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Experimental matrix on radionuclide migration and retention

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Liesbeth Van Laer, Norbert Maes

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SCK•CEN
Boeretang 200
BE-2400 Mol
Belgium

© SCK•CEN
Studiecentrum voor Kernenergie
Centre d'Etude de l'énergie Nucléaire
Boeretang 200
BE-2400 Mol
Belgium

Phone +32 14 33 21 11
Fax +32 14 31 50 21

http://www.sckcen.be

Contact:
Knowledge Centre
library@sckcen.be

Configuration Control

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Abstract

This report gives an update of the earlier defined experimental matrix on the radionuclide migration and retention (R-4792 – Bruggeman, 2009), which is supporting the report on 'Radionuclide migration and retention in the Boom Clay' (ER-0345 –Bruggeman & Maes, 2017) and 'Compilation of technical notes on less known elements' (ER-0323 – Salah et al., 2017).

An overview of the experiments performed is given per element. It is mentioned which types of experiments (i.e. migration, sorption, solubility and complexation experiments) were performed and which type of parameters were obtained from these experiments. The different elements are grouped as follows: non-retarded, anion exclusion, cation exchange, NOM and NOM related transport. This last group is subdivided in (i) transition metals and some actinides, (ii) trivalent lanthanides and actinides and (iii) tetra- and pentavalent actinides.

The overview contains the experiments performed until 2014. An update with the experiments from 2014 to 2017 will follow end of this year. A yearly update of this document is foreseen to keep track of the future experiments.

Keywords

Experimental matrix, migration, sorption, solubility, complexation

1 Introduction

This report serves to give an update of the earlier defined experimental matrix on the radionuclide migration and retention (R-4792 – Bruggeman, 2009), which is supporting the report on 'Radionuclide migration and retention in the Boom Clay' (ER-0345 –Bruggeman & Maes, 2017) and 'Compilation of technical notes on less known elements' (ER-0323 – Salah et al., 2017). The first version gives an overview of the experiments performed until 2014. The document will be updated with the experiments from 2014 to 2017 before the end of December 2017. A yearly update of this document is foreseen to keep track of the future experiments.

2 Experimental matrix

The updated experimental matrix is presented in Table 1. The table gives an overview of the experiments performed on migration, sorption, solubility and complexation with organic matter and it is also mentioned if parameters could be derived from these experiments or not.

RN solubility and complexation are sometimes investigated in a combined experiment (solubility in presence of organic matter).

RN sorption has been studied by performing sorption isotherms, sorption edges, single point measurements or sorption experiments at different S/L ratios.

Regarding RN migration experiments, there are two general types of experiments carried out: (1) pure diffusion tests, with a concentration gradient only; and (2) column migration tests with an additional hydraulic pressure gradient creating a small advective flow.

Two types of migration tests with advective flow are performed. In the *percolation experiments*, also referred as C4, the source is added between two clay plugs, while in the *pulse injection experiments*, also referred as D2, the tracer is injected as a pulse at the inlet filter at the start of the experiment.

The two variants of pure diffusion tests (*i.e*, without advection) are through-diffusion or in-diffusion experiments and back-to-back diffusion experiments. *Through-diffusion experiments*, or in-diffusion, consist of a clay core confined between two well-stirred water compartments. Initially, tracer is added to the inlet compartment and diffuses through the clay core towards the outlet compartment. back-to-back, are similar to percolation experiments but without advective flow.

Next to these type of experiments, also *electromigration experiments* have been performed for some elements. In these experiments the transport is accelerated by an electrical gradient.

At last, migration is/has been studied also *in-situ* by means of injection in a filter in a piezometer and follow-up of the activity in the surrounding piezometer filters.

Table 1 Experimenal matrix updated according to the radionuclide grouping proposed in Bruggeman & Maes (2016).

Group	Element	Equilibrium species	DATA			
			Migration	Sorption	Solubility	Complexation OM
Non-retarded	HTO	HTO	p	n.a.	n.a.	n.a.
Anion exclusion	I	I(-)	p	n.a.	n.a.	n.a.
	Cl	Cl(-)	-	n.a.	n.a.	n.a.
	Se(1)	HSe(-)	x	-	p	p
	Se(2)	SeO4(2-)	p	-	n.a.	p
	Mo	MoO4(2-)	-	-	-	-
	C	HCO3(-)	p	-	n.a.	n.a.
Cation exchange	Cs	Cs(+)	p	p	n.a.	-
	Rb	Rb(+)	-	-	n.a.	-
	Sr	Sr(2+)	p	p	n.a.	-
	Ca	Ca(2+)	p	-	n.a.	-
	Ra	Ra(2+)	p	-	n.a.	-
NOM	NOM	NOM small	p	p	-	n.a.
		NOM large				
NOM related transport	Tc	TcO(OH)2(aq)	x	p	p	p
	Ag	AgHS(aq)	-	-	-	-
	Be	BeO2(2-)	x	-	-	-
	Ni	Ni(CO3)2(2-)	-	-	-	-
	Nb	Nb(OH)6(-)	-	-	-	-
	Pd	Pd(OH)2(aq)	x	-	-	-
	Zr(IV)	Zr(OH)4(aq)	x	-	-	-
	Sn(IV)	Sn(OH)5(-)	-	-	-	-
	Am	Am(CO3)2(-)	x	p	p	p
	Cm	Cm(CO3)2(-)	x	-	-	-
	Pu	Pu(CO3)2(-)	x	-	p	-
	Sm	Sm(CO3)2(-)	-	-	-	-
	Ac	Ac(CO3)2(-)	-	-	-	-
	U	U(OH)4(aq)/ UO2(CO3)3(4-)	x	-	p	x
	Th	Th(OH)3(CO3)(-)	-	p	p	-
	Np	Np(OH)4(aq)	x	-	p	-
	Pa	Pa(OH)5(aq)	x	-	-	-

in blue	representative for a group
n.a.	not applicable
-	no data available
x	data available, but no parameters could be determined
p	data and parameters available

For each element, a more detailed overview of the experiments is given, i.e. the type and amount of experiments and the parameters obtained from these experiments, as listed below.

Symbols	**Description parameters**
C_{RN}	Experimentally determined concentration in equilibrium with solid phase
K_d	Solid/solution distribution coefficient at equilibrium
R_d	Solid/solution distribution coefficient measured in non-equilibrium conditions
R	Retardation factor of the RN
η	Porosity
D_{app}	Apparent diffusion coefficient
$R_{RN\text{-}DOM}$	Retardation factor or the RN bound to DOM
$K_{RN\text{-}DOM}$	Complexation constant between radionuclide RN and DOM
k_{diss}	Dissociation constant of the RN-DOM complex

2.1 Non-retarded

2.1.1 HTO

Migration of HTO has been investigated intensively over time with different types of experiments.

HTO	Clay	#exp	Parameters	
Migration				
Through-diffusion	illite	17	D_{app}	ηR
	BC	3	D_{app}	ηR
Pulse injection	BC (Mol)	79	D_{app}	ηR
	BC (Doel-2b)	10	D_{app}	ηR
	YC (Doel-1)	25	D_{app}	ηR
	YC (Kallo)	9	D_{app}	ηR
Percolation	BC (Essen)	7	D_{app}	ηR
Back-to-back	BC	1	D_{app}	
Electromigration	BC	9	D_{app}	
In-situ	BC	3	blind prediction	

Old experiments using consolidated clay pastes are not taken up in the overview.

The majority of experiments on Boom Clay were the pulse injection experiments performed on clay cores with different origin and under different conditions (orientation, solution type).

2.2 Anion exclusion

2.2.1 Iodine

Under Boom Clay conditions, iodide or I^-, is assumed as the predominant iodine species. Iodide is not solubility limited, will not react with DOM and can be considered as non-sorbing on the clay under Boom Clay conditions. Hence, only migration experiments were performed. Below the overview of migration experiments is given. Old experiments with consolidated pastes are not taken up in the table.

	Clay	#exp	Parameters	
Migration				
Through-diffusion	BC	3	D_{app}	ηR
Pulse injection	BC (Mol)	96	D_{app}	ηR
	BC (Doel-2b)	10	D_{app}	ηR
	YC (Doel-1)	25	D_{app}	ηR
	YC (Kallo)	9	D_{app}	ηR
Percolation	BC (Essen)	7	D_{app}	ηR
Electromigration	BC	5	D_{app}	
Back-to-back	BC	5	D_{app}	
In-situ	BC	2	blind prediction	

Recently, more I^- through-diffusion and percolations were performed. These experiments will be taken up in the next version of the document.

2.2.2 Chlorine

For Cl no in-house experiments have been performed.

2.2.3 Selenium

Two different oxidation states have been observed for Se (De Cannière et al., 2010). Se(-II), in form of HSe^-, represents the predicted stable species under the reducing conditions imposed by the Boom Clay. It is however established, that selenate species, i.e. SeO_4^{2-} (Se(VI)) might also persist in reducing porewaters due to redox disequilibrium and/or slow reduction kinetics (De Cannière et al., 2010). Both species, Se(-II) and Se(VI) are taken as representative equilibrium species of the anion exlusion group. In addition, experiments with Se(IV), as selenite SeO_3^{2-}, have been performed. Selenite is found to reduce to selenide under Boom Clay conditions.

Regarding Se(-II), no experiments have been performed to study HSe^- uptake on Boom Clay or its constituting phases. Due to the difficulty in preparing (pure) Se(-II) solutions, similar uptake experiments are also rarely found in scientific literature. There was also no sorption observed in batch tests for selenate (SeO_4^{2-}). Under Boom Clay conditions, amongst the selenium oxyanions, only selenite (SeO_3^{2-}) forms stable inner-sphere complexes at neutral and slightly alkaline pH and can sorb on oxide surfaces by surface complexation reaction. Sorption experiments with illite and Boom Clay have been performed, but they provide only R_d values instead of K_d, because the solid-liquid distribution is not only determined by sorption, but also by solubility (reduction with solubility limited release of HSe^-).

No solubility limits are reached for SeO_4^{2-} and SeO_3^{2-} in Boom Clay water, hence solublility experiments were not applicable. Solubility of Se(-II) has been determined.

For the three different redox states of Se, migration experiments have been performed.The migration of SeO_4^{2-} has been investigated with a series of electromigration experiments. In addition, several types of migration experiments have been performed with $^{35}SO_4^{2-}$, its chemical analogue. It was also attempted to determine the migration behaviour of selenide (HSe^-) and selenite (SeO_3^{2-}), but it was not possible to derive migration parameters from these percolation experiments.

Se(-II)	Clay	#exp	Parameters
Migration			
Percolation[a]	BC	2	no parameters
Sorption		/	
Solubility/Complexation			
Solubility		1	C_{RN}
Complexation OM		1	no complexation observed

[a] double-labelled exp with ^{14}C-OM

Se(VI)		Clay	#exp	Parameters	
Migration					
Percolation	SO_4^{2-}	BC	2	D_{app}	ηR
Pulse injection	SO_4^{2-}	BC	2	D_{app}	ηR
Pure diffusion	SO_4^{2-}	BC	1	D_{app}	
Electromigration		BC	7	D_{app}	
Sorption			/		
Solubility/Complexation					
Solubility				not solubility limited	
Complexation			1	no complexation observed	

Se(IV)	Clay	#exp	Parameters
Migration			
Percolation	BC	2	no parameters
Sorption			
Sorption isotherm+edge	illite	2	R_d
Sorption isotherm	BC	1	R_d
Solubility/Complexation			
Complexation		1	no parameter

2.2.4 Molybdenum

No in-house experiments have been performed for Mo.

2.2.5 Carbon

Only inorganic carbon is considered in the anion exclusion group. The organic carbon is described in 2.4. In Boom Clay porewater, C occurs as HCO_3^-. This equilibrium species is not solubility limited. From the percolation experiments it is clear that HCO_3^- is slightly retarded. However, no sorption experiments have been performed yet.

HCO_3^-	Clay	#exp	Parameters	
Migration				
Through-diffusion	BC	5	D_{app}	ηR
Pulse injection	BC	8	D_{app}	ηR
In-situ	BC	1	D_{app}	fixed*
Sorption		/		

* ηR was fixed in order to determine the D_{app}

Migration experiments have been performed in different forms, both in lab-scale setups and *in situ* on a larger scale in the HADES underground research facility (URF, Mol, Belgium).

2.3 Cation exchange

2.3.1 Cesium

Cesium is representative for the monovalent cation of the cation exchange group and is already intensively studied. There is no concentration limit expected for this element. Sorption has been measured in batch and compacted systems.

The migration behaviour of Cs in Boom Clay has been studied with different types of migration experiments. They provided reliable and reproducible values for D_{app}, but for ηR the uncertainty was quite high.

Cs	Clay	#exp	Parameters	
Migration				
In-diffusion	BC	5	D_{app}	ηR
Percolation	BC	2	D_{app}	ηR
In-situ percolation	BC	1	D_{app}	ηR
Electromigration	BC	5	D_{app}	
Sorption				
Isotherm	BC	1	K_d	
Compacted clay	BC	1	R_d	
Solubility/Complexation		/		

2.3.2 Rubidium

For Rb no in-house experiments have been performed.

2.3.3 Strontium

Strontium is the representative element for the divalent cations of the cation exchange group. According to speciation calculations, Sr is solubility limited under Boom Clay conditions (in equilibrium with strontianite). No solubility experiments have been performed however. Due to the concentration limit, sorption was only studied at one concentration (single point K_d) in batch instead of performing an isotherm. In addition, sorption was studied on a compacted clay.

Migration has been studied in five different ways. All these experiments provided reproducible and reliable values for D_{app} value, but they did not lead to trustworthy *R* values. However, by combining the data of the different experiments, a good approximation of the *R* value was possible.

Sr	Clay	#exp	Parameters	
Migration				
Through-diffusion	BC	2	D_{app}	ηR
	illite	11	D_{app}	ηR
Percolation	BC	1	D_{app}	ηR
In-situ percolation	BC	1	D_{app}	ηR
Pure diffusion	BC	2	D_{app}	
Electromigration	BC	6	D_{app}	
Sorption				
Sorption edge	illite	1	K_d	
Single point	BC	2	K_d	
Compacted clay	BC	1	K_d	
Solubility/Complexation		/		

2.3.4 Calcium

Until now, only migration experiments with Ca were performed. The type of migration experiments (pure diffusion and electromigration) allowed only to determine the D_{app} and not the ηR.

Ca	Clay	#exp	Parameters
Migration			
Pure diffusion	BC	2	D_{app}
Electromigration	BC	6	D_{app}
Sorption		/	
Solubility/Complexation		/	

2.3.5 Radium

Sorption and solubility data with Boom Clay are not yet available. The migration of Ra was studied by electromigration experiments, which provided a D_{app} value.

Ra	Clay	#exp	Parameters
Migration			
Electromigration	BC	5	D_{app}
Sorption		/	
Solubility/Complexation		/	

2.4 NOM

Natural organic matter has a widespread size distribution (Durce et al., xxx). It is reasonable to expect that the migration or retardation behaviour of small and large molecules is quite different. However, it is difficult to define arbitrarely what is 'small' and what is 'large'. In addition, a lot of experiments have been performed with the full fraction of the dissolved organic matter. Therefore, we provide an overview of all the NOM experiments, but always specifying the size fraction in an extra column.

The table contains the experiments performed before 2012. The recent work of Durce (ER-0xxx Durce et al., xxx) will be added in the next version.

NOM	Clay	Size fraction OM*	#exp	Parameters	
Migration					
Percolation	BC	F	27	D^i_{app}	ηR
Pulse injection	BC	F	13	D^i_{app}	ηR
	BC	<1kDa	6	D^i_{app}	ηR
	BC	>100 kDa	6	D^i_{app}	ηR
Pure diffusion	BC	F	1	D_{pore}	
Electromigration	BC	F	5	D_{app}	
Through-diffusion	BC	F	1	no parameters	
	BC	<1kDa	1	no parameters	
In-situ	BC	F	1	blind prediction	
Sorption					
at ≠ S/L	BC	F	1	K_d	
sequential	BC	F	1	K_d	
3 DOM pools	illite	F	1	K_d	
Solubility/Complexation			/		

* F=full fraction

The sorption of OM on Boom Clay was tested with two different experiments. In addition, the sorption of three different DOM pools on illite has been investigated.

The migration of NOM has been studied with all possible migration experiments, mostly percolation and pulse injection experiments. The majority of the experiments is performed with the full fraction of the organic matter, but there are also experiments which were performed with the isolated, small organic molecules (< 1 kDa) or the large molecules (> 100 kDa).

2.5 NOM related transport

A general remark for the elements of this group is that from the data of the 'normal' percolation experiments no transport parameters could be derived. Therefore, sequential migration experiments have been started up for most of the elements by coupling the clay core of the percolation experiment with a second clay core. These experiments, referred as sequential migration, have been modelled with a transport model developed by Maes et al. (2011) to interpret the organic matter linked transport in the Boom Clay. With this model it was possible to derive the dissociation constant of the organic matter, k_{diss}, the retardation factor R for the inorganic species, R_{RN}, and the retardation factor of the RN-DOM complex, R_{RN-DOM}. The apparent diffusion coefficient D_{app} can be calculated then with the fitted R value and a fixed value of the D_{pore}.

Transition metals and some actinides

2.5.1 Technetium

Solubility experiments were performed with and without organic matter and sorption was measured at different S/L ratios (which means also different OM concentrations).

Migration has been investigated with four percolation experiments, of whom two in-situ. Only the retardation factor R could be derived from these experiments. This R value corresponds however not to the inorganic Tc, but to the organic colloids.

One percolation experiment is coupled to a second clay core for the sequential migration experiment and is still running. Different parameters could be fitted with the transport model to describe the experimental data of this transport experiment.

Tc	Clay	#exp	Parameters			
Migration						
Percolation	BC	2	R			
Sequential migration	BC	1	D_{app}*	R	R_{RN-DOM}	k_{diss}
In-situ percolation	BC	2				
Sorption						
at ≠ [Tc], ≠ S/L	BC	1	K_d			
Solubility/Complexation						
Solubility		1	C_{RN}			
Complexation OM		1	K_{RN-DOM}			

* calculated with fixed value of D_{pore} and fitted value of R

2.5.2 Silver

For Ag no in-house experiments have been performed.

2.5.3 Beryllium

No in-house data on sorption or solubility are available for Boom Clay.

Two percolation experiments have been performed with Be. However, it was not possible to derive parameters from the obtained data. From the clay profile and the outlet concentrations, it appears that two separate processes are dominating Be transport in Boom Clay, one immobilizing and one mobilization mechanism.

The complexation with OM was not studied, but the rapid breakthrough observed in the percolation experiments is indicating DOM linked transport process.

Be	Clay	#exp	Parameters
Migration			
Percolation	BC	2	no parameters
Sorption		/	
Solubility/Complexation		/	

2.5.4 Nickel

Sorption of Ni has been investigated on illite (effect of organic and inorganic carbon) and on Boom Clay (in SBCW and RBCW).

Two percolation experiments have been started in 2013 and are still ongoing. No parameters could yet be derived.

Ni	Clay	#exp	Parameters
Migration			
Percolation	BC	2	no parameters
Sorption			
Sorption edge + isotherms	illite	7	K_d
Sorption isotherms	BC	2	K_d
Solubility/Complexation		/	

Zn is not a safety relevant radionuclide. However, as it is considered to be similar as Ni, an overview of the experiments with Zn is provided too. In-diffusion of Zn in illite has been studied at pH ≤7. At higher pH it is not possible to perform diffusion experiments due to the sorption of Zn on the material. This is also the reason why no parameters could be derived for Zn diffusion in Boom Clay (lack of good data).

Two percolation experiments were stopped after two years, but the clay profile was too narrow to analyze (only a few mm). Therefore, two new percolation experiments have been started in 2013 (still ongoing).

Zn	Clay	#exp	Parameters	
Migration				
Percolation	BC	4	no parameters	
In-diffusion	illite	6	D_{app}	ηR
	BC	1	no parameters	
Sorption				
Sorption edge + isotherms	illite	10	K_d	
Sorption isotherms	BC	2	K_d	
Solubility/Complexation		/		

2.5.5 Nyobium

For Nb no in-house experiments have been performed.

2.5.6 Palladium

No in-house data on sorption or solubility are available for Boom Clay.

Two percolation experiments have been started in 1994 with inactive Pd. However, the concentration in the outlet stays below the detection limit. Hence, no useful data are available.

Pd	Clay	#exp	Parameters
Migration			
Percolation	BC	2	no useful data
Sorption		/	
Solubility/Complexation		/	

2.5.7 Zirconium

No in-house data on sorption or solubility are available for Boom Clay.

Two percolation experiments have been performed with ^{95}Zr. Data of the clay profiles are available, but not could be used to derive robust parameters.

Zr	Clay	#exp	Parameters
Migration			
Percolation	BC	2	no parameters
Sorption		/	
Solubility/Complexation		/	

2.5.8 Tin

For Sn no in-house experiments have been performed.

Trivalent lanthanides and actinides

2.5.9 Americium

Since Am and Eu are chemical analogues, all the experiments for Eu are considered too. Next to the solubility experiments of Am with and without organic matter, a series of experiments with Eu were performed to derive the solublility limited concentration and the complexation constant with organic matter.

Sorption of Eu has been measured on illite, kerogen and Boom Clay. Also here the effect of organic matter on the sorption has been investigated.

Multiple migration experiments have been set up of whom the majority is still running, but no parameters could be derived from the experimental data yet.

Am	Clay	#exp	Parameters	
Migration				
Percolation	BC	9	no parameters	
Sequential migration	BC	1	no parameters	
Pulse injection	BC	3/4	no parameters	
In situ percolation	BC	1	no parameters	
Electromigration Eu	BC	1	no parameters	
Sorption				
Sorption edge Eu	illite	2	K_d	
Sorption isotherm Eu[a]	illite	3	K_d	
Sorption edge Eu	kerogen	1	K_d	
Sorption at ≠ S/L Eu/Am	BC	2	K_d	
Solubility/Complexation				
Solubility Am[a]		2	C_{RN}	
Solubility/Complexation Eu[b]		20	C_{RN}	$K_{RN\text{-}DOM}$

[a] with and without organic matter

[b] Solubility and complexation were measured in the same experiments

2.5.10 Curium

No sorption and solubility experiments have been performed yet.

To study the migration behaviour, two percolation experiments have been started long ago. One of them has been coupled to a second clay core (sequential migration experiment). Only from the latter, parameters could be derived.

Cm	Clay	#exp	Parameters			
Migration						
Percolation	BC	2	no parameters			
Sequential migration	BC	1	D_{app*}	R	R_{RN-DOM}	k_{diss}
Sorption		/				
Solubility/Complexation		/				

* calculated with fixed value of D_{pore} and fitted value of R

2.5.11 Plutonium

Solubility of Pu was measured in pure water, SBCW with organic matter and RBCW.

For sorption, no in-house data are available.

Regarding migration, five percolation experiments have been started, of whom two in combination with ^{14}C. One of the clay cores of the percolation experiments has been coupled to a second clay core (sequential migration experiment). One experiment has been stopped, but was not sliced. The other ones are still running. Parameters could only be derived from the sequential migration experiment.

Pu	Clay	#exp	Parameters			
Migration						
Percolation	BC	5	no parameters			
Sequential migration	BC	1	D_{app*}	R	R_{RN-DOM}	k_{diss}
Sorption		/				
Solubility/Complexation						
Solubility		3	C_{RN}			

* calculated with fixed value of D_{pore} and fitted value of R

2.5.12 Samarium

No in-house experiments were performed for Sm.

2.5.13 Actinium

No in-house experiments were performed for Ac.

Tetra- and pentavalent actinides

2.5.14 Uranium

In Boom Clay conditions, uranium occurs as U(IV) and U(VI). Solubility experiments with U(IV) in absence and presence of organic matter provides as well solubility limit as a complexation constant with organic matter. In addition, complexation experiments with both species were performed in order to determine a complexation constant.

For U(IV) sorption experiments were performed on Boom Clay constituting clay minerals, while the sorption of U(VI) has been investigated on Boom Clay itself.

Regarding migration, four percolation experiments have been set-up, of whom two double labelled with ^{14}C. One experiment is yet stopped and sliced, but the other three are still runnig.

U	Clay	#exp	Parameters	
Migration				
Percolation	BC	4	no parameters	
Electromigration U(VI)	BC	1	no parameters	
Sorption				
Single point U(IV)	≠ clays[a]	1	K_d	
Sorption at ≠ S/L U(VI)	BC	1	K_d	
Solubility/Complexation				
Solubility[b] U(IV)		3	C_{RN}	$K_{RN\text{-}DOM}$
Complexation U(IV)/U(VI)		5	$K_{RN\text{-}DOM}$	

[a] kaolinite, illite, illite-smectite layer, montmorillonite, chlorite

[b] with and without organic matter

2.5.15 Thorium

The solubility of Th(IV) in absence and presence of organic matter has been investigated twice at SCK•CEN. Sorption was studied as well on Boom Clay, as on the Boom Clay constituting minerals illite and montmorillonite.

With respect to transport unfortunately no in-house data are available, but we consider U(IV) to be a good analogue for Th(IV). Based on the experimental results and the U(IV)/Th(IV) analogy, transport of U through undisturbed BC is also considered to be organic matter (DOM) mediated.

Th	Clay	#exp	Parameters	
Migration		/		
Sorption				
Sorption edge	illite	1	K_d	
Sorption edge	montmorillonite	1	K_d	
Sorption isotherms	BC	2	K_d	
Solubility/Complexation				
Solubility		2	C_{RN}	K_{RN-DOM}

2.5.16 Neptunium

Two percolation experiments have been started and one of them is coupled to a second clay core (sequential migration experiment). From the latter it was possible to derive transport parameters.

Np	Clay	#exp	Parameters			
Migration						
Percolation	BC	2		no parameters		
Sequential migration	BC	1	D_{app*}	R	R_{RN-DOM}	k_{diss}
Sorption		/				
Solubility/Complexation						
Complexation OM		3	K_{RN-DOM}			

* calculated with fixed value of D_{pore} and fitted value of R

2.5.17 Proactinium

No in-house data on sorption or solubility are available for Boom Clay.

Two percolation experiments of whom one is coupled with a second clay core (sequential migration experiment) are running with ^{231}Pa in confined Boom Clay cores. The data of the sequential migration experiment allowed to derive parameters.

Pa	Clay	#exp	Parameters			
Migration						
Percolation	BC	2	no parameters			
Sequential migration	BC	1	D_{app*}	R	R_{RN-DOM}	k_{diss}
Sorption		/				
Solubility/Complexation		/				

* calculated with fixed value of D_{pore} and fitted value of R

References

Bruggeman, C. (2009). Preparation of topical reports as a basis for the Data Collection Forms. SCK•CEN-R-4792.

Bruggeman, C. & Maes, N. (2016). Radionuclide migration and retention in Boom Clay. SCK•CEN-ER-0345.

Durce, D. et al. (2017). Water-soluble organic matter in Boom Clay: 2012-2016 studies. SCK•CEN-ER-0382.

Maes, N., Bruggeman, C., Govaerts, J., Martens, E., Salah, S., and van Gompel, M. (2011). A consistent phenomenological model for natural organic matter linked migration of Tc(IV), Cm(III), Np(IV), Pu(III/IV) and Pa(V) in the Boom Clay. *Physics and Chemistry of the Earth*, **36**, 1590-1599.