

Outline of the experimental approach adopted by SCK•CEN for developing radionuclide sorption parameters

-period 2007-2013-

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Report to ONDRAF/NIRAS prepared in the frame of contract CCHO 2004-2470/00/00, DS251-A44/2.1 RP.WD.037 Radionuclide Migration and Retention in Boom Clay

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RDD

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September, 2008

Status: Unclassified

ISSN 1782-2335

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Operational Office: Boeretang 200 – BE-2400 MOL

GLOSSARY

CA	Component Additivity
GC	Generalised Composite
K_d	Distribution coefficient, assuming equilibrium between solid and solution phase
PA	Performance assessment
R	Retardation factor
R_d	Distribution coefficient, not assuming equilibrium between solid and solution phase
RBCW	Real Boom Clay water
RN	Radionuclide
SBCW	Synthetic Boom Clay water
TSM	Thermodynamic sorption model

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1. Introduction

The present document refers to the experimental programme on sorption (or, more general, "uptake"¹) that is developed and adopted by SCK•CEN in the framework of the deep geological disposal research financed by NIRAS/ONDRAF. The present experimental programme is considered as part of the scientific underbuilding of the Safety and Feasibility Case 1 (SFC-1), foreseen for 2013. For a general approach on the development of a migration parameter database within this SFC-1, and the selection strategy for such parameters, the reader is referred to the appropriate document (currently labelled "Integrated strategy for selection of radionuclide migration parameters", Bruggeman *et al.*, in preparation as a L5 Topical Report for SFC-1).

For a more historic overview on how sorption of radionuclides has been treated in the Belgian R&D programme for geological disposal before the currently considered period, the reader is referred to the Sorption Position Paper (Wang *et al.*, 2004). Following an expert review (Ochs, 2007) on this past approach by M.Ochs as requested by NIRAS/ONDRAF (review meeting held in October 2006), the general strategy on sorption was reconsidered and accumulated in the drafting of the present document. This document has again been reviewed by M.Ochs (review meeting held in September 2007).

¹ By using the more general term "uptake", all processes related to chemical interactions between dissolved radionuclide species and the solid Boom Clay phase are comprised. These processes include adsorption to mineral phases, complexation with the solid kerogen fraction, surface precipitation/solid solution formation, etc.

2. Underlying strategy for the experimental programme on uptake

The principal objective of any facility for the disposal of radioactive waste is to provide long-term safety by isolation and confinement. Dose rates resulting from the release of such waste under disposal conditions can be estimated by performance assessment (PA) calculations that rely on knowledge and understanding of the migration behaviour of radionuclides in the geological formation. In Belgium, the Boom Clay is currently considered as the reference host formation for phenomenological studies. Therefore performance assessment calculations are focussed on transport predictions under the currently prevailing Boom Clay geochemical conditions. As a simplifying assumption within the present experimental programme, we consider these conditions to be homogeneous both in vertical as in horizontal direction (in the Campine area), and stable in time.

Radionuclide sorption is one of the most important processes which provide the prevention (or sufficient retardation) of radionuclide migration to the biosphere (NEA, 2001). The performance assessment parameter which relates to this process, is the retardation factor (R). This factor is a function of the partitioning of a radionuclide between the solid phase and the solution, the bulk dry density and the diffusion accessible porosity of the considered medium (see below). Retardation factors applicable for *in situ* conditions can in principle be determined directly from migration (diffusion) experiments on undisturbed compacted clay cores. However, it often proves to be impossible to derive this factor unambiguously, while on the other hand the nominal value of R does not provide any information concerning 1) the underlying uptake processes, and 2) its sensitivity towards possibly occurring changing conditions.

As an input to PA, retardation factors are in general coupled to distribution coefficients (NEA, 2005). These coefficients may be K_d values, assuming that radionuclide uptake during transport is in (thermodynamic) equilibrium:

$$K_d \text{ [l/kg]} = \{C\}_{\text{solid,eq}} / [C]_{\text{liquid,eq}}$$

with $\{C\}_{\text{solid,eq}}$ the concentration of a contaminant adsorbed on the solid phase (mol/kg) at equilibrium, and $[C]_{\text{liquid,eq}}$ the corresponding concentration of the contaminant in the liquid phase (mol/l); or they may be R_d values, when equilibrium conditions are not met due to e.g. kinetic factors

$$R_d \text{ [l/kg]} = \{C\}_{\text{solid,exp}} / [C]_{\text{liquid,exp}}$$

with $\{C\}_{\text{solid,exp}}$ and $[C]_{\text{liquid,exp}}$ the experimentally determined concentrations of a contaminant on the solid phase (mol/kg) and in the liquid phase (mol/l), respectively.

Experiments to determine K_d/R_d values are of the batch/suspension-type and can be classified into two groups. In the first group, the *in situ* geochemical conditions are mimicked in the highest possible way by using suspensions of solid Boom Clay in Synthetic Boom Clay Water (SBCW)² or in Real Boom Clay Water (RBCW)³. Such experiments allow for limited parameter variation (e.g. isotherms) and can provide some process understanding. The main challenge lies

² Synthetic Boom Clay Water (SBCW): inorganic, synthetic equivalent of Boom Clay (piezometer) pore water; mainly used in laboratory experiments to mimic the real Boom Clay water.

³ Real Boom Clay Water (RBCW): Boom Clay pore water, containing dissolved organic carbon; the pore water is mainly collected from the EG/BS vertical piezometer.

in the upscaling of these batch K_d/R_d values to uptake during transport in the compacted state. In the second group, simplified batch systems containing relatively pure and well-characterised geochemical phases are used to elucidate uptake processes by allowing variation of different geochemical parameters (pH, ionic strength, sorbate concentration, presence of competing sorbates). The geochemical phases are selected as they represent or mimic the principle components of Boom Clay which control or dominate uptake of the radionuclide in question. The study of such well-characterised and relatively simple systems will allow for the development of Thermodynamic Sorption Models (TSMs) which can support the selection of a K_d/R_d value for performance assessment, and which can predict or rather interpolate uptake values under changed geochemical conditions. However, because of the relative simplicity of these systems compared to the intrinsic complex nature of the host rock, transferability of results from these experiments to parameters needed for PA is mostly a very delicate task, and often relies on expert judgement.

In the ideal case, where sorption is linear and reversible, the K_d value is a unique value for all radionuclide concentrations. In the latter case, the relationship between K_d and R is given by the following relationship:

$$R [-] = 1 + \rho \times K_d / \eta$$

with ρ the bulk dry density of Boom Clay ($\sim 1.7 \text{ kg/dm}^3$) and η the diffusion accessible porosity (0.37 for HTO). Ideally, the results of all above-mentioned types of experiments (migration experiments, batch studies on complex substrates, batch studies on "pure" geochemical phases) would lead to a single coherent model which allows to describe and predict sorption under variable geochemical conditions.

Taking into account the above observations, the present experimental programme for studying radionuclide uptake on the Boom Clay, is based on a two-pronged strategy, which is also developed in the framework of SFC-1 for other transport parameters: 1) to **acquire and select sorption parameters** (essentially $K_d/R_d/R$'s) that serve as an input for PA calculations; 2) to scientifically underpin the selected $K_d/R_d/R$ values by **demonstrating a comprehensive understanding** of the radionuclide uptake processes in the host formation ("confidence building"). This latter goal necessitates the ability to sufficiently describe the most relevant radionuclide uptake processes by means of (quasi-)mechanistic chemical formulations. The scheme given below ([figure 1](#)) summarises the types of laboratory experiments performed to come to a sorption database, which will allow for a selection of retardation parameters for SFC-1.

Although the above-mentioned two-pronged strategy of the research programme on uptake is quite straightforward, it is not possible to achieve especially the second goal for all radionuclides that are considered in PA calculations due to the limited time and resources with respect to the SFC-1 finalisation date in 2013. Therefore, the (quasi-)mechanistic understanding of the geochemical behaviour in the host formation will at the moment be demonstrated only for a limited set of radionuclides. The latter are selected because of their representativeness for a whole group of nuclides which display similar chemical and/or uptake behaviour.

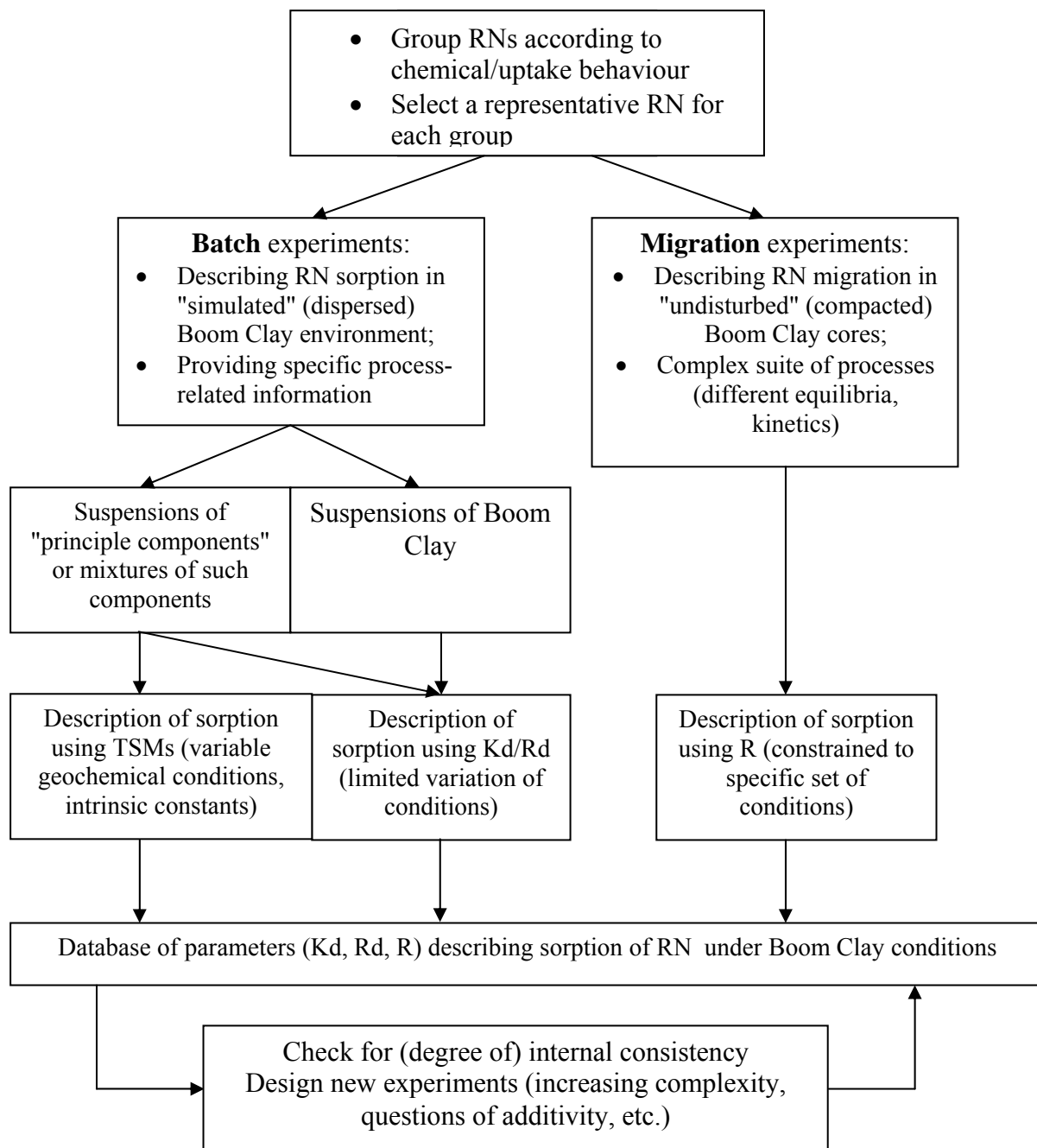


Figure 1: Scheme describing the experimental strategy to come to a scientifically underbuilt sorption database

Currently, four different groups of RN's are considered, characterized by an increasing range of complexity in interaction mechanisms, which roughly coincides with their charge and oxidation state (I, II, III, and IV-valent cations). More specifically, the division of nuclides in such groups is based on scientific considerations (chemical analogy, *in situ* speciation, literature data, etc.), while representatives for each group are chosen based on the availability of transport data from previous migration experiments:

1. Behaviour dominated by ion exchange – represented by Cs⁺
2. Behaviour dominated by ion exchange and isotopic exchange⁴ – represented by Sr²⁺
3. Behaviour dominated by adsorption (ion exchange, surface complexation) on clay minerals and interaction with natural organic matter – represented by Eu³⁺ and Am³⁺
4. Behaviour dominated by adsorption (ion exchange, surface complexation) on clay minerals and interaction with natural organic matter, with special attention to colloidal behaviour – represented by Th⁴⁺ and U⁴⁺

These four groups are by no means exhaustive, and other groups may be added when considered necessary. Such groups may include, for example:

5. (Oxy)anions
6. Transition metals whose behaviour cannot be linked to one of the above groups
7. Amphoteric elements

A crucial step in the strategy consists in setting up an experimental "bottom-up" approach which will allow to gain both qualitative and quantitative insight into the uptake mechanisms of a selected radionuclide (RN) on (the relevant geochemical phases of) the Boom Clay. In such an approach the geochemical behaviour of a RN in Boom Clay will be described by considering in the first instance only the geochemical phase(s) which is (are) dominant for the speciation of the RN in question. This "bottom-up" approach represents in fact a "slim-line" version of a completely exhaustive "Component Additivity" (CA) approach⁵, which due to the geochemical complexity of the Boom Clay is considered not feasible. More specifically, major objections to the elaboration of an exhaustive CA approach are, amongst others, the present lack of knowledge on interactions between different geochemical phases (and the way to describe them), the vast amount of work which would be needed to fully describe uptake for all RN's on all Boom Clay components which may have an influence on speciation, and the current lack of scientific consensus on the applicability of a CA approach to predict geochemical behaviour in complex matrices (Payne *et al.*, 2004).

The rationale which is derived from the adoption of the "bottom-up" approach can be described as follows: for each RN the principal geochemical components of the Boom Clay which are assumed to dominate its uptake (and therefore speciation), are selected based on 1) in-house knowledge on the (mineralogical) composition of Boom Clay; 2) a review of available literature data; and 3) experiences from migration experiments. Hereafter, interaction studies (surface complexation, organic complexation, ion exchange) with each of these assumed principal components can be designed and performed in order to obtain model parameters which are necessary to describe the interaction mechanism. In case of pure clay minerals (illite, montmorillonite), the available literature may already provide good quality data sets (and consistent models) and no additional in-house experiences are needed. The focus of interaction

⁴ There is isotopic exchange when, for instance, a radioactive tracer (e.g. Co-60) is added to a system already containing non-active isotopes of the same element (e.g. Co-59). Subsequently, an exchange will take place between the radioactive isotopes and the (readily available) non-active isotopes until an equal partitioning is observed (Kd[Co-60] = Kd[Co-59])

⁵ Component additivity approach: modelling approach in which it is attempted to *predict* adsorption on a complex mineral assemblage using the results of a surface characterisation of the assemblage and collected data for adsorption by pure, reference minerals or organic phases. It is assumed in this approach that the wetted surface of the complex mineral assemblage is composed of a mixture of one or more mineral (or organic) phases whose surface properties are known from independent studies of the individual phases (Davis *et al.*, 1998)

studies is therefore expected to be centered on geochemical phases which are specific for Boom Clay, e.g. kerogen, dissolved organic matter, interlayered illite-smectite, etc.

Geochemical modelling, applying a straight-forward additivity rule when several phases are involved, is then used to predict the results from experiments on Boom Clay batch systems (the latter were in the past frequently analysed using a "generalised composite" (GC) approach⁶). The outcome of this comparison can then be coupled back to the initial assumptions which were made on the choice of the principal component(s), and if necessary, CA model parameters can be adapted to better fit the uptake on the complex assemblage (Payne *et al.*, 2004).

Therefore, the major benefit of the "bottom-up" approach is that it will allow to provide more insight into the main speciation-controlling mechanisms for each (class of) radionuclide(s), and to obtain information on the parameter sensitivity upon changing geochemical conditions, while limiting the amount of experimental work necessary to understand the geochemical behaviour in the complex Boom Clay. The major flaw is related to (the presently unresolved question of) the applicability of the additivity rule in very complex phase assemblages such as the Boom Clay. This applicability may be proven to be valid for some radionuclides, but may not be generalized and lead to large under- or overestimates on uptake for others (e.g. Vermeer *et al.*, 1999).

⁶ Generalised composite approach: modelling approach in which it is assumed that the surface composition of the mineral assemblage is inherently too complex to be quantified in terms of the contributions of individual phases to adsorption. Instead, it is assumed that the adsorptive reactivity of the surface can be described by surface complexation equilibria written with "generic" surface functional groups, with the stoichiometry and formation constants for each surface complex equilibria determined by fitting experimental data (Davis *et al.*, 1998).

3. Details for the experimental programme

The *geochemical codes* used for describing experimental systems, and for making predictions, are Phreeqc (version 2) (Parkhurst and Appelo, 1999) and The Geochemist's Workbench (GWB) (release 7.0) (Bethke, 2006).

Primary *thermodynamic data sources* used in the geochemical codes are the LLNL, NAPS and NEA databases. Within the frame of the so-called MOLDATA project, these sources are currently being compiled (finalisation date December 2008) to enable a selection of internally consistent aqueous species and solid phases that are considered to be relevant for geochemical calculations of the Boom Clay system.

The *principle components* of the Boom Clay, or model geochemical phases mimicking such components, which have been and will be used in the interaction studies, and which are presumed to be of importance for understanding, describing and predicting uptake for most radionuclides, are listed in [table 1](#).

Table 1: Principle Boom Clay components that are currently considered to be used in the "bottom-up" approach

Phase type	Specific phase	Interaction type	Comments
Inorganic	Illite ⁷	Adsorption: surface complexation and ion exchange	Main constituting clay phase in Boom Clay
	Montmorillonite ⁸		Taken as a possible alternative model for illite-smectite mixed-layer
	Bentonite MX-80 (Volclay)		Will only occasionally be used; relevant phase in backfill material studies, reference phase in international literature
Organic	Humic substances in Real Boom Clay water (RBCW) ⁹	Complexation, colloidal interaction	Non-purified
	Boom Clay humic acid		Purified based on IHSS ¹⁰ method
	Aldrich humic acid		Reference phase for comparison with international literature
	Boom Clay kerogen (IFP) ¹¹	Complexation (?)	Purified (as received)
	Boom Clay kerogen ¹²		

⁷ Illite du Puy, FR, obtained from CEA, or IMt-2 Illite, Silver Hill Montana, USA, purchased from the The Clay Minerals Society.

⁸ SWy-2 Na-Montmorillonite, Crook County, Wyoming, USA, purchased from the Clay Minerals Society

⁹ Real Boom Clay water (RBCW) sampled from the underground laboratory at the Mol site

¹⁰ IHSS: International Humic Substance Society, <http://www.ihss.gatech.edu/>

¹¹ Kerogen extracted from Boom Clay by the Institut Français du Pétrole (IFP - FR) (Durand and Nicaise, 1980)

The components list in [table 1](#) is not exhaustive: new phases (e.g. mixed-layer illite-smectite, calcite, pyrite) may be included if considered necessary. However, for the time being we assume that such phases only serve to impose geochemical conditions (e.g. E_h for pyrite, carbonate buffer for calcite) which can be treated during geochemical modelling without taking into account the phase as a surface available for uptake.

Table 2: Geochemical models currently considered and evaluated for describing interactions with selected geochemical phases. Models that are already used in the present status of the uptake programme, are highlighted in grey.

Geochemical model	Literature reference	Interaction type and geochemical phase	RN concerned
3-site Ion exchange model (3S IEX)	Bradbury M.H. and Baeyens B., <i>J. Cont. Hydrol.</i> , 42 , 141-163 (2000)	Ion exchange on illite	Cs
2-site protolysis non-electrostatic surface complexation/cation exchange (2 SPNE SC/CE)	Bradbury M.H. and Baeyens B., <i>J. Cont. Hydrol.</i> , 27 , 223-248 (1997) Bradbury M.H. <i>et al.</i> , <i>Geochim. Cosmochim. Acta</i> , 69 , 5403-5412 (2005)	Ion exchange and surface complexation on illite and montmorillonite	Most cations and anions
Charge distribution – Multi Site Complexation (CD-MUSIC)	Hiemstra <i>et al.</i> , <i>J. Colloid Interface Sci.</i> , 131 , 91-104 (1989) Hiemstra T. and van Riemsdijk W.H., <i>J. Colloid Interface Sci.</i> , 179 , 488-508 (1996)	Surface complexation on (hydr)oxide surfaces <i>Currently only available for metal (hydr) oxides, not for clays; probably too complicated to be of reference for Boom Clay</i>	
Humic Ion-binding model VI (Tipping VI)	Tipping E., <i>Aquatic Geochem.</i> , 4 , 3-47 (1998)	Complexation with humic substances	Most cations
Non-ideal competitive (consistent) adsorption (NIC(C)A)-Donnan	Koopal <i>et al.</i> , <i>Colloids Surf. A</i> , 265 , 40–54 (2005) Kinniburgh <i>et al.</i> , <i>Colloids Surf. A</i> , 151 , 147-166 (1999)	Complexation with humic substances <i>Currently unavailable within Phreeqc and GWB codes</i>	Most cations
Ligand and Charge Distribution model (LCD)	Weng <i>et al.</i> , <i>J. Coll. Interface Sci.</i> , 302 , 442-457 (2006).	Ternary systems: metals + oxide + humic substances <i>Currently in a developing phase; it will be investigated whether more conventional models can be used to describe ternary systems</i>	

¹² In case new extraction procedures become available (e.g. PhD performed at Université de Nancy – R. Michels) and/or when it appears that IFP kerogen is not useful

To describe the specific interaction mechanisms, also given in [table 1](#), of RN's with each of these phases, appropriate *geochemical models* are selected. These models must ideally fulfill the following two requirements: 1) provide intrinsic parameters which are independent of geochemical conditions¹³ (RN concentration, pH, ionic strength, competition with other metals and RN's); 2) be internally (e.g. thermodynamically) consistent. The selected geochemical models are given in [table 2](#). The models that are at present actively explored are highlighted.

By using these wide-spread and well-established models, whose value has already been appreciated through numerous international literature citations, SCK•CEN's programme on uptake is simultaneously benchmarked with respect to other (international) research groups who are working in the same scientific field. International cooperation, stimulating the verification and validity of model use and the exchange of parameters obtained, will equally be strived for (e.g. with M.H. Bradbury and B. Baeyens of the Paul Scherrer Institute (PSI) – CH). It is remarked that currently, to our knowledge no models are available for describing RN interactions with kerogen. However, by considering that kerogen is a type of humic substance (albeit immobile), we assume for now that complexation-type models may be applicable.

The *experimental design* that complies to the strategy on uptake, is aimed at providing appropriate database parameters for the geochemical models listed in [table 2](#). Generally speaking, these parameters fall into two broad categories: 1) parameters that are intrinsic for the component under consideration and describe its geochemical behaviour, such as protonation/deprotonation constants of clay and oxide surfaces, site capacities (ion exchange, surface complexation) of clay minerals, humic substance functional group deprotonation constants, humic substance proton exchange capacity, etc.; 2) parameters that describe the uptake of radionuclides on the component under consideration, such as surface complexation constants on clay/oxide surface sites, complexation constants with carboxylic and phenolic functional groups of humic substances, etc.

Based on the two categories of parameters needed as an input for geochemical modelling, the experimental design similarly consists of two experiment types: 1) experiments to characterise the selected geochemical reference phases (principal Boom Clay components), examples include surface acid-base titrations, ion-exchange capacity measurements, etc.; 2) experiments to provide radionuclide-specific interaction parameters, examples include sorption measurements on surfaces as a function of pH (sorption edges), sorption measurements as a function of RN concentration (sorption isotherms), solubility measurements in presence of humic substances above a well-characterised precipitate, etc. In general it will not be necessary to determine all parameters for all phases and involved elements. Indeed, as the selected geochemical models are already extensively used in international literature, for a number of phases and radionuclides it will be possible to select "generic" parameter values a priori, and then to compare them for applicability to the site-specific (Boom Clay) situation.

The experiments that have been performed, are currently ongoing, and are foreseen for the near future are presented in a separate (attached) Excel-file.

¹³ Intrinsic parameters, as opposed to conditional parameters, are independent of the geochemical conditions in which a particular experiment is performed. Thermodynamic constants are examples of intrinsic parameters, while K_d is a typical example of a conditional parameter.

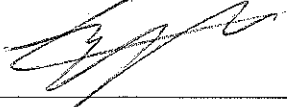

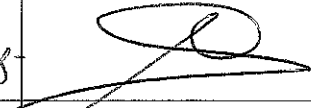
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Annex I

Uptake experimental matrix, status December 2007

Radionuclide	Batch/Migration	Interaction mechanism	Geochemical phase	Parameters	Source of parameter value	Comment	Status	Modelling result
Cesium (Cs)	Batch	Ion exchange	Illite	Exchange capacities	Literature	<i>Bradbury and Baeyens, J.Cont.Hydrol., 42 (2000), 141-163</i>	Done	OK
				Selectivity coefficients	Literature		Done	OK
				Exchange capacities	Adsorption isotherm		Done	OK, successful fit
				Selectivity coefficients	Adsorption isotherm		Done	OK, successful fit
	Migration	Ion exchange	Boom Clay	Retardation factor	Column migraton	Compare with model prediction for Boom Clay uptake in batch	Done	Fit after parameter adaptation
	Migration	Ion exchange	Boom Clay	Retardation factor	In-diffusion	Compare with model prediction for Boom Clay uptake in batch	In progress	
Strontium (Sr)	Batch	Ion exchange	Illite, montmorillonite	Exchange capacities	Literature	<i>Bradbury and Baeyens, TECHNISCHER BERICHT- NAGRA NTB</i>	Done	OK
				Selectivity coefficients	Literature		Done	OK
	<i>Not selected yet</i>	Isotopic exchange	Strontianite	<i>Not selected yet</i>	<i>Not selected yet</i>	Compare with model prediction for illite uptake	<i>Not appl.</i>	
	Batch			Ion exchange	Boom Clay		Exchange capacities	Isotopic dilution
	Migration	Ion exchange, isotopic exchange	Boom Clay	Retardation factor	Column migraton	Compare with model prediction for Boom Clay uptake in batch	Done	Needs to be validated
	Migration	Ion exchange, isotopic exchange	Boom Clay	Retardation factor	In-diffusion	Compare with model prediction for Boom Clay uptake in batch	In progress	
Europium (Eu)	Batch	Ion exchange/Surface complexation	Illite	Site capacities	Literature	<i>Bradbury and Baeyens, TECHNISCHER BERICHT- NAGRA NTB</i>	Done	OK
				Surface complexation constants	Literature		Done	OK
				Exchange capacities	Literature		Done	OK
				Selectivity coefficients	Literature		Done	OK
	Batch	Ion exchange/Surface complexation	Illite	Site capacities	Adsorption isotherm	Compare with model prediction for illite uptake	Done	OK, successful fit
				Surface complexation constants	Adsorption isotherm		Done	OK, successful fit
				Exchange capacities	Adsorption isotherm		Done	OK, successful fit
				Selectivity coefficients	Adsorption isotherm		Done	OK, successful fit
	Batch	Complexation	RBCW	Functional group capacities	Trancom-Clay	Compare with literature value for generic HS	In preparation	
				Deprotonation constants	Trancom-Clay		In preparation	
				Complexation constants	Solubility above Eu(OH) ₃		Done	OK, generic HS values used, further evaluation needed
				Site capacities	Adsorption isotherm		Done	Rule does not seem to work
	Batch	IE/SC and complexation	Illite + RBCW	Surface complexation constants	Adsorption isotherm	Validation of additivity rule, ultrafiltration for small/large HS	Done	Rule does not seem to work
				Exchange capacities	Adsorption isotherm		Done	Rule does not seem to work
				Selectivity coefficients	Adsorption isotherm		Done	Rule does not seem to work
				Functional group capacities	Adsorption isotherm		Done	Rule does not seem to work
				Deprotonation constants	Adsorption isotherm		Done	Rule does not seem to work
				Complexation constants	Adsorption isotherm		Done	Rule does not seem to work
Batch	"Complexation"	Kerogen	Functional group capacities	Literature	Use generic HS parameters, <i>Tipping, Aquatic Geochem., 4 (1998), 3-47</i>	Done	OK	
			Deprotonation constants	Literature		Done	OK	
			Complexation constants	Adsorption isotherm		Done	Not modelled yet	
			Migration	IE/SC and complexation		Boom Clay	Retardation factor	In-diffusion
Migration	IE/SC and complexation	Boom Clay	Retardation factor	Column migraton	Data with Am(III)	Done	Needs to be validated; problem: colloidal transport	
Radionuclide	Batch/Migration	Interaction mechanism	Geochemical phase	Parameters	Source of parameter value	Comment	Status	Modelling result
Americium (Am)	Batch	Ion exchange/Surface complexation	Illite	Site capacities	Literature	<i>Bradbury and Baeyens, TECHNISCHER BERICHT- NAGRA NTB</i>	Done	OK
				Surface complexation constants	Literature		Done	OK
				Exchange capacities	Literature		Done	OK
				Selectivity coefficients	Literature		Done	OK
	Batch	Ion exchange/Surface complexation	Illite	Site capacities	Adsorption edge	Compare with model prediction for illite uptake	In progress	
				Surface complexation constants	Adsorption edge		In progress	
				Exchange capacities	Adsorption edge		In progress	
				Selectivity coefficients	Adsorption edge		In progress	
	Batch	IE/SC and complexation	Illite + RBCW	Site capacities	Adsorption isotherm	Validation of additivity rule, ultrafiltration for small/large HS	Done	Modelling in progress
				Surface complexation constants	Adsorption isotherm		Done	Modelling in progress
				Exchange capacities	Adsorption isotherm		Done	Modelling in progress
				Selectivity coefficients	Adsorption isotherm		Done	Modelling in progress
Migration	IE/SC and complexation	Boom Clay	Functional group capacities	Adsorption isotherm	Data taken from Eu(III) experiments	Done	Modelling in progress	
			Deprotonation constants	Adsorption isotherm		Done	Modelling in progress	
			Complexation constants	Adsorption isotherm		Done	Modelling in progress	
			Retardation factor	Column migraton		Done	Needs to be validated; problem: colloidal transport	
Thorium (Th)	Batch	Ion exchange/Surface complexation	Illite, montmorillonite	Site capacities	Literature	<i>Bradbury and Baeyens, TECHNISCHER BERICHT- NAGRA NTB</i>	Done	OK
				Surface complexation constants	Literature		Done	OK
				Exchange capacities	Literature		Done	OK
				Selectivity coefficients	Literature		Done	OK
	Batch	Ion exchange/Surface complexation	Illite	Site capacities	Adsorption edge	Compare with model prediction for illite uptake	In progress	
				Surface complexation constants	Adsorption edge		In progress	
				Exchange capacities	Adsorption edge		In progress	
				Selectivity coefficients	Adsorption edge		In progress	
	Batch	Ion exchange/Surface complexation	Montmorillonite	Site capacities	Adsorption edge	Compare with model prediction for montmorillonite uptake	Done	Modelling in progress
				Surface complexation constants	Adsorption edge		Done	Modelling in progress
				Exchange capacities	Adsorption edge		Done	Modelling in progress
				Selectivity coefficients	Adsorption edge		Done	Modelling in progress
	Batch	Ion exchange/Surface complexation	Volclay MX-80	Site capacities	Adsorption isotherm	Compare with model prediction for montmorillonite uptake	Done	Modelling in progress
				Surface complexation constants	Adsorption isotherm		Done	Modelling in progress
				Exchange capacities	Adsorption isotherm		Done	Modelling in progress
				Selectivity coefficients	Adsorption isotherm		Done	Modelling in progress
	Batch	Complexation	RBCW	Functional group capacities	Trancom-Clay	Compare with literature value for generic HS	In preparation	
				Deprotonation constants	Trancom-Clay		In preparation	
Complexation constants				Solubility above ThO ₂	Done		OK, generic HS values used, further evaluation needed	
Site capacities				Adsorption isotherm	Done		Modelling in progress	
Batch	IE/SC and complexation	Boom Clay	Surface complexation constants	Adsorption isotherm	Ultrafiltration to distinguish between small and large HS	Done	Modelling in progress	
			Exchange capacities	Adsorption isotherm		Done	Modelling in progress	
			Selectivity coefficients	Adsorption isotherm		Done	Modelling in progress	
			Functional group capacities	Adsorption isotherm		Done	Modelling in progress	
			Deprotonation constants	Adsorption isotherm		Done	Modelling in progress	
			Complexation constants	Adsorption isotherm		Done	Modelling in progress	

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