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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., 2017b).

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 beryllium (Be), (ii) trivalent lanthanides and actinides and (iii) tetra- and pentavalent actinides.

Version 1 gave an overview of the experiments performed until 2014. This second version includes the experiments from 2014 to 2017. A yearly update of this document is foreseen to keep track of the future experiments.

Keywords

Experimental matrix, radionuclides, natural organic matter, migration, sorption, solubility, complexation

1 Introduction

This report serves to give an update of the earlier defined experimental matrix on the radionuclide (RN) 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).

Version 1 gave an overview of the experiments performed until 2014. This second version includes the experiments from 2014 to 2017. A yearly update of this document is foreseen to keep track of the future experiments. Newly added data compared to previous versions are listed in the tables in italic.

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. Further, it is mentioned if parameters could be derived from these experiments or not.

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 beryllium (Be), (ii) trivalent lanthanides and actinides and (iii) tetra- and pentavalent actinides.

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 to as C4, the source is added between two clay plugs, while in the *pulse injection experiments*, also referred to 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 (2017).

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 NOM large	p	p	n.a.	n.a.
NOM related transport	Tc	TcO(OH)2(aq)	p	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(-)	p	p	p	p
	Cm	Cm(CO3)2(-)	p	-	-	-
	Pu	Pu(CO3)2(-)	p	-	p	-
	Sm	Sm(CO3)2(-)	-	-	-	-
	Ac	Ac(CO3)2(-)	-	-	-	-
	U	U(OH)4(aq)/ UO2(CO3)3(4-)	p	-	p	x
	Th	Th(OH)3(CO3)(-)	-	p	p	-
	Np	Np(OH)4(aq)	p	-	p	-
	Pa	Pa(OH)5(aq)	p	-	-	-

in blue	representative of the 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_a	Apparent diffusion coefficient
D_p	Pore 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

Abbreviations	Full names
BC	Boom Clay
YC	Ypresian clays
ES	Eigenbilzen sands

When BC is mentioned, it refers to Boom Clay of the Mol site. In case it is referring to Boom Clay from other locations, it is explicitly mentioned.

2.1 Non-retarded

2.1.1 HTO

Migration of HTO has been investigated intensively over time with different types of experiments, different clay samples, different orientation, different lengths, different chemical conditions (e.g. ionic strength),…

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).

Since 2014 a lot of new experiments were performed. Jacops et al. (2017a&b) performed through-diffusion experiments in five Boom Clay samples and four Eigenbilzen Sand samples. Durce (2017) performed nine through-diffusion experiments and 13 pulse injection experiments during the last years. Van Laer performed six through-diffusions on Boom Clay (Durce & Van Laer, 2017; Van Laer et al. 2015) and two on Ypresian clay samples (Van Laer et al., 2015).

HTO	Clay	#exp	Parameters	
Migration				
Through-diffusion	illite	*20*	D_a	ηR
	BC	*23*	D_a	ηR
	ES*	*4*	D_a	ηR
Pulse injection	BC (Mol)	*92*	D_a	ηR
	BC (Doel-2b)	10	D_a	ηR
	YC (Doel-1)	25	D_a	ηR
	YC (Kallo)	*11*	D_a	ηR
Percolation	BC (Essen)	7	D_a	ηR
Back-to-back	BC	2	D_a	
Electromigration	BC	9	D_a	
In-situ	BC	3	blind prediction	

* ES=Eigenbilzen Sands

References:

Aertsens (2013), Aertsens et al. (2005a, 2005b, 2009, 2010b, 2013, 2015), Altmann et al. (2015), Bruggeman et al. (2013, 2017c), De Cannière et al. (1996), Maes et al. (1999, 2004), Moors (2005), Put et al. (1991), Van Laer et al. (2018), Wang et al. (2004)

Update 2017: Jacops et al. (2017a & b), Durce & Van Laer (2017), Durce (2017), Van Laer et al. (2015)

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.

Iodine	Clay	#exp	Parameters	
Migration				
Through-diffusion	BC	18	D_a	ηR
Pulse injection	BC (Mol)	*105*	D_a	ηR
	BC (Doel-2b)	10	D_a	ηR
	YC (Doel-1)	25	D_a	ηR
	YC (Kallo)	*11*	D_a	ηR
Percolation	BC (Essen)	7	D_a	ηR
Electromigration	BC	5	D_a	
Back-to-back	BC	5	D_a	
In-situ	BC	2	blind prediction	

Since 2014 a lot of new experiments were performed. Durce performed nine through-diffusion experiments and nine pulse injection experiments during the last years (Durce, 2017). Van Laer performed six through-diffusions on Boom Clay (Durce & Van Laer, 2017; Van Laer et al. 2015) and two on Ypresian clay samples (Van Laer et al., 2015).

References:

Aertsens (2013), Aertsens et al. (2005a, 2005b, 2009, 2010, 2013, 2015), Bruggeman et al. (2010b, 2017d), De Cannière et al. (1996), Maes et al. (1999), Moors et al. (2005), Van Laer et al. (2018), Wang et al. (2004)

Update 2017: Durce & Van Laer (2017), Durce (2017), Van Laer et al. (2015)

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 on 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
Solubility/Complexation			
Solubility		1	C_{RN}
Complexation OM		1	no complexation observed

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

References

Migration: Dierckx et al. (2000)

Solubility/Complexation: De Cannière et al. (2010)

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

References

Migration: De Cannière et al. (2010), Beauwens et al. (2005)

Complexation: De Cannière et al. (2010)

Se(IV)	Clay	#exp	Parameters
Migration			
Percolation	BC	2	no parameters
Sorption			
Sorption isotherm+edge	illite	8	R_d
Sorption isotherm	BC	4	R_d
Solubility/Complexation			
Complexation		1	no parameter

References

Migration: De Cannière et al. (2010)

Sorption: Bruggeman (2006), De Cannière et al. (2010)

Complexation: De Cannière et al. (2010)

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.

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). The in-situ experiments are still ongoing (TRIBICARB).

HCO_3^-	Clay	#exp	Parameters	
Migration				
Through-diffusion	BC	5	D_a	ηR
Pulse injection	BC	8	D_a	ηR
In-situ	BC	2	D_a	fixed*

* ηR was fixed in order to determine the D_a

References

Aertsens (2013), Aertsens et al. (2008, 2010a, 2013), Salah et al. (2017b), Van Laer et al. (2018), Weetjens et al. (2011)

2.3 Cation exchange

2.3.1 Cesium

Cesium is representative for the monovalent cations featuring in the "cation exchange group" and is already intensively studied. There is no concentration limit expected for this element. Sorption on Boom Clay 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_a, but for ηR the uncertainty was quite high.

Before 2014, Cs sorption was studied once in batch (isotherm) and once on compacted Boom Clay sample. After 2014, three more sorption isotherms were performed on three different Boom Clay samples of well known mineralogical composition (Maes et al., 2015).

Cs	Clay	#exp	Parameters	
Migration				
In-diffusion	BC	5	D_a	ηR
Percolation	BC	2	D_a	ηR
In-situ percolation	BC	1	D_a	ηR
Electromigration	BC	5	D_a	
Sorption				
Isotherm	BC	6	K_d	
Compacted clay	BC	1	R_d	

References

Maes et al. (2008, 2009, 2011a, 2017a), Van Laer et al. (2016)

Update 2017: Maes et al. (2015)

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 on Boom Clay (batch and compacted) was only studied at one concentration (single point K_d) in batch instead of performing an isotherm. In addition, a sorption edge on illite was studied.

Migration in Boom Clay has been studied in five different ways. All these experiments provided reproducible and reliable values for D_a 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	*4*	D_a	ηR
	illite	*14*	D_a	ηR
	YC	*2*	D_a	ηR
Percolation	BC	1	D_a	ηR
In-situ percolation	BC	1	D_a	ηR
Pure diffusion	BC	2	D_a	
Electromigration	BC	6	D_a	
Sorption				
Sorption edge	illite	1	K_d	
Single point	BC	2	K_d	
Compacted clay	BC	1	K_d	

After 2014, Van Laer et al. (2015) performed through-diffusion experiments on two Boom Clay samples and two Ypresian clay samples (Kallo). Further, three more through-diffusion experiments were performed with Sr in compacted illite, but these results are not yet published (Van Laer, unpublished).

References

Altmann et al. (2015), Beaufays et al. (1994), Maes et al. (1999, 2009, 2012, 2017b), Montoya et al. (2018), Van Laer et al. (2016)

Update 2017: Van Laer et al. (2015)

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 D_a and not ηR.

Ca	Clay	#exp	Parameters
Migration			
Pure diffusion	BC	2	D_a
Electromigration	BC	6	D_a

References

Ecoclay-II (2005), Salah et al. (2017b)

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_a value.

Ra	Clay	#exp	Parameters
Migration			
Electromigration	BC	5	D_a

References

Maes et al. (2001), Salah et al. (2017)

2.4 NOM

Natural organic matter has a widespread size distribution (Durce et al., 2017). 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"[1] 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.

Version 1 of this report the table contained the experiments performed before 2012. The recent work of Durce (ER-0382 Durce et al., 2017) is included in this version 2 (status 2017).

NOM	Clay	Size fraction OM*	#exp	Parameters		
Migration						
Percolation	BC	F	21	D^i_a	ηR	
	BC	<1kDa	1			
	BC	F	*6*	D_a	ηR	ηk_f[a]
Pulse injection	BC	F	13	D^i_a	ηR	
	BC	<1kDa	6	D^i_a	ηR	
	BC	>100 kDa	6	D^i_a	ηR	
Electromigration	BC	F	5	D^i_a		
Pure diffusion	BC	F	1	D_p		
Back-to-back diffusion	BC			D_p		
Through-diffusion	BC	F	*2*	D_a	ηR	
In-situ 14C	BC	F	2	blind prediction		
Sorption						
Experiments with 14-C						
at ≠ S/L	BC	F	1	K_d		
sequential	BC	F	1	K_d		
single point	BC	F	1	K_d		
sorption/desorption	BC	F	1	K_d		
Experiments with NOM						
3 DOM pools	illite	F	1	K_d		
Isotherm + kinetics	BC	<30kDa[b]	1	K_d		
Isotherm + kinetics	BC	>100kDa[c]	1	K_d		

* F=full fraction

[a] fitration rate (Durce et al., 2017)

[b] pore water DOM filtered at 30 kDa = "mobile DOM"

[c] extracted water-soluble OM filtered at 100 kDa

[1] OM concentrated from EG/BS piezometer and subsequently radiolabelled with ^{14}C, no size fractionation performed, hence considered as representative for wide size distribution of OM present in Boom Clay.

The migration of NOM has been studied with all possible migration experiments, mostly percolation and pulse injection experiments (Put et al., 1992, 1998; Dierckx et al., 2000, Maes et al., 2004, 2006). The majority of the experiments is performed with the full fraction of the organic matter, but there are also experiments which were performed with a specific size fraction (<1 kDa, < 30 kDa, > 100kDa). These experiments include as well single as double-label experiments. In the double-label experiments the migration behaviour of ^{14}C is studied together with a DOM-facilitated RN (Am, Pu, U: described in 2.5).

During EC projects TRANCOM-Clay (Dierckx et al., 2000) and TRANCOM-II (Maes et al., 2004) a lot of migration experiments have been launched. A part of them are still ongoing, namely three single pulse injection experiments and 11 double-labelled percolation experiments.

In 1997/1998, two in-situ experiments were launched in HADES, namely TROM R41V and H, which are still being monitored (Martens et al., 2010).

Recently, Durce et al. (2017) performed six percolation experiments with ^{14}C at three different hydraulic gradients. Keeping the conceptual model for the RN-colloidal facilitated transport described by Maes et al. (2011b) in mind, the MW/size contribution was integrated by decomposing the MW distribution of the inlet and outlet solutions in discrete points and solving the advection-dispersion equation for each point. An extra parameter, ηk_f or filtration rate, was introduced. In addition, two through-diffusion experiments were performed.

The sorption of OM on Boom Clay was tested in the past during TRANCOM-Clay (Dierckx et al., 2000) and TRANCOM-II (Maes et al., 2004) with batch experiments at different solid/liquid distribution with radio-labelled DOM (^{14}C). Durce et al. (2017) performed batch sorption experiments on two Boom Clay samples. On one of them also desorption was investigated. In addition, the sorption of natural DOM was investigated. Two different DOM pools were used: 1) pore water DOM filtered at 30 kDa which accounts for the small DOM that is assumed to be the mobile DOM, and 2) extracted water-soluble OM filtered at 100 kDa, which accounts for the large and less mobile NOM.

At last, the sorption of three different DOM pools on illite has been investigated too (Bruggeman et al., 2010c).

References

Migration: Put (1992, 1998), Dierckx et al. (2000), Durce et al. (2017), Maes et al. (2004, 2006), Martens et al. (2010), Van Laer et al. (2018)

Sorption: Bruggeman et al. (2010c), Dierckx et al. (2000), Durce et al. (2017), Maes et al. (2004), Van Laer et al. (2016)

Update 2017: Durce et al. (2017), Van Laer et al. (2018)

2.5 NOM related transport

A general remark for the elements of this group is that in general no transport parameters could be derived from the data of the 'normal' percolation experiments. 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 to as "sequential migration experiments", have been modelled with a transport model described by Maes et al. (2011b) to interpret the organic matter facilitated RN 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, $R_{RN\text{-}DOM}$. The apparent diffusion coefficient D_a can be calculated then with the fitted R value and a fixed value of the D_p.

This group of DOM facilitated RN transport is subdivided in (i) transition metals and beryllium (Be), (ii) trivalent lanthanides and actinides and (iii) tetra- and pentavalent actinides.

Transition metals and Be

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).

The sorption of Tc(IV) was studied on Boom Clay by Henrion et al. (1985).

Migration has been investigated with four percolation experiments, of which two in-situ (one for disturbed conditions). 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 lab 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 (Maes et al., 2011b).

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

* calculated with fixed value of D_p and fitted value of R

References

Migration: Bruggeman (2010a, 2017b), Maes et al. (2011b), Van Laer et al. (2018)

Sorption: Bruggeman (2010a, 2017b), Henrion (1985), Van Laer et al. (2016)

Solubility/complexation: Bruggeman (2010a, 2017b)

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 mobilizing mechanism.

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

Be	Clay	#exp	Parameters
Migration			
Percolation	BC	2	no parameters

References

Salah et al. (2017b), Van Laer et al. (2018)

2.5.4 Nickel

Sorption of Ni has been investigated on two types of illite. On the Silver Hill Illite the effect of organic and inorganic carbon was studied. On Illite du Puy, the effect of pH was investigated (sorption edge). Further, sorption isotherms on Boom Clay were performed 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

References

Van Laer et al. (2016, 2018)

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 Zn tends to sorb on the materials used in a diffusion set up, hence attempts were not successfull. 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_a ηR
	BC	1	no parameters
Sorption			
Sorption edge + isotherms	illite	10	K_d
Sorption isotherms	BC	2	K_d

References

Altmann et al. (2015), Montoya et al. (2018), Van Laer et al. (2016, 2018)

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

References

Van Laer et al. (2018)

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 could not be used to derive robust parameters.

Zr	Clay	#exp	Parameters
Migration			
Percolation	BC	2	no parameters

References

Van Laer et al. (2018)

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 conducted with 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 for Am. Two single-labelled percolation experiments were launched in 1996. One experiment was stopped after ±3 years and the other was coupled to a second clay core in 2009 (SCC) and is still running. The other percolation experiments (7) are all double-labelled experiments (with ^{14}C) which are still running. Five of these percolation experiments were modelled with the organic matter facilitated RN transport model described by Maes et al. (2011b) and provided transport parameters (Govaersts, unpublished data). The other migration experiments (sequential migration, pulse injection, in-situ percolation and electromigration) did not provide parameters.

Am	Clay	#exp	Parameters			
Migration						
Percolation	BC	9	D_{a*}	R_{RN}	R_{RN-DOM}	k_{diss}
Sequential migration[a]	BC	4	no parameters			
Pulse injection	BC	3	no parameters			
In situ percolation	BC	2	no parameters			
Electromigration Eu	BC	1	no parameters			
Sorption						
Sorption edge Am/Eu	illite	5	K_d			
Sorption isotherm Eu	illite	3	K_d			
Sorption edge Eu	BC kerogen	2	K_d			
Sorption isotherm	BC	2	K_d			
Sorption at ≠ S/L Eu/Am	BC	2	K_d			
Solubility/Complexation						
Solubility Am/Eu		2	C_{RN}			
Solubility/Complexation Am/Eu[a]		20	C_{RN}	K_{RN-DOM}		

* calculated with fixed value of D_p and fitted value of R

[a] 3 exp are coupled to pulse injection exp, 1 exp with percolation exp

References

Migration: Aertsens (2013), Bruggeman (2012, 2017a), Dierckx et al. (2000), Maes et al. (2004, 2009), Van Laer et al. (2018), Govaerts (unpublished results).

Sorption: Bruggeman (2010c, 2012, 2017a), Henrion et al. (1985), Maes et al. (2009), Salah et al. (2009), Van Laer et al. (2016), Wang et al. (1998)

Solubility/Complexation: Bruggeman (2012, 2017a), Liu et al. (2008), Maes et al. (2004)

2.5.10 Curium

No sorption and solubility experiments have been performed yet.

To study the migration behaviour, two long-term 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 experiment, parameters could be derived.

Cm	Clay	#exp	Parameters			
Migration						
Percolation	BC	2	no parameters			
Sequential migration	BC	1	D_a*	R_{RN}	$R_{RN\text{-}DOM}$	k_{diss}

* calculated with fixed value of D_p and fitted value of R

References

Maes et al. (2011b); Salah et al. (2017b); Van Laer et al. (2018)

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 which two in combination with ^{14}C (double-labelled experiments). One single-labelled experiment has been stopped, but was not sliced. Another percolation experiment has been coupled to a second clay core (sequential migration experiment) and is still running, just as the third single-labelled and the two double-labelled experiments. 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_{a*}	R	$R_{RN\text{-}DOM}$	k_{diss}
Solubility/Complexation						
Solubility		3	C_{RN}			

* calculated with fixed value of D_p and fitted value of R

References

Migration: Maes et al. (2004, 2011b); Salah et al. (2017b); Van Laer et al. (2018)

Solubility/Complexation: Maes et al. (2004)

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 which two double labelled with ^{14}C. One (single label ^{233}U) experiment is stopped and sliced but from the pattern, no parameters could be derived. The second experiment is coupled to a second clay core for a "sequential migration" experiment. From the latter experiment transport parameters were derived (Salah et al., 2017a; Van Laer et al., 2018).

U	Clay	#exp	Parameters			
Migration						
Percolation	BC	4	D_p	R		
Sequential migration	BC	1	D_a*	R	R_{RN-DOM}	k_{diss}
Electromigration U(VI)	BC	1	D_a			
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-DOM}		
Complexation U(IV)/U(VI)		5	K_{RN-DOM}			

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

[b] with and without organic matter

* calculated with fixed value of D_p and fitted value of R

References

Migration: Maes et al. (2002, 2004); Salah et al. (2015, 2017a); Van Laer et al. (2018)

Sorption: Dierckx et al. (2000), Delécaut (2004), Van Laer et al. (2016)

Solubility/Complexation: Dierckx et al. (2000), Delécaut (2004)

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 (Salah et al., 2007), as on the Boom Clay constituting minerals illite (unpublished data) and montmorillonite (Salah et al., 2010).

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	
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}	$K_{RN\text{-}DOM}$

References

Sorption: Salah et al. (2007, 2010, 2017b); Van Laer et al. (2016)

Solubility: Delécaut (2004); Maes et al. (2011b), Salah et al. (2013), Salah et al. (2017b)

2.5.16 Neptunium

Two long-term percolation experiments are ongoing for long time. 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_{a*}	R	$R_{RN\text{-}DOM}$	k_{diss}
Solubility/Complexation						
Complexation OM		3	$K_{RN\text{-}DOM}$			

* calculated with fixed value of D_p and fitted value of R

References

Migration: Maes et al. (2011b), Salah et al. (2017b), Van Laer et al. (2018)

Complexation: Pirlet et al. (1998), Pirlet & Van Iseghem (2003), Salah et al. (2017b)

2.5.17 Protactinium

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

Two percolation experiments of which 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_{a*}	R	$R_{RN\text{-}DOM}$	k_{diss}

* calculated with fixed value of D_p and fitted value of R

References

Maes et al. (2011b); Salah et al. (2017b); Van Laer et al. (2018)

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