$	(mol/kg)	1.09E-16	1.27E-10
∑ competitive Eu complexes (Eu-cmp) with Eu-CO_3	(mol/kg)	8.19E-16	3.62E-07
Eu tot-(Eu-cmp)	(mol/kg)	6.95E-19	1.44E-08
(Eu tot- (Eu-cmp)) / Eu tot (CF-spec/a)		**8.48E-04**	**3.83E-02**
$EuCO_3^+$	(mol/kg)	1.72E-16	1.79E-07
$Eu(CO_3)_2^-$	(mol/kg)	4.66E-16	1.11E-09
$EuSO_4^+$	(mol/kg)	1.81E-28	9.10E-08
$Eu(SO_4)_2^-$	(mol/kg)	4.12E-38	2.28E-08
$EuCl^{2+}$	(mol/kg)	2.93E-20	1.18E-08
$EuCl_2^+$	(mol/kg)	3.70E-21	1.32E-09
EuF^{2+}	(mol/kg)		3.28E-09
EuF_2^+	(mol/kg)		4.08E-11
$EuSiO(OH)_3^{2+}$	(mol/kg)	7.10E-17	5.16E-08
$Eu(SiO(OH)_3)_2^+$	(mol/kg)	1.09E-16	1.27E-10
∑ competitive Eu complexes (Eu-cmp) without Eu-CO_3	(mol/kg)	1.80E-16	1.82E-07
Eu tot-(Eu-cmp) without Eu-CO_3	(mol/kg)	6.39E-16	1.94E-07
(Eu tot- (Eu-cmp)) / Eu tot (CF-spec/b)		**7.80E-01**	**5.16E-01**

K_d-Derivation Eu(III) on Opalinus Clay, pH 6.3, Lauber et al. (2000)

Overview Pred. K_d [m^3/kg]	Mudstone
	HDB-6
Pred. K_d: CF-1 (% clay)	367.40
Pred. K_d: CF-2a (% clay)	8.13
Pred. K_d: CF-2b (% clay)	555.23

Eu(III) K_d-derivation		Conditions				Conversion (C) / Uncertainty (U)
		Data source[1]	Additional information[1]		Application[2]	CF
Solution		synthetic porewater			HDB-6	HDB-6
Substrate		OPA	Ca-SWy-1	Ca-SWy-1	Mudstone	Mudstone
Experimental K_d	(m^3/kg)	6.0				
Conversion Mineralogy (CF-min)						
%clay		70			27	0.39
Conversion pH (CF-pH)						
pH		6.3	6.3	8.5	8.5	
Kd (pH conversion)[1]	(m^3/kg)		1.6	251.2		158.49
Conversion Speciation (CF-spec)						
CF-spec/a		0.04			0.00	0.02
CF-spec/b		0.52			0.78	1.51
Conversion Competition (Anionic or Cationic) (CF-cmp)	*irrelevant*					

(1) K_d values for Eu on Ca-Swy-1(Bradbury and Baeyens, 2002)
(2) Values of the avarage composition

Eu(III) K_d-derivation via % clay						CF
CF-1 (%clay)						61.13
Speciation						
CF-2a (% clay)						1.35
CF-2b (% clay)						92.38
						UF
Predicted K_d: CF-1 (% clay)	(m^3/kg)				**367.40**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-total						5.5
K_d upper limit	(m^3/kg)				2038.02	
K_d lower limit	(m^3/kg)				66.23	
Predicted K_d: CF-2a (% clay)	(m^3/kg)				**8.13**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-speciation						1.4
UF-total						7.8
K_d upper limit	(m^3/kg)				63.17	
K_d lower limit	(m^3/kg)				1.05	
Predicted K_d: CF-2b (% clay)	(m^3/kg)				**555.23**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-speciation						1.4
UF-total						7.8
K_d upper limit	(m^3/kg)				4311.90	
K_d lower limit	(m^3/kg)				71.50	

Speciation Case I

Filename Th1_equilibrium_case_I_120606_DAT Th1_B&B_opalinus_pH7.8_120618_DAT
Preequilibration_case_I_120511_DAT

TDB: JAEA, version 100331c1

Th(IV) speciation (Bradbury&Baeyens, 2003b)

System		Application	Data Source
Substrate		Horonobe Mudstone	Mont Terri Opalinus Clay
Solution		HDB-6	substrate equilibrium soln.
pH		6.752	7.80
pCO_2		-	-
Th(IV) dissolved	(mol/kg)	2.42E-11	1.02E-09
Th^{4+}	(mol/kg)	1.10E-21	2.76E-21
$Th(OH)_4$	(mol/kg)	3.05E-14	1.01E-09
$ThOH^{3+}$	(mol/kg)	2.29E-18	6.09E-17
$Th(OH)_2^{2+}$	(mol/kg)	5.56E-16	1.59E-13
$Th(CO_3)_2(OH)_2^{2-}$	(mol/kg)	2.34E-11	1.38E-11
$Th(CO_3)_5^{6-}$	(mol/kg)	5.77E-14	3.20E-20
$Th(CO_3)_4OH^{5-}$	(mol/kg)	6.85E-13	8.63E-17
$ThCO_3(OH)_4^{2-}$	(mol/kg)	4.46E-17	7.16E-14
ThF_3^{+}	(mol/kg)	2.38E-18	2.11E-15
ThF_4	(mol/kg)	1.03E-18	6.38E-15
ThF_2^{2+}	(mol/kg)	7.07E-18	8.74E-16
ThF^{3+}	(mol/kg)	7.37E-19	1.29E-17
$Th(OH)_3(H_3SiO_4)_3^{2-}$	(mol/kg)	2.48E-19	1.08E-12
$ThCl^{3+}$	(mol/kg)	1.10E-21	3.91E-21
$Th(SO_4)_3^{2-}$	(mol/kg)		1.42E-19
$Th(SO_4)_2$	(mol/kg)		9.43E-19
$ThSO_4^{2+}$	(mol/kg)		3.25E-19
∑ competitive Th complexes (Th-cmp) with Th-CO_3	(mol/kg)	2.42E-11	1.49E-11
Th tot-(Th-cmp)	(mol/kg)	3.73E-14	1.01E-09
(Th tot- (Th-cmp)) / Th tot (CF-spec/a)		**1.54E-03**	**9.85E-01**
$Th(CO_3)_2(OH)_2^{2-}$	(mol/kg)	2.34E-11	1.38E-11
$Th(CO_3)_5^{6-}$	(mol/kg)	5.77E-14	3.20E-20
$Th(CO_3)_4OH^{5-}$	(mol/kg)	6.85E-13	8.63E-17
$ThCO_3(OH)_4^{2-}$	(mol/kg)	4.46E-17	7.16E-14
ThF_3^{+}	(mol/kg)	2.38E-18	2.11E-15
ThF_4	(mol/kg)	1.03E-18	6.38E-15
ThF_2^{2+}	(mol/kg)	7.07E-18	8.74E-16
ThF^{3+}	(mol/kg)	7.37E-19	1.29E-17
$Th(OH)_3(H_3SiO_4)_3^{2-}$	(mol/kg)	2.48E-19	1.08E-12
$ThCl^{3+}$	(mol/kg)	1.10E-21	3.91E-21
$Th(SO_4)_3^{2-}$	(mol/kg)		1.42E-19
$Th(SO_4)_2$	(mol/kg)		9.43E-19
$ThSO_4^{2+}$	(mol/kg)		3.25E-19
∑ competitive Th complexes (Th-cmp) without Th-CO_3	(mol/kg)	1.15E-17	1.09E-12
Th tot-(Th-cmp) without Th-CO_3	(mol/kg)	2.42E-11	1.02E-09
(Th tot- (Th-cmp)) / Th tot (CF-spec/b)		**1.00E+00**	**9.99E-01**

K_d-Derivation Th(IV) on Mont Terri Opalinus Clay, pH 7.8, Bradbury&Baeyens (2003b)

Overview Pred. K_d [m^3/kg]	Mudstone
	HDB-6
Pred. K_d: CF-1 (% clay)	9.55
Pred. K_d: CF-2a (% clay)	0.015
Pred. K_d: CF-2b (% clay)	9.56

Th(IV) K_d-derivation		Conditions				Conversion (C) / Uncertainty (U)
		Data source	Additional information[1]		Application	CF
Solution		Dist. equil. water	0.1 M $NaClO_4$		HDB-6	HDB-6
Substrate		Opalinus Clay	SWy-1	SWy-1	Mudstone	Mudstone
Experimental K_d	(m^3/kg)	55.4				
Conversion Mineralogy (CF-min)						
%clay		63			19	0.30
Conversion pH (CF-pH)						
pH		7.8	7.8	6.75	6.752	
Kd for 0.1 M NaClO4 (pH conversion)[2]		55.4	350	200		0.57
Conversion Speciation (CF-spec)						
CF-spec/a		0.99			1.54E-03	1.56E-03
CF-spec/b		1.00			1.00	1.00
Conversion Competition (Anionic or Cationic) (CF-cmp)	*irrelevant*					

(1) K_d values from Th(IV) sorption edge (Bradbury and Baeyens, 2005)
(2) Values of the avarage composition

Th(IV) K_d-derivation via % clay						CF
CF-1 (%clay)						0.17
Speciation						
CF-2a (% clay)						0.00
CF-2b (% clay)						0.17
						UF
Predicted K_d: CF-1 (% clay)	(m^3/kg)				**9.55**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-total						5.5
K_d upper limit	(m^3/kg)				52.96	
K_d lower limit	(m^3/kg)				1.72	
Predicted K_d: CF-2a (% clay)	(m^3/kg)				**0.01**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-speciation						1.4
UF-total						7.8
K_d upper limit	(m^3/kg)				0.12	
K_d lower limit	(m^3/kg)				0.002	
Predicted K_d: CF-2b (% clay)	(m^3/kg)				**9.56**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-speciation						1.4
UF-total						7.8
K_d upper limit	(m^3/kg)				74.22	
K_d lower limit	(m^3/kg)				1.23	

Speciation Case I

Filename Th1_equilibrium_case_I_120606_DAT Th1_B&B_opalinus_pH6.3_120618_DAT
Preequilibration_case_I_120511_DAT

TDB: JAEA,version 100331c1

Th(IV) speciation (Bradbury&Baeyens, 2003b)

System		Application	Data Source
Substrate		Horonobe Mudstone	Mont Terri Opalinus Clay
Solution		HDB-6	substrate equilibrium soln.
pH		6.75	6.30
pCO_2		-	-
Th(IV) dissolved	(mol/kg)	2.42E-11	1.02E-09
Th^{4+}	(mol/kg)	1.10E-21	3.84E-17
$Th(OH)_4$	(mol/kg)	3.05E-14	1.55E-11
$ThOH^{3+}$	(mol/kg)	2.29E-18	2.76E-14
$Th(OH)_2^{2+}$	(mol/kg)	5.56E-16	2.32E-12
$Th(CO_3)_2(OH)_2^{2-}$	(mol/kg)	2.34E-11	7.69E-10
$Th(CO_3)_5^{6-}$	(mol/kg)	5.77E-14	1.20E-14
$Th(CO_3)_4OH^{5-}$	(mol/kg)	6.85E-13	5.43E-13
$ThCO_3(OH)_4^{2-}$	(mol/kg)	4.46E-17	2.06E-15
ThF_3^{+}	(mol/kg)	2.38E-18	4.61E-11
ThF_4	(mol/kg)	1.03E-18	1.65E-10
ThF_2^{2+}	(mol/kg)	7.07E-18	1.65E-11
ThF^{3+}	(mol/kg)	7.37E-19	2.10E-13
$Th(OH)_3(H_3SiO_4)_3^{2-}$	(mol/kg)	2.48E-19	1.63E-17
$ThCl^{3+}$	(mol/kg)	1.10E-21	3.31E-17
$Th(SO_4)_3^{2-}$	(mol/kg)		5.35E-15
$Th(SO_4)_2$	(mol/kg)		2.71E-14
$ThSO_4^{2+}$	(mol/kg)		6.48E-15
∑ competitive Th complexes (Th-cmp) with Th-CO_3	(mol/kg)	2.42E-11	9.98E-10
Th tot-(Th-cmp)	(mol/kg)	3.73E-14	1.79E-11
(Th tot- (Th-cmp)) / Th tot (CF-spec/a)		**1.54E-03**	**1.76E-02**
$Th(CO_3)_2(OH)_2^{2-}$	(mol/kg)	2.34E-11	7.69E-10
$Th(CO_3)_5^{6-}$	(mol/kg)	5.77E-14	1.20E-14
$Th(CO_3)_4OH^{5-}$	(mol/kg)	6.85E-13	5.43E-13
$ThCO_3(OH)_4^{2-}$	(mol/kg)	4.46E-17	2.06E-15
ThF_3^{+}	(mol/kg)	2.38E-18	4.61E-11
ThF_4	(mol/kg)	1.03E-18	1.65E-10
ThF_2^{2+}	(mol/kg)	7.07E-18	1.65E-11
ThF^{3+}	(mol/kg)	7.37E-19	2.10E-13
$Th(OH)_3(H_3SiO_4)_3^{2-}$	(mol/kg)	2.48E-19	1.63E-17
$ThCl^{3+}$	(mol/kg)	1.10E-21	3.31E-17
$Th(SO_4)_3^{2-}$	(mol/kg)		5.35E-15
$Th(SO_4)_2$	(mol/kg)		2.71E-14
$ThSO_4^{2+}$	(mol/kg)		6.48E-15
∑ competitive Th complexes (Th-cmp) without Th-CO_3	(mol/kg)	1.15E-17	2.28E-10
Th tot-(Th-cmp) without Th-CO_3	(mol/kg)	2.42E-11	7.88E-10
(Th tot- (Th-cmp)) / Th tot (CF-spec/b)		**1.00E+00**	**7.75E-01**

K_d-Derivation Th(IV) on Mont Terri Opalinus Clay, pH 6.3, Bradbury&Baeyens (2003b)

Overview Pred. K_d [m^3/kg]	Mudstone
	HDB-6
Pred. K_d: CF-1 (% clay)	7.37
Pred. K_d: CF-2a (% clay)	0.64
Pred. K_d: CF-2b (% clay)	9.51

Th(IV) K_d-derivation		Conditions				Conversion (C) / Uncertainty (U)
		Data source	Additional information[1]		Application	CF
Solution		Dist. equil. water	0.1 M $NaClO_4$		HDB-6	HDB-6
Substrate		Opalinus Clay	SWy-1	SWy-1	Mudstone	Mudstone
Experimental K_d	(m^3/kg)	22.0				
Conversion Mineralogy (CF-min)						
%clay		63			19	0.30
Conversion pH (CF-pH)						
pH		6.3	6.3	6.75	6.75	
Kd for 0.1 M NaClO4 (pH conversion)[2]		22.0	180	200		1.11
Conversion Speciation (CF-spec)						
CF-spec/a		1.76E-02			0.00	0.09
CF-spec/b		0.78			1.00	1.29
Conversion Competition (Anionic or Cationic) (CF-cmp)	*irrelevant*					

(1) Kd values from Th(IV) sorption edge (Bradbury and Baeyens, 2005)
(2) Values of the avarage composition

Th(IV) K_d-derivation via % clay						CF
CF-1 (%clay)						0.34
Speciation						
CF-2a (% clay)						0.03
CF-2b (% clay)						0.43
						UF
Predicted K_d: CF-1 (% clay)	(m^3/kg)				**7.37**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-total						5.5
K_d upper limit	(m^3/kg)				40.89	
K_d lower limit	(m^3/kg)				1.33	
Predicted K_d: CF-2a (% clay)	(m^3/kg)				**0.64**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-speciation						1.4
UF-total						7.8
K_d upper limit	(m^3/kg)				5.01	
K_d lower limit	(m^3/kg)				0.08	
Predicted K_d: CF-2b (% clay)	(m^3/kg)				**9.51**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-speciation						1.4
UF-total						7.8
K_d upper limit	(m^3/kg)				73.84	
K_d lower limit	(m^3/kg)				1.22	

Speciation Case I

Filename Th1_equilibrium_case_I_120606_DAT Th1_B&B_MX-80_pH7.25_120618_DAT
Preequilibration_case_I_120511_DAT

TDB: JAEA,version 100331c1

Th(IV) speciation (Bradbury&Baeyens, 2003a)

System		Application	Data Source
Substrate		Horonobe Mudstone	MX-80
Solution		HDB-6	substrate equilibrium soln.
pH		6.752	7.25
pCO_2		-	-
Th(IV) dissolved	(mol/kg)	2.42E-11	1.02E-09
Th^{4+}	(mol/kg)	1.10E-21	9.12E-20
$Th(OH)_4$	(mol/kg)	3.05E-14	2.17E-10
$ThOH^{3+}$	(mol/kg)	2.29E-18	5.69E-16
$Th(OH)_2^{2+}$	(mol/kg)	5.56E-16	4.19E-13
$Th(CO_3)_2(OH)_2^{2-}$	(mol/kg)	2.34E-11	7.99E-10
$Th(CO_3)_5^{6-}$	(mol/kg)	5.77E-14	2.40E-15
$Th(CO_3)_4OH^{5-}$	(mol/kg)	6.85E-13	3.90E-13
$ThCO_3(OH)_4^{2-}$	(mol/kg)	4.46E-17	7.08E-14
ThF_3^{+}	(mol/kg)	2.38E-18	5.43E-13
ThF_4	(mol/kg)	1.03E-18	3.32E-12
ThF_2^{2+}	(mol/kg)	7.07E-18	1.14E-13
ThF^{3+}	(mol/kg)	7.37E-19	8.45E-16
$Th(OH)_3(H_3SiO_4)_3^{2-}$	(mol/kg)	2.48E-19	1.84E-14
$ThCl^{3+}$	(mol/kg)	1.10E-21	7.37E-20
$Th(SO_4)_3^{2-}$	(mol/kg)		1.75E-16
$Th(SO_4)_2$	(mol/kg)		3.56E-16
$ThSO_4^{2+}$	(mol/kg)		3.59E-17
∑ competitive Th complexes (Th-cmp) with Th-CO_3	(mol/kg)	2.42E-11	8.03E-10
Th tot-(Th-cmp)	(mol/kg)	3.73E-14	2.17E-10
(Th tot- (Th-cmp)) / Th tot (CF-spec/a)		**1.54E-03**	**2.13E-01**
$Th(CO_3)_2(OH)_2^{2-}$	(mol/kg)	2.34E-11	7.99E-10
$Th(CO_3)_5^{6-}$	(mol/kg)	5.77E-14	2.40E-15
$Th(CO_3)_4OH^{5-}$	(mol/kg)	6.85E-13	3.90E-13
$ThCO_3(OH)_4^{2-}$	(mol/kg)	4.46E-17	7.08E-14
ThF_3^{+}	(mol/kg)	2.38E-18	5.43E-13
ThF_4	(mol/kg)	1.03E-18	3.32E-12
ThF_2^{2+}	(mol/kg)	7.07E-18	1.14E-13
ThF^{3+}	(mol/kg)	7.37E-19	8.45E-16
$Th(OH)_3(H_3SiO_4)_3^{2-}$	(mol/kg)	2.48E-19	1.84E-14
$ThCl^{3+}$	(mol/kg)	1.10E-21	7.37E-20
$Th(SO_4)_3^{2-}$	(mol/kg)		1.75E-16
$Th(SO_4)_2$	(mol/kg)		3.56E-16
$ThSO_4^{2+}$	(mol/kg)		3.59E-17
∑ competitive Th complexes (Th-cmp) without Th-CO_3	(mol/kg)	1.15E-17	4.00E-12
Th tot-(Th-cmp) without Th-CO_3	(mol/kg)	2.42E-11	1.02E-09
(Th tot- (Th-cmp)) / Th tot (CF-spec/b)		**1.00E+00**	**9.96E-01**

K_d-Derivation Th(IV) on MX-80, pH 7.25, Bradbury&Baeyens (2003a)

Overview Pred. K_d [m^3/kg]	Mudstone
	HDB-6
Pred. K_d: CF-1 (% clay)	14.51
Pred. K_d: CF-2a (% clay)	0.11
Pred. K_d: CF-2b (% clay)	14.57

Th(IV) K_d-derivation		Conditions				Conversion (C) / Uncertainty (U)
		Data source	Additional information[1]		Application[2]	CF
Solution		Dist. equil. water	0.1 M $NaClO_4$		HDB-6	HDB-6
Substrate		MX-80	SWy-1	SWy-1	Mudstone	Mudstone
Experimental K_d	(m^3/kg)	63.0				
Conversion Mineralogy (CF-min)						
%clay		75			19	0.25
Conversion pH (CF-pH)						
pH		7.25	7.25	6.75	6.752	
Kd for 0.1 M NaClO4 (pH conversion)		63.0	220	200		0.91
Conversion Speciation (CF-spec)						
CF-spec/a		0.21			0.00	0.01
CF-spec/b		1.00			1.00	1.00
Conversion Competition (Anionic or Cationic) (CF-cmp)	*irrelevant*					

(1) Kd values from Th(IV) sorption edge (Bradbury and Baeyens, 2005)
(2) Values of the avarage composition

Th(IV) K_d-derivation via % clay						CF
CF-1 (%clay)						0.23
Speciation						
CF-2a (% clay)						0.00
CF-2b (% clay)						0.23
						UF
Predicted K_d: CF-1 (% clay)	(m^3/kg)				**14.51**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-total						5.5
K_d upper limit	(m^3/kg)				80.48	
K_d lower limit	(m^3/kg)				2.62	
Predicted K_d: CF-2a (% clay)	(m^3/kg)				**0.11**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-speciation						1.4
UF-total						7.8
K_d upper limit	(m^3/kg)				0.82	
K_d lower limit	(m^3/kg)				0.01	
Predicted K_d: CF-2b (% clay)	(m^3/kg)				**14.57**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-speciation						1.4
UF-total						7.8
K_d upper limit	(m^3/kg)				113.12	
K_d lower limit	(m^3/kg)				1.88	

Speciation Case II

Filename Th1_equilibrium_case_II_120606_DAT Th1_B&B_opalinus_pH7.8_120618_DAT
Preequilibration_case_II_120509_DAT

TDB: JAEA,version 100331c1

Th(IV) speciation (Bradbury&Baeyens, 2003b)

System		Application	Data Source
Substrate		Horonobe Mudstone	Mont Terri Opalinus Clay
Solution		HDB-6	substrate equilibrium soln.
pH		8.500	7.80
pCO_2		-	-
Th(IV) dissolved	(mol/kg)	2.00E-11	1.02E-09
Th^{4+}	(mol/kg)	8.71E-29	2.76E-21
$Th(OH)_4$	(mol/kg)	2.40E-14	1.01E-09
$ThOH^{3+}$	(mol/kg)	1.02E-23	6.09E-17
$Th(OH)_2^{2+}$	(mol/kg)	1.39E-19	1.59E-13
$Th(CO_3)_5^{6-}$	(mol/kg)	5.71E-13	3.20E-20
$Th(CO_3)_2(OH)_2^{2-}$	(mol/kg)	1.02E-11	1.38E-11
$Th(CO_3)_4OH^{5-}$	(mol/kg)	9.15E-12	8.63E-17
$ThCO_3(OH)_4^{2-}$	(mol/kg)	1.46E-15	7.16E-14
$Th(OH)_3(H_3SiO_4)_3^{2-}$	(mol/kg)	6.11E-16	1.08E-12
ThF_3^{+}	(mol/kg)		2.11E-15
ThF_4	(mol/kg)		6.38E-15
ThF_2^{2+}	(mol/kg)		8.74E-16
ThF^{3+}	(mol/kg)		1.29E-17
$Th(SO_4)_3^{2-}$	(mol/kg)		1.42E-19
$Th(SO_4)_2$	(mol/kg)		9.43E-19
$ThSO_4^{2+}$	(mol/kg)		3.25E-19
∑ competitive Th complexes (Th-cmp) with Th-CO_3	(mol/kg)	1.99E-11	1.49E-11
Th tot-(Th-cmp)	(mol/kg)	1.51E-14	1.01E-09
(Th tot- (Th-cmp)) / Th tot (CF-spec/a)		**7.58E-04**	**9.85E-01**
$Th(CO_3)_5^{6-}$	(mol/kg)	5.71E-13	3.20E-20
$Th(CO_3)_2(OH)_2^{2-}$	(mol/kg)	1.02E-11	1.38E-11
$Th(CO_3)_4OH^{5-}$	(mol/kg)	9.15E-12	8.63E-17
$ThCO_3(OH)_4^{2-}$	(mol/kg)	1.46E-15	7.16E-14
$Th(OH)_3(H_3SiO_4)_3^{2-}$	(mol/kg)	6.11E-16	1.08E-12
ThF_3^{+}	(mol/kg)		2.11E-15
ThF_4	(mol/kg)		6.38E-15
ThF_2^{2+}	(mol/kg)		8.74E-16
ThF^{3+}	(mol/kg)		1.29E-17
$Th(SO_4)_3^{2-}$	(mol/kg)		1.42E-19
$Th(SO_4)_2$	(mol/kg)		9.43E-19
$ThSO_4^{2+}$	(mol/kg)		3.25E-19
∑ competitive Th complexes (Th-cmp) without Th-CO_3	(mol/kg)	6.11E-16	1.09E-12
Th tot-(Th-cmp) without Th-CO_3	(mol/kg)	1.99E-11	1.02E-09
(Th tot- (Th-cmp)) / Th tot (CF-spec/b)		**1.00E+00**	**9.99E-01**

K_d-Derivation Th(IV) on Mont Terri Opalinus Clay, pH 7.8, Bradbury&Baeyens (2003b)

Overview Pred. K_d [m^3/kg]	Mudstone
	HDB-6
Pred. K_d: CF-1 (% clay)	19.09
Pred. K_d: CF-2a (% clay)	0.01
Pred. K_d: CF-2b (% clay)	19.11

Th(IV) K_d-derivation		Conditions				Conversion (C) / Uncertainty (U)
		Data source	Additional information[1]		Application	CF
Solution		Dist. equil. water	0.1 M $NaClO_4$		HDB-6	HDB-6
Substrate		Opalinus Clay	SWy-1	SWy-1	Mudstone	Mudstone
Experimental K_d	(m^3/kg)	55.4				
Conversion Mineralogy (CF-min)						
%clay		63			19	0.30
Conversion pH (CF-pH)						
pH		7.8	7.8	8.5	8.5	
Kd for 0.1 M NaClO4 (pH conversion)[2]		55.4	350	400		1.14
Conversion Speciation (CF-spec)						
CF-spec/a		0.99			7.58E-04	7.70E-04
CF-spec/b		1.00			1.00	1.00
Conversion Competition (Anionic or Cationic) (CF-cmp)	*irrelevant*					

(1) K_d values from Th(IV) sorption edge (Bradbury and Baeyens, 2005)
(2) Values of the avarage composition

Th(IV) K_d-derivation via % clay						CF
CF-1 (%clay)						0.34
Speciation						
CF-2a (% clay)						0.00
CF-2b (% clay)						0.35
						UF
Predicted K_d: CF-1 (% clay)	(m^3/kg)				**19.09**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-total						5.5
K_d upper limit	(m^3/kg)				105.92	
K_d lower limit	(m^3/kg)				3.44	
Predicted K_d: CF-2a (% clay)	(m^3/kg)				**0.01**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-speciation						1.4
UF-total						7.8
K_d upper limit	(m^3/kg)				0.11	
K_d lower limit	(m^3/kg)				0.002	
Predicted K_d: CF-2b (% clay)	(m^3/kg)				**19.11**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-speciation						1.4
UF-total						7.8
K_d upper limit	(m^3/kg)				148.44	
K_d lower limit	(m^3/kg)				2.46	

Speciation Case II

Filename Th1_equilibrium_case_II_120606_DAT Th1_B&B_opalinus_pH6.3_120618_DAT
Preequilibration_case_II_120509_DAT

TDB: JAEA,version 100331c1

Th(IV) speciation (Bradbury&Baeyens, 2003b)

System		Application	Data Source
Substrate		Horonobe Mudstone	Mont Terri Opalinus Clay
Solution		HDB-6	substrate equilibrium soln.
pH		8.50	6.30
pCO_2		-	-
Th(IV) dissolved	(mol/kg)	2.00E-11	1.02E-09
Th^{4+}	(mol/kg)	8.71E-29	3.84E-17
$Th(OH)_4$	(mol/kg)	2.40E-14	1.55E-11
$ThOH^{3+}$	(mol/kg)	1.02E-23	2.76E-14
$Th(OH)_2^{2+}$	(mol/kg)	1.39E-19	2.32E-12
$Th(CO_3)_5^{6-}$	(mol/kg)	5.71E-13	1.20E-14
$Th(CO_3)_2(OH)_2^{2-}$	(mol/kg)	1.02E-11	7.69E-10
$Th(CO_3)_4OH^{5-}$	(mol/kg)	9.15E-12	5.43E-13
$ThCO_3(OH)_4^{2-}$	(mol/kg)	1.46E-15	2.06E-15
$Th(OH)_3(H_3SiO_4)_3^{2-}$	(mol/kg)	6.11E-16	1.63E-17
ThF_3^{+}	(mol/kg)		4.61E-11
ThF_4	(mol/kg)		1.65E-10
ThF_2^{2+}	(mol/kg)		1.65E-11
ThF^{3+}	(mol/kg)		2.10E-13
$Th(SO_4)_3^{2-}$	(mol/kg)		5.35E-15
$Th(SO_4)_2$	(mol/kg)		2.71E-14
$ThSO_4^{2+}$	(mol/kg)		6.48E-15
∑ competitive Th complexes (Th-cmp) with Th-CO_3	(mol/kg)	1.99E-11	9.98E-10
Th tot-(Th-cmp)	(mol/kg)	1.51E-14	1.79E-11
(Th tot- (Th-cmp)) / Th tot (CF-spec/a)		**7.58E-04**	**1.76E-02**
$Th(CO_3)_5^{6-}$	(mol/kg)	5.71E-13	1.20E-14
$Th(CO_3)_2(OH)_2^{2-}$	(mol/kg)	1.02E-11	7.69E-10
$Th(CO_3)_4OH^{5-}$	(mol/kg)	9.15E-12	5.43E-13
$ThCO_3(OH)_4^{2-}$	(mol/kg)	1.46E-15	2.06E-15
$Th(OH)_3(H_3SiO_4)_3^{2-}$	(mol/kg)	6.11E-16	1.63E-17
ThF_3^{+}	(mol/kg)		4.61E-11
ThF_4	(mol/kg)		1.65E-10
ThF_2^{2+}	(mol/kg)		1.65E-11
ThF^{3+}	(mol/kg)		2.10E-13
$Th(SO_4)_3^{2-}$	(mol/kg)		5.35E-15
$Th(SO_4)_2$	(mol/kg)		2.71E-14
$ThSO_4^{2+}$	(mol/kg)		6.48E-15
∑ competitive Th complexes (Th-cmp) without Th-CO_3	(mol/kg)	6.11E-16	2.28E-10
Th tot-(Th-cmp) without Th-CO_3	(mol/kg)	1.99E-11	7.88E-10
(Th tot- (Th-cmp)) / Th tot (CF-spec/b)		**1.00E+00**	**7.75E-01**

K_d-Derivation Th(IV) on Mont Terri Opalinus Clay, pH 6.3, Bradbury&Baeyens (2003b)

Overview Pred. K_d [m^3/kg]	Mudstone
	HDB-6
Pred. K_d: CF-1 (% clay)	14.74
Pred. K_d: CF-2a (% clay)	0.63
Pred. K_d: CF-2b (% clay)	19.02

Th(IV) K_d-derivation		Conditions				Conversion (C) / Uncertainty (U)
		Data source	Additional information[1]		Application	CF
Solution		Dist. equil. water	0.1 M $NaClO_4$		HDB-6	HDB-6
Substrate		Opalinus Clay	SWy-1	SWy-1	Mudstone	Mudstone
Experimental K_d	(m^3/kg)	22.0				
Conversion Mineralogy (CF-min)						
%clay		63			19	0.30
Conversion pH (CF-pH)						
pH		6.3	6.3	8.5	8.50	
Kd for 0.1 M NaClO4 (pH conversion)[2]		22.0	180	400		2.22
Conversion Speciation (CF-spec)						
CF-spec/a		1.76E-02			7.58E-04	0.04
CF-spec/b		0.78			1.00	1.29
Conversion Competition (Anionic or Cationic) (CF-cmp)	*irrelevant*					

(1) Kd values from Th(IV) sorption edge (Bradbury and Baeyens, 2005)
(2) Values of the avarage composition

Th(IV) K_d-derivation via % clay						CF
CF-1 (%clay)						0.67
Speciation						
CF-2a (% clay)						0.03
CF-2b (% clay)						0.86
						UF
Predicted K_d: CF-1 (% clay)	(m^3/kg)				**14.74**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-total						5.5
K_d upper limit	(m^3/kg)				81.79	
K_d lower limit	(m^3/kg)				2.66	
Predicted K_d: CF-2a (% clay)	(m^3/kg)				**0.63**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-speciation						1.4
UF-total						7.8
K_d upper limit	(m^3/kg)				4.92	
K_d lower limit	(m^3/kg)				0.08	
Predicted K_d: CF-2b (% clay)	(m^3/kg)				**19.02**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-speciation						1.4
UF-total						7.8
K_d upper limit	(m^3/kg)				147.67	
K_d lower limit	(m^3/kg)				2.45	

Speciation Case II

Filename Th1_equilibrium_case_II_120606_DAT Th1_B&B_MX-80_pH7.25_120618_DAT
Preequilibration_case_II_120509_DAT

TDB: JAEA,version 100331c1

Th(IV) speciation (Bradbury&Baeyens, 2003a)

System		Application	Data Source
Substrate		Horonobe Mudstone	MX-80
Solution		HDB-6	substrate equilibrium soln.
pH		8.50	7.25
pCO_2		-	-
Th(IV) dissolved	(mol/kg)	2.00E-11	1.02E-09
Th^{4+}	(mol/kg)	8.71E-29	9.12E-20
$Th(OH)_4$	(mol/kg)	2.40E-14	2.17E-10
$ThOH^{3+}$	(mol/kg)	1.02E-23	5.69E-16
$Th(OH)_2^{2+}$	(mol/kg)	1.39E-19	4.19E-13
$Th(CO_3)_5^{6-}$	(mol/kg)	5.71E-13	2.40E-15
$Th(CO_3)_2(OH)_2^{2-}$	(mol/kg)	1.02E-11	7.99E-10
$Th(CO_3)_4OH^{5-}$	(mol/kg)	9.15E-12	3.90E-13
$ThCO_3(OH)_4^{2-}$	(mol/kg)	1.46E-15	7.08E-14
$Th(OH)_3(H_3SiO_4)_3^{2-}$	(mol/kg)	6.11E-16	1.84E-14
ThF_3^{+}	(mol/kg)		5.43E-13
ThF_4	(mol/kg)		3.32E-12
ThF_2^{2+}	(mol/kg)		1.14E-13
ThF^{3+}	(mol/kg)		8.45E-16
$Th(SO_4)_3^{2-}$	(mol/kg)		1.75E-16
$Th(SO_4)_2$	(mol/kg)		3.56E-16
$ThSO_4^{2+}$	(mol/kg)		3.59E-17
∑ competitive Th complexes (Th-cmp) with Th-CO_3	(mol/kg)	1.99E-11	8.03E-10
Th tot-(Th-cmp)	(mol/kg)	1.51E-14	2.17E-10
(Th tot- (Th-cmp)) / Th tot (CF-spec/a)		**7.58E-04**	**2.13E-01**
$Th(CO_3)_5^{6-}$	(mol/kg)	5.71E-13	2.40E-15
$Th(CO_3)_2(OH)_2^{2-}$	(mol/kg)	1.02E-11	7.99E-10
$Th(CO_3)_4OH^{5-}$	(mol/kg)	9.15E-12	3.90E-13
$ThCO_3(OH)_4^{2-}$	(mol/kg)	1.46E-15	7.08E-14
$Th(OH)_3(H_3SiO_4)_3^{2-}$	(mol/kg)	6.11E-16	1.84E-14
ThF_3^{+}	(mol/kg)		5.43E-13
ThF_4	(mol/kg)		3.32E-12
ThF_2^{2+}	(mol/kg)		1.14E-13
ThF^{3+}	(mol/kg)		8.45E-16
$Th(SO_4)_3^{2-}$	(mol/kg)		1.75E-16
$Th(SO_4)_2$	(mol/kg)		3.56E-16
$ThSO_4^{2+}$	(mol/kg)		3.59E-17
∑ competitive Th complexes (Th-cmp) without Th-CO_3	(mol/kg)	6.11E-16	4.00E-12
Th tot-(Th-cmp) without Th-CO_3	(mol/kg)	1.99E-11	1.02E-09
(Th tot- (Th-cmp)) / Th tot (CF-spec/b)		**1.00E+00**	**9.96E-01**

K_d-Derivation Th(IV) on MX-80, pH 7.25, Bradbury&Baeyens (2003a)

Overview Pred. K_d [m^3/kg]	Mudstone
	HDB-6
Pred. K_d: CF-1 (% clay)	29.02
Pred. K_d: CF-2a (% clay)	0.10
Pred. K_d: CF-2b (% clay)	29.13

Th(IV) K_d-derivation		Conditions				Conversion (C) / Uncertainty (U)
		Data source	Additional information[1]		Application[2]	CF
Solution		Dist. equil. water	0.1 M $NaClO_4$		HDB-6	HDB-6
Substrate		MX-80	SWy-1	SWy-1	Mudstone	Mudstone
Experimental K_d	(m^3/kg)	63.0				
Conversion Mineralogy (CF-min)						
%clay		75			19	0.25
Conversion pH (CF-pH)						
pH		7.25	7.25	8.5	8.5	
Kd for 0.1 M NaClO4 (pH conversion)		63.0	220	400		1.82
Conversion Speciation (CF-spec)						
CF-spec/a		2.13E-01			7.58E-04	0.00
CF-spec/b		1.00			1.00	1.00
Conversion Competition (Anionic or Cationic) (CF-cmp)	*irrelevant*					

(1) Kd values from Th(IV) sorption edge (Bradbury and Baeyens, 2005)
(2) Values of the avarage composition

Th(IV) K_d-derivation via % clay						CF
CF-1 (%clay)						0.46
Speciation						
CF-2a (% clay)						0.00
CF-2b (% clay)						0.46
						UF
Predicted K_d: CF-1 (% clay)	(m^3/kg)				**29.02**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-total						5.5
K_d upper limit	(m^3/kg)				160.97	
K_d lower limit	(m^3/kg)				5.23	
Predicted K_d: CF-2a (% clay)	(m^3/kg)				**0.10**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-speciation						1.4
UF-total						7.8
K_d upper limit	(m^3/kg)				0.80	
K_d lower limit	(m^3/kg)				0.01	
Predicted K_d: CF-2b (% clay)	(m^3/kg)				**29.13**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-speciation						1.4
UF-total						7.8
K_d upper limit	(m^3/kg)				226.23	
K_d lower limit	(m^3/kg)				3.75	

Speciation Case III

Filename Th1_equilibrium_case_III_120609_DAT Th1_B&B_opalinus_pH7.8_120618_DAT
Preequilibration_case_III_120509_DAT

TDB: JAEA,version 100331c1

Th(IV) speciation (Bradbury&Baeyens, 2003b)

System		Application	Data Source
Substrate		Horonobe Mudstone	Mont Terri Opalinus Clay
Solution		HDB-6	substrate equilibrium soln.
pH		8.500	7.80
pCO_2		-	-
Th(IV) dissolved	(mol/kg)	1.51E-11	1.02E-09
Th^{4+}	(mol/kg)	6.20E-29	2.76E-21
$Th(OH)_4$	(mol/kg)	1.69E-14	1.01E-09
$ThOH^{3+}$	(mol/kg)	7.24E-24	6.09E-17
$Th(OH)_2^{2+}$	(mol/kg)	9.83E-20	1.59E-13
$Th(CO_3)_5^{6-}$	(mol/kg)	4.54E-13	3.20E-20
$Th(CO_3)_2(OH)_2^{2-}$	(mol/kg)	7.49E-12	1.38E-11
$Th(CO_3)_4OH^{5-}$	(mol/kg)	7.09E-12	8.63E-17
$ThCO_3(OH)_4^{2-}$	(mol/kg)	1.05E-15	7.16E-14
ThF_3^{+}	(mol/kg)		2.11E-15
ThF_4	(mol/kg)		6.38E-15
ThF_2^{2+}	(mol/kg)		8.74E-16
ThF^{3+}	(mol/kg)		1.29E-17
$Th(SO_4)_3^{2-}$	(mol/kg)		1.42E-19
$Th(SO_4)_2$	(mol/kg)		9.43E-19
$ThSO_4^{2+}$	(mol/kg)		3.25E-19
$Th(OH)_3(H_3SiO_4)_3^{2-}$	(mol/kg)	4.31E-16	1.08E-12
∑ competitive Th complexes (Th-cmp) with Th-CO_3	(mol/kg)	1.50E-11	1.491E-11
Th tot-(Th-cmp)	(mol/kg)	1.64E-14	1.01E-09
(Th tot- (Th-cmp)) / Th tot (CF-spec/a)		**1.09E-03**	**9.85E-01**
$Th(CO_3)_5^{6-}$	(mol/kg)	4.54E-13	3.20E-20
$Th(CO_3)_2(OH)_2^{2-}$	(mol/kg)	7.49E-12	1.38E-11
$Th(CO_3)_4OH^{5-}$	(mol/kg)	7.09E-12	8.63E-17
$ThCO_3(OH)_4^{2-}$	(mol/kg)	1.05E-15	7.16E-14
ThF_3^{+}	(mol/kg)		2.11E-15
ThF_4	(mol/kg)		6.38E-15
ThF_2^{2+}	(mol/kg)		8.74E-16
ThF^{3+}	(mol/kg)		1.29E-17
$Th(SO_4)_3^{2-}$	(mol/kg)		1.42E-19
$Th(SO_4)_2$	(mol/kg)		9.43E-19
$ThSO_4^{2+}$	(mol/kg)		3.25E-19
$Th(OH)_3(H_3SiO_4)_3^{2-}$	(mol/kg)	4.31E-16	1.08E-12
∑ competitive Th complexes (Th-cmp) without Th-CO_3	(mol/kg)	4.31E-16	1.09E-12
Th tot-(Th-cmp) without Th-CO_3	(mol/kg)	1.50E-11	1.02E-09
(Th tot- (Th-cmp)) / Th tot (CF-spec/b)		**1.00E+00**	**9.99E-01**

K_d-Derivation Th(IV) on Mont Terri Opalinus Clay, pH 7.8, Bradbury&Baeyens (2003b)

Overview Pred. K_d [m^3/kg]	Mudstone
	HDB-6
Pred. K_d: CF-1 (% clay)	26.13
Pred. K_d: CF-2a (% clay)	0.03
Pred. K_d: CF-2b (% clay)	26.16

Th(IV) K_d-derivation		Conditions				Conversion (C) / Uncertainty (U)
		Data source	Additional information[1]		Application	CF
Solution		Dist. equil. water	0.1 M $NaClO_4$		HDB-6	HDB-6
Substrate		Opalinus Clay	SWy-1	SWy-1	Mudstone	Mudstone
Experimental K_d	(m^3/kg)	55.4				
Conversion Mineralogy (CF-min)						
%clay		63			26	0.41
Conversion pH (CF-pH)						
pH		7.8	7.8	8.5	8.5	
Kd for 0.1 M NaClO4 (pH conversion)[2]		55.4	350	400		1.14
Conversion Speciation (CF-spec)						
CF-spec/a		0.99			1.09E-03	1.11E-03
CF-spec/b		1.00			1.00	1.00
Conversion Competition (Anionic or Cationic) (CF-cmp)	*irrelevant*					

(1) Kd values from Th(IV) sorption edge (Bradbury and Baeyens, 2005)
(2) Values of the avarage composition

Th(IV) K_d-derivation via % clay						CF
CF-1 (%clay)						0.47
Speciation						
CF-2a (% clay)						0.00
CF-2b (% clay)						0.47
						UF
Predicted K_d: CF-1 (% clay)	(m^3/kg)				**26.13**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-total						5.5
K_d upper limit	(m^3/kg)				144.94	
K_d lower limit	(m^3/kg)				4.71	
Predicted K_d: CF-2a (% clay)	(m^3/kg)				**0.03**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-speciation						1.4
UF-total						7.8
K_d upper limit	(m^3/kg)				0.22	
K_d lower limit	(m^3/kg)				0.004	
Predicted K_d: CF-2b (% clay)	(m^3/kg)				**26.16**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-speciation						1.4
UF-total						7.8
K_d upper limit	(m^3/kg)				203.13	
K_d lower limit	(m^3/kg)				3.37	

Speciation Case III

Filename Th1_equilibrium_case_III_120609_DAT Th1_B&B_opalinus_pH6.3_120618_DAT
Preequilibration_case_III_120509_DAT

TDB: JAEA,version 100331c1

Th(IV) speciation (Bradbury&Baeyens, 2003b)

System		Application	Data Source
Substrate		Horonobe Mudstone	Mont Terri Opalinus Clay
Solution		HDB-6	substrate equilibrium soln.
pH		8.50	6.30
pCO_2		-	-
Th(IV) dissolved	(mol/kg)	1.51E-11	1.02E-09
Th^{4+}	(mol/kg)	6.20E-29	3.84E-17
$Th(OH)_4$	(mol/kg)	1.69E-14	1.55E-11
$ThOH^{3+}$	(mol/kg)	7.24E-24	2.76E-14
$Th(OH)_2^{2+}$	(mol/kg)	9.83E-20	2.32E-12
$Th(CO_3)_5^{6-}$	(mol/kg)	4.54E-13	1.20E-14
$Th(CO_3)_2(OH)_2^{2-}$	(mol/kg)	7.49E-12	7.69E-10
$Th(CO_3)_4OH^{5-}$	(mol/kg)	7.09E-12	5.43E-13
$ThCO_3(OH)_4^{2-}$	(mol/kg)	1.05E-15	2.06E-15
ThF_3^{+}	(mol/kg)		4.61E-11
ThF_4	(mol/kg)		1.65E-10
ThF_2^{2+}	(mol/kg)		1.65E-11
ThF^{3+}	(mol/kg)		2.10E-13
$Th(SO_4)_3^{2-}$	(mol/kg)		5.35E-15
$Th(SO_4)_2$	(mol/kg)		2.71E-14
$ThSO_4^{2+}$	(mol/kg)		6.48E-15
$Th(OH)_3(H_3SiO_4)_3^{2-}$	(mol/kg)	4.31E-16	1.63E-17
∑ competitive Th complexes (Th-cmp) with Th-CO_3	(mol/kg)	1.50E-11	9.98E-10
Th tot-(Th-cmp)	(mol/kg)	1.64E-14	1.79E-11
(Th tot- (Th-cmp)) / Th tot (CF-spec/a)		**1.09E-03**	**1.76E-02**
$Th(CO_3)_5^{6-}$	(mol/kg)	4.54E-13	1.20E-14
$Th(CO_3)_2(OH)_2^{2-}$	(mol/kg)	7.49E-12	7.69E-10
$Th(CO_3)_4OH^{5-}$	(mol/kg)	7.09E-12	5.43E-13
$ThCO_3(OH)_4^{2-}$	(mol/kg)	1.05E-15	2.06E-15
ThF_3^{+}	(mol/kg)		4.61E-11
ThF_4	(mol/kg)		1.65E-10
ThF_2^{2+}	(mol/kg)		1.65E-11
ThF^{3+}	(mol/kg)		2.10E-13
$Th(SO_4)_3^{2-}$	(mol/kg)		5.35E-15
$Th(SO_4)_2$	(mol/kg)		2.71E-14
$ThSO_4^{2+}$	(mol/kg)		6.48E-15
$Th(OH)_3(H_3SiO_4)_3^{2-}$	(mol/kg)	4.31E-16	1.63E-17
∑ competitive Th complexes (Th-cmp) without Th-CO_3	(mol/kg)	4.31E-16	2.28E-10
Th tot-(Th-cmp) without Th-CO_3	(mol/kg)	1.50E-11	7.88E-10
(Th tot- (Th-cmp)) / Th tot (CF-spec/b)		**1.00E+00**	**7.75E-01**

K_d-Derivation Th(IV) on Mont Terri Opalinus Clay, pH 6.3, Bradbury&Baeyens (2003)

Overview Pred. K_d [m^3/kg]	Mudstone
	HDB-6
Pred. K_d: CF-1 (% clay)	20.18
Pred. K_d: CF-2a (% clay)	1.25
Pred. K_d: CF-2b (% clay)	26.02

Th(IV) K_d-derivation		Conditions				Conversion (C) / Uncertainty (U)
		Data source	Additional information[1]		Application	CF
Solution		Dist. equil. water	0.1 M $NaClO_4$		HDB-6	HDB-6
Substrate		Opalinus Clay	SWy-1	SWy-1	Mudstone	Mudstone
Experimental K_d	(m^3/kg)	22.0				
Conversion Mineralogy (CF-min)						
%clay		63			26	0.41
Conversion pH (CF-pH)						
pH		6.3	6.3	8.5	8.50	
Kd for 0.1 M NaClO4 (pH conversion)[2]		22.0	180	400		2.22
Conversion Speciation (CF-spec)						
CF-spec/a		1.76E-02			1.09E-03	0.06
CF-spec/b		0.78			1.00	1.29
Conversion Competition (Anionic or Cationic) (CF-cmp)	*irrelevant*					

(1) Kd values from Th(IV) sorption edge (Bradbury and Baeyens, 2005)
(2) Values of the avarage composition

Th(IV) K_d-derivation via % clay						CF
CF-1 (%clay)						0.92
Speciation						
CF-2a (% clay)						0.06
CF-2b (% clay)						1.18
						UF
Predicted K_d: CF-1 (% clay)	(m^3/kg)				**20.18**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-total						5.5
K_d upper limit	(m^3/kg)				111.92	
K_d lower limit	(m^3/kg)				3.64	
Predicted K_d: CF-2a (% clay)	(m^3/kg)				**1.25**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-speciation						1.4
UF-total						7.8
K_d upper limit	(m^3/kg)				9.69	
K_d lower limit	(m^3/kg)				0.16	
Predicted K_d: CF-2b (% clay)	(m^3/kg)				**26.02**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-speciation						1.4
UF-total						7.8
K_d upper limit	(m^3/kg)				202.08	
K_d lower limit	(m^3/kg)				3.35	

Speciation Case III

Filename Th1_equilibrium_case_III_120609_DAT mudstone_HDB-6_MX-80 (7.25)_Th(1)
Preequilibration_case_III_120509_DAT

TDB: JAEA,version 100331c1

Th(IV) speciation (Bradbury&Baeyens, 2003a)

System		Application	Data Source
Substrate		Horonobe Mudstone	MX-80
Solution		HDB-6	substrate equilibrium soln.
pH		8.50	7.25
pCO_2		-	-
Th(IV) dissolved	(mol/kg)	1.51E-11	1.02E-09
Th^{4+}	(mol/kg)	6.20E-29	9.12E-20
$Th(OH)_4$	(mol/kg)	1.69E-14	2.17E-10
$ThOH^{3+}$	(mol/kg)	7.24E-24	5.69E-16
$Th(OH)_2^{2+}$	(mol/kg)	9.83E-20	4.19E-13
$Th(CO_3)_5^{6-}$	(mol/kg)	4.54E-13	2.40E-15
$Th(CO_3)_2(OH)_2^{2-}$	(mol/kg)	7.49E-12	7.99E-10
$Th(CO_3)_4OH^{5-}$	(mol/kg)	7.09E-12	3.90E-13
$ThCO_3(OH)_4^{2-}$	(mol/kg)	1.05E-15	7.08E-14
ThF_3^{+}	(mol/kg)		5.43E-13
ThF_4	(mol/kg)		3.32E-12
ThF_2^{2+}	(mol/kg)		1.14E-13
ThF^{3+}	(mol/kg)		8.45E-16
$Th(SO_4)_3^{2-}$	(mol/kg)		1.75E-16
$Th(SO_4)_2$	(mol/kg)		3.56E-16
$ThSO_4^{2+}$	(mol/kg)		3.59E-17
$Th(OH)_3(H_3SiO_4)_3^{2-}$	(mol/kg)	4.31E-16	1.84E-14
∑ competitive Th complexes (Th-cmp) with Th-CO_3	(mol/kg)	1.50E-11	8.03E-10
Th tot-(Th-cmp)	(mol/kg)	1.64E-14	2.17E-10
(Th tot- (Th-cmp)) / Th tot (CF-spec/a)		**1.09E-03**	**2.13E-01**
$Th(CO_3)_5^{6-}$	(mol/kg)	4.54E-13	2.40E-15
$Th(CO_3)_2(OH)_2^{2-}$	(mol/kg)	7.49E-12	7.99E-10
$Th(CO_3)_4OH^{5-}$	(mol/kg)	7.09E-12	3.90E-13
$ThCO_3(OH)_4^{2-}$	(mol/kg)	1.05E-15	7.08E-14
ThF_3^{+}	(mol/kg)		5.43E-13
ThF_4	(mol/kg)		3.32E-12
ThF_2^{2+}	(mol/kg)		1.14E-13
ThF^{3+}	(mol/kg)		8.45E-16
$Th(SO_4)_3^{2-}$	(mol/kg)		1.75E-16
$Th(SO_4)_2$	(mol/kg)		3.56E-16
$ThSO_4^{2+}$	(mol/kg)		3.59E-17
$Th(OH)_3(H_3SiO_4)_3^{2-}$	(mol/kg)	4.31E-16	1.84E-14
∑ competitive Th complexes (Th-cmp) without Th-CO_3	(mol/kg)	4.31E-16	4.00E-12
Th tot-(Th-cmp) without Th-CO_3	(mol/kg)	1.50E-11	1.02E-09
(Th tot- (Th-cmp)) / Th tot (CF-spec/b)		**1.00E+00**	**9.96E-01**

K_d-Derivation Th(IV) on MX-80, pH 7.25, Bradbury&Baeyens (2003a)

Overview Pred. K_d [m^3/kg]	Mudstone
	HDB-6
Pred. K_d: CF-1 (% clay)	39.71
Pred. K_d: CF-2a (% clay)	0.20
Pred. K_d: CF-2b (% clay)	39.86

Th(IV) K_d-derivation		Conditions				Conversion (C) / Uncertainty (U)
		Data source	Additional information[1]		Application[2]	CF
Solution		Dist. equil. water	0.1 M $NaClO_4$		HDB-6	HDB-6
Substrate		MX-80	SWy-1	SWy-1	Mudstone	Mudstone
Experimental K_d	(m^3/kg)	63.0				
Conversion Mineralogy (CF-min)						
%clay		75			26	0.35
Conversion pH (CF-pH)						
pH		7.25	7.25	8.5	8.5	
Kd for 0.1 M NaClO4 (pH conversion)		63.0	220	400		1.82
Conversion Speciation (CF-spec)						
CF-spec/a		2.13E-01			1.09E-03	0.01
CF-spec/b		1.00			1.00	1.00
Conversion Competition (Anionic or Cationic) (CF-cmp)	*irrelevant*					

(1) Kd values from Th(IV) sorption edge (Bradbury and Baeyens, 2005)
(2) Values of the avarage composition

Th(IV) K_d-derivation via % clay						CF
CF-1 (%clay)						0.63
Speciation						
CF-2a (% clay)						0.00
CF-2b (% clay)						0.63
						UF
Predicted K_d: CF-1 (% clay)	(m^3/kg)				**39.71**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-total						5.5
K_d upper limit	(m^3/kg)				220.27	
K_d lower limit	(m^3/kg)				7.16	
Predicted K_d: CF-2a (% clay)	(m^3/kg)				**0.20**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-speciation						1.4
UF-total						7.8
K_d upper limit	(m^3/kg)				1.58	
K_d lower limit	(m^3/kg)				0.03	
Predicted K_d: CF-2b (% clay)	(m^3/kg)				**39.86**	
UF-starting K_d						1.6
UF-pH conversion						2.5
UF-min						1.4
UF-speciation						1.4
UF-total						7.8
K_d upper limit	(m^3/kg)				309.59	
K_d lower limit	(m^3/kg)				5.13	

Speciation

Filename MIZ1_solution speciation Kitamura 1999a_solution speciation

TDB: Hummel et al. (2002)

Am(II) speciation (Kitamura et al., 1999a)

System		**Application**	**Data Source**
Substrate		**MIZ1**	**Granite**
Solution			**0.01M NaClO4**
$Am(II)_{initial}$	(mol/kg)	1.80E-10	1.80E-10
pH		8.70	8.70
pCO_2		-	-
Am(II)	(mol/kg)	1.80E-10	1.80E-10
Am^{+3}	(mol/kg)	8.35E-14	3.76E-13
$AmOH^{+2}$	(mol/kg)	1.59E-12	2.48E-12
$Am(OH)_2^{+}$	(mol/kg)	8.28E-12	4.12E-12
$Am(OH)_3$	(mol/kg)	1.23E-13	1.73E-14
$AmCO_3^{+}$	(mol/kg)	6.07E-12	
$Am(CO_3)_2^{-}$	(mol/kg)	3.53E-13	
$Am(CO_3)_3^{-3}$	(mol/kg)	8.66E-16	
AmF^{+2}	(mol/kg)	8.55E-14	
AmF_2^{+}	(mol/kg)	9.49E-15	
$AmSO_{4+}$	(mol/kg)	2.75E-14	
$Am(SO_4)_2^{-}$	(mol/kg)	7.27E-17	
$AmCl^{+2}$	(mol/kg)	1.60E-15	
$AmSiO(OH)_3^{+2}$	(mol/kg)	1.63E-10	1.73E-10
∑ competitive Am complexes (Am-cmp) with Am-CO_3	(mol/kg)	1.70E-10	1.73E-10
Am tot-Am-cmp)	(mol/kg)	1.02E-11	7.00E-12
(Am tot- (Am-cmp)) / Am tot (CF-spec/a)		**5.64E-02**	**3.89E-02**
$AmCO_3^{+}$	(mol/kg)	6.07E-12	
$Am(CO_3)_2^{-}$	(mol/kg)	3.53E-13	
$Am(CO_3)_3^{-3}$	(mol/kg)	8.66E-16	
AmF^{+2}	(mol/kg)	8.55E-14	
AmF_2^{+}	(mol/kg)	9.49E-15	
$AmSO_4^{+}$	(mol/kg)	2.75E-14	
$Am(SO_4)_2^{-}$	(mol/kg)	7.27E-17	
$AmCl^{+2}$	(mol/kg)	1.60E-15	
$AmSiO(OH)_3^{+2}$	(mol/kg)	1.63E-10	1.73E-10
	(mol/kg)		
Am complexes (Am-cmp) without Am-CO_3	(mol/kg)	1.63E-10	1.73E-10
Am tot-(Am-cmp) without Am-CO4	(mol/kg)	1.66E-11	7.00E-12
(Am tot- (Am-cmp)) / Am tot (CF-spec/b)		**9.214E-02**	**3.889E-02**

K_d-Derivation Am(III) on garanite, pH 8.7, Kitamura et al. (1999a)

Overview Pred. K_d [m^3/kg]	via BET
Pred. K_d: CF-1 (% clay)	111.36
Pred. K_d: CF-2a (% clay)	161.63
Pred. K_d: CF-2b (% clay)	263.85

Am(II) K_d-derivation		Conditions		Conversion (C) / Uncertainty (U) Factor(F)
		Data source[1]	Application[2]	CF
Solution		**0.01M NaClO4**	**granite water**	**granite water**
Substrate		**Granite**	**MIZ1**	**MIZ1**
Experimental K_d	(m^3/kg)	35		
Conversion Mineralogy (CF-min)				
CEC	(meq/100g)		1.80	1.0
BET	(m^2/g)	0.11	0.35	3.18
% mica			4.0	1.0
Conversion pH (CF-pH)				
pH		8.70	8.7	
Kd for 1.0 M NaClO4 (pH conversion)		160		1.00
Conversion Speciation (CF-spec)				
CF-spec/a		0.04	0.06	1.45
CF-spec/b		0.04	0.09	2.37
Conversion Competition (Anionic or Cationic) (CF-cmp)	*irrelevant*			

(1) K_d value from Am(II) sorption datapoint Kitamura et al. (1999a)
(2) Values provided by JAEA

Am(II) K_d-derivation via BET				CF
CF-1 (BET)				3.18
Speciation				
CF-2a (BET)				4.62
CF-2b (BET)				7.54
				UF
Predicted K_d: CF-1 (BET)	(m^3/kg)		**111.36**	
UF-starting K_d				1.6
UF-pH conversion				
UF-min				1.4
UF-total				2.2
Kd upper limit	(m^3/kg)		247.10	
Kd lower limit	(m^3/kg)		50.19	
positive error bar			*135.74*	
negative error bar			*61.17*	
Predicted K_d: CF-2a (BET)	(m^3/kg)		**161.63**	
UF-starting K_d				1.6
UF-pH conversion				
UF-min				1.4
UF-speciation				1.4
UF-total				3.1
K_d upper limit	(m^3/kg)		502.08	
K_d lower limit	(m^3/kg)		52.03	
positive error bar			*340.45*	
negative error bar			*109.60*	
Predicted K_d: CF-2b (BET)	(m^3/kg)		**263.85**	
UF-starting K_d				1.6
UF-pH conversion				
UF-min				1.4
UF-speciation				1.4
UF-total				3.1
K_d upper limit	(m^3/kg)		819.63	
K_d lower limit	(m^3/kg)		84.94	
positive error bar			*555.78*	
negative error bar			*178.91*	

国際単位系（SI）

表１．SI 基本単位

基本量	SI 基本単位	
	名称	記号
長さ	メートル	m
質量	キログラム	kg
時間	秒	s
電流	アンペア	A
熱力学温度	ケルビン	K
物質量	モル	mol
光度	カンデラ	cd

表２．基本単位を用いて表されるSI組立単位の例

組立量	SI 基本単位	
	名称	記号
面積	平方メートル	m^2
体積	立法メートル	m^3
速さ，速度	メートル毎秒	m/s
加速度	メートル毎秒毎秒	m/s^2
波数	毎メートル	m^{-1}
密度，質量密度	キログラム毎立方メートル	kg/m^3
面積密度	キログラム毎平方メートル	kg/m^2
比体積	立方メートル毎キログラム	m^3/kg
電流密度	アンペア毎平方メートル	A/m^2
磁界の強さ	アンペア毎メートル	A/m
量濃度[a]，濃度	モル毎立方メートル	mol/m^3
質量濃度	キログラム毎立法メートル	kg/m^3
輝度	カンデラ毎平方メートル	cd/m^2
屈折率[b]	（数字の） 1	1
比透磁率[b]	（数字の） 1	1

(a) 量濃度（amount concentration）は臨床化学の分野では物質濃度（substance concentration）ともよばれる。
(b) これらは無次元量あるいは次元１をもつ量であるが、そのことを表す単位記号である数字の１は通常は表記しない。

表３．固有の名称と記号で表されるSI組立単位

組立量	SI 組立単位			
	名称	記号	他のSI単位による表し方	SI基本単位による表し方
平面角	ラジアン[b]	rad	1 [b]	m/m
立体角	ステラジアン[b]	sr[c]	1 [b]	m^2/m^2
周波数	ヘルツ[d]	Hz		s^{-1}
力	ニュートン	N		$m\ kg\ s^{-2}$
圧力，応力	パスカル	Pa	N/m^2	$m^{-1}\ kg\ s^{-2}$
エネルギー，仕事，熱量	ジュール	J	N m	$m^2\ kg\ s^{-2}$
仕事率，工率，放射束	ワット	W	J/s	$m^2\ kg\ s^{-3}$
電荷，電気量	クーロン	C		s A
電位差（電圧），起電力	ボルト	V	W/A	$m^2\ kg\ s^{-3}\ A^{-1}$
静電容量	ファラド	F	C/V	$m^{-2}\ kg^{-1}\ s^4\ A^2$
電気抵抗	オーム	Ω	V/A	$m^2\ kg\ s^{-3}\ A^{-2}$
コンダクタンス	ジーメンス	S	A/V	$m^{-2}\ kg^{-1}\ s^3\ A^2$
磁束	ウエーバ	Wb	Vs	$m^2\ kg\ s^{-2}\ A^{-1}$
磁束密度	テスラ	T	Wb/m^2	$kg\ s^{-2}\ A^{-1}$
インダクタンス	ヘンリー	H	Wb/A	$m^2\ kg\ s^{-2}\ A^{-2}$
セルシウス温度	セルシウス度[e]	℃		K
光束	ルーメン	lm	cd sr[c]	cd
照度	ルクス	lx	lm/m^2	$m^{-2}\ cd$
放射性核種の放射能[f]	ベクレル[d]	Bq		s^{-1}
吸収線量, 比エネルギー分与, カーマ	グレイ	Gy	J/kg	$m^2\ s^{-2}$
線量当量, 周辺線量当量, 方向性線量当量, 個人線量当量	シーベルト[g]	Sv	J/kg	$m^2\ s^{-2}$
酸素活性	カタール	kat		$s^{-1}\ mol$

(a)SI接頭語は固有の名称と記号を持つ組立単位と組み合わせても使用できる。しかし接頭語を付した単位はもはやコヒーレントではない。
(b)ラジアンとステラジアンは数字の１に対する単位の特別な名称で、量についての情報をつたえるために使われる。実際には、使用する時には記号rad及びsrが用いられるが、習慣として組立単位としての記号である数字の１は明示されない。
(c)測光学ではステラジアンという名称と記号srを単位の表し方の中に、そのまま維持している。
(d)ヘルツは周期現象についてのみ、ベクレルは放射性核種の統計的過程についてのみ使用される。
(e)セルシウス度はケルビンの特別な名称で、セルシウス温度を表すために使用される。セルシウス度とケルビンの単位の大きさは同一である。したがって、温度差や温度間隔を表す数値はどちらの単位で表しても同じである。
(f)放射性核種の放射能（activity referred to a radionuclide）は、しばしば誤った用語で"radioactivity"と記される。
(g)単位シーベルト（PV,2002,70,205）についてはCIPM勧告2（CI-2002）を参照。

表４．単位の中に固有の名称と記号を含むSI組立単位の例

組立量	SI 組立単位		
	名称	記号	SI 基本単位による表し方
粘度	パスカル秒	Pa s	$m^{-1}\ kg\ s^{-1}$
力のモーメント	ニュートンメートル	N m	$m^2\ kg\ s^{-2}$
表面張力	ニュートン毎メートル	N/m	$kg\ s^{-2}$
角速度	ラジアン毎秒	rad/s	$m\ m^{-1}\ s^{-1}=s^{-1}$
角加速度	ラジアン毎秒毎秒	rad/s^2	$m\ m^{-1}\ s^{-2}=s^{-2}$
熱流密度，放射照度	ワット毎平方メートル	W/m^2	$kg\ s^{-3}$
熱容量，エントロピー	ジュール毎ケルビン	J/K	$m^2\ kg\ s^{-2}\ K^{-1}$
比熱容量，比エントロピー	ジュール毎キログラム毎ケルビン	J/(kg K)	$m^2\ s^{-2}\ K^{-1}$
比エネルギー	ジュール毎キログラム	J/kg	$m^2\ s^{-2}$
熱伝導率	ワット毎メートル毎ケルビン	W/(m K)	$m\ kg\ s^{-3}\ K^{-1}$
体積エネルギー	ジュール毎立方メートル	J/m^3	$m^{-1}\ kg\ s^{-2}$
電界の強さ	ボルト毎メートル	V/m	$m\ kg\ s^{-3}\ A^{-1}$
電荷密度	クーロン毎立方メートル	C/m^3	$m^{-3}\ sA$
表面電荷	クーロン毎平方メートル	C/m^2	$m^{-2}\ sA$
電束密度，電気変位	クーロン毎平方メートル	C/m^2	$m^{-2}\ sA$
誘電率	ファラド毎メートル	F/m	$m^{-3}\ kg^{-1}\ s^4\ A^2$
透磁率	ヘンリー毎メートル	H/m	$m\ kg\ s^{-2}\ A^{-2}$
モルエネルギー	ジュール毎モル	J/mol	$m^2\ kg\ s^{-2}\ mol^{-1}$
モルエントロピー, モル熱容量	ジュール毎モル毎ケルビン	J/(mol K)	$m^2\ kg\ s^{-2}\ K^{-1}\ mol^{-1}$
照射線量（X線及びγ線）	クーロン毎キログラム	C/kg	$kg^{-1}\ sA$
吸収線量率	グレイ毎秒	Gy/s	$m^2\ s^{-3}$
放射強度	ワット毎ステラジアン	W/sr	$m^4\ m^{-2}\ kg\ s^{-3}=m^2\ kg\ s^{-3}$
放射輝度	ワット毎平方メートル毎ステラジアン	$W/(m^2\ sr)$	$m^2\ m^{-2}\ kg\ s^{-3}=kg\ s^{-3}$
酵素活性濃度	カタール毎立方メートル	kat/m^3	$m^{-3}\ s^{-1}\ mol$

表５．SI 接頭語

乗数	接頭語	記号	乗数	接頭語	記号
10^{24}	ヨタ	Y	10^{-1}	デシ	d
10^{21}	ゼタ	Z	10^{-2}	センチ	c
10^{18}	エクサ	E	10^{-3}	ミリ	m
10^{15}	ペタ	P	10^{-6}	マイクロ	μ
10^{12}	テラ	T	10^{-9}	ナノ	n
10^{9}	ギガ	G	10^{-12}	ピコ	p
10^{6}	メガ	M	10^{-15}	フェムト	f
10^{3}	キロ	k	10^{-18}	アト	a
10^{2}	ヘクト	h	10^{-21}	ゼプト	z
10^{1}	デカ	da	10^{-24}	ヨクト	y

表６．SIに属さないが、SIと併用される単位

名称	記号	SI 単位による値
分	min	1 min=60s
時	h	1h =60 min=3600 s
日	d	1 d=24 h=86 400 s
度	°	1°=(π/180) rad
分	’	1’=(1/60)°=(π/10800) rad
秒	”	1”=(1/60)’=(π/648000) rad
ヘクタール	ha	$1ha=1hm^2=10^4m^2$
リットル	L，l	$1L=1=1dm^3=10^3cm^3=10^{-3}m^3$
トン	t	$1t=10^3\ kg$

表７．SIに属さないが、SIと併用される単位で、SI単位で表される数値が実験的に得られるもの

名称	記号	SI 単位で表される数値
電子ボルト	eV	$1eV=1.602\ 176\ 53(14)\times10^{-19}J$
ダルトン	Da	$1Da=1.660\ 538\ 86(28)\times10^{-27}kg$
統一原子質量単位	u	1u=1 Da
天文単位	ua	$1ua=1.495\ 978\ 706\ 91(6)\times10^{11}m$

表８．SIに属さないが、SIと併用されるその他の単位

名称	記号	SI 単位で表される数値
バール	bar	$1\ bar=0.1MPa=100kPa=10^5Pa$
水銀柱ミリメートル	mmHg	1mmHg=133.322Pa
オングストローム	Å	$1\ Å=0.1nm=100pm=10^{-10}m$
海里	M	1 M=1852m
バーン	b	$1\ b=100fm^2=(10^{-12}cm)2=10^{-28}m^2$
ノット	kn	1 kn=(1852/3600)m/s
ネーパ	Np	SI単位との数値的な関係は、対数量の定義に依存。
ベル	B	
デジベル	dB	

表９．固有の名称をもつCGS組立単位

名称	記号	SI 単位で表される数値
エルグ	erg	$1\ erg=10^{-7}\ J$
ダイン	dyn	$1\ dyn=10^{-5}N$
ポアズ	P	$1\ P=1\ dyn\ s\ cm^{-2}=0.1Pa\ s$
ストークス	St	$1\ St=1cm^2\ s^{-1}=10^{-4}m^2\ s^{-1}$
スチルブ	sb	$1\ sb=1cd\ cm^{-2}=10^4cd\ m^{-2}$
フォト	ph	$1\ ph=1cd\ sr\ cm^{-2}\ 10^4lx$
ガル	Gal	$1\ Gal=1cm\ s^{-2}=10^{-2}ms^{-2}$
マクスウェル	Mx	$1\ Mx = 1G\ cm^2=10^{-8}Wb$
ガウス	G	$1\ G =1Mx\ cm^{-2} =10^{-4}T$
エルステッド[c]	Oe	$1\ Oe \triangleq (10^3/4\pi)A\ m^{-1}$

(c) ３元系のCGS単位系とSIでは直接比較できないため、等号「 ≙ 」は対応関係を示すものである。

表10．SIに属さないその他の単位の例

名称	記号	SI 単位で表される数値
キュリー	Ci	$1\ Ci=3.7\times10^{10}Bq$
レントゲン	R	$1\ R = 2.58\times10^{-4}C/kg$
ラド	rad	$1\ rad=1cGy=10^{-2}Gy$
レム	rem	$1\ rem=1\ cSv=10^{-2}Sv$
ガンマ	γ	1 γ =1 nT=10-9T
フェルミ		1フェルミ=1 fm=10-15m
メートル系カラット		1メートル系カラット = 200 mg = 2×10-4kg
トル	Torr	1 Torr = (101 325/760) Pa
標準大気圧	atm	1 atm = 101 325 Pa
カロリー	cal	1cal=4.1858J（「15℃」カロリー），4.1868J（「IT」カロリー）4.184J（「熱化学」カロリー）
ミクロン	μ	$1\ \mu =1\mu m=10^{-6}m$

（第8版，2006年改訂）