Comparison between *in situ* and laboratory diffusion studies of HTO and halides in Opalinus Clay from the Mont Terri

By E. Tevissen^{1,2,*}, J. M. Soler³, P. Montarnal⁴, A. Gautschi⁵ and L. R. Van Loon⁶

¹ CEA Saclay, bat 450 DPC/SECR/L3MR, F-91191 Gif-sur-Yvette, France

² Andra DS/TR, 1–7 rue J. Monnet, F-92298 Châtenay-Malabry Cedex, France

³ Inst. Ciències de la Terra “Jaume Almera” (CSIC), Lluís Solé i Sabarís s/n, E-08028 Barcelona, Spain

⁴ CEA Saclay, DM2S/MTMS, F-91191 Gif-sur-Yvette, France

⁵ NAGRA, Hardstrasse 73, CH-5430 Wetztingen, Switzerland

⁶ Paul Scherrer Institut, CH-5232 Villigen PSI, Switzerland

(Received August 11, 2003; accepted March 22, 2004)

Diffusion / Tritium / Iodide / Claystone / Inverse modelling

Summary. A long-term single-borehole diffusion experiment (DI) using tritiated water (HTO) and stable iodide ($^{127}\text{I}^-$) was carried out in the Opalinus Clay of the Mont Terri Underground Rock Laboratory (URL). Diffusion coefficients D_L and accessible porosity for HTO, $^{36}\text{Cl}^-$ and $^{125}\text{I}^-$ were also measured on centimetric Opalinus clay samples using the through diffusion technique. The evolution of tritium and iodide concentration in the injection system over time and *in situ* profiles were interpreted with a 3-D numerical simulation. A detailed analysis of the results pointed out the effect of a disturbed zone around the borehole with higher diffusion coefficients. The best estimate values for HTO and iodide in the undisturbed rock are $D_L = 5 \times 10^{-11} \text{ m}^2/\text{s}$ and $D_L = 1.5 \times 10^{-11} \text{ m}^2/\text{s}$ respectively. For the laboratory tests, D_L values for HTO are in the range of $5 \times 10^{-11} \text{ m}^2/\text{s}$ to $8.5 \times 10^{-11} \text{ m}^2/\text{s}$. For $^{125}\text{I}^-$ and $^{36}\text{Cl}^-$ the measured values are $D_L = 1.4 \times 10^{-11}$ and $D_L = 1.6 \times 10^{-11} \text{ m}^2/\text{s}$ respectively.

All HTO results obtained with a through diffusion technique are within the same range as those obtained in the *in situ* tests. The D_L values obtained in diffusion cells with $^{125}\text{I}^-$ and $^{36}\text{Cl}^-$ and the value drawn from the interpretation of stable $^{127}\text{I}^-$ concentration profiles from the *in situ* tests are very close. In fact, some significant uncertainties could be identified (*i.e.* a likely chemical retention of iodide on argillites, effect of the disturbed zone).

1. Introduction

Various countries are considering consolidated clay or claystone formations as a suitable host rock for the deep disposal of radioactive waste. The Mont Terri tunnel, located in the Swiss Jura provides an opportunity to collect relevant *in situ* data for safety assessment by offering access to such a formation, *i.e.* Opalinus clay. The multidisciplinary work programme of the International Mont Terri Project is devoted to the development of investigation techniques, the characterisation of the Opalinus clay and to the assessment of the repository-induced changes in the medium [1].

Diffusion is the main transport mechanism for released radionuclides in argillaceous formations. Diffusion parameters may be measured on small scale rock samples in the laboratory. The values however need to be confirmed by proper field experiments. For that reason, the DI program included three types of activities: laboratory tests on Opalinus-clay samples (through-diffusion tests), an *in situ* experiment carried out in a borehole in the Mont-Terri URL, and modelling [2].

2. Long term *in situ* diffusion test: DI

2.1 Experimental procedure

The Opalinus Clay was formed around 180 millions years ago in the Jurassic Sea by deposition of fine mud particles. Between 40 and 80% of the Opalinus Clay is made up of clay minerals. Around 10% of those minerals are capable of swelling and may increase in volume in the case of water influx. Other components include quartz (sand and silt), calcite, siderite, pyrite, feldspar and organic carbon. Several facies have been defined inside the Opalinus Clay formation: silty-shaly facies, sandy facies and sandy carbonate rich facies. Table 1 gives the composition of Opalinus Clay formation Water [3].

Table 1. Composition of Opalinus Clay formation water as taken from [3].

Water composition	
$\log p_{\text{CO}_2}$	−2.2
pH	7.24
Ionic strength (eq/L)	0.23
Dissolved species (mol/L)	
CO_3	2.70×10^{-3}
Na	1.69×10^{-1}
Ca	1.05×10^{-2}
Mg	7.48×10^{-3}
K	5.65×10^{-3}
Cl	1.60×10^{-1}
SO_4	2.40×10^{-2}

* Author for correspondence (E-mail: etienne.tevissen@cea.fr).

Only a brief description of the *in situ* diffusion experiment is given here. More detailed information has been published previously for tritiated water [2]. The *in situ* experiment was carried out in a 76-mm-diameter, 8.3-m-deep borehole. Because the test focused on diffusion processes, the injection borehole was maintained at the local pore-water pressure in order to avoid any advective flow. After one year of hydraulic stabilisation, the tracers (HTO and stable iodide) were added to the borehole liquid. This water was synthetic pore water with NaCl as the major constituent. A continuous circulation between the borehole equipment and a water tank located outside the borehole maintained the traced fluid homogeneous. It is important to notice that there are four readjustments of the tritium concentration in the injection chamber after the initial spike. The iodide concentration is not readjusted. After one year of diffusion, the area where the tracers were expected to have diffused was removed by overcoring the diffusion borehole. The cores were cut into pieces to collect undisturbed centimeter-sized rock samples and to obtain several tracer-concentration profiles with different orientations with respect to the axis of the borehole and the bedding anisotropy. Chemical-dilution analysis was used to determine tracer concentrations in the rock samples. Tritium was quantified by liquid-scintillation analysis. Iodide was analysed using an ion-chromatography device linked to an electrical conductivity detector. The pore-water content in the rock was derived from weight loss measurements.

2.2 Numerical modelling of the DI test

The modelled domain is three-dimensional: a claystone cylinder around the injection chamber. The size of the domain is adjusted in order to ignore boundary-condition effects on the outer surface of the domain: tracer concentrations on the outer surface must be negligible during simulation [4]. Diameter variations of the borehole equipment were ignored.

Tracer transport in the clay domain is described by the following diffusion equation [5]:

$$\omega \frac{\partial C}{\partial t} - \nabla(\bar{D} \nabla C) + \omega \lambda C = 0, \quad (1)$$

where C represents the concentration, ω the porosity accessible to diffusion, $\bar{D}$ the effective orthotropic diffusion tensor, λ and the radioactive decay coefficient. The expression of the effective orthotropic diffusion tensor $\bar{D}$ is the following:

$$\bar{D} = \begin{pmatrix} \cos^2 \nu D_L + \sin^2 \nu D_T & 0 & \sin \nu \cos \nu (D_L - D_T) \\ 0 & D_L & 0 \\ \sin \nu \cos \nu (D_L - D_T) & 0 & \cos^2 \nu D_T + \sin^2 \nu D_L \end{pmatrix}, \quad (2)$$

where D_L is the longitudinal effective diffusion coefficient (parallel to stratification) D_T the transverse effective diffusion coefficient (perpendicular to stratification), and ν the angle between the horizontal axis and the bedding plane (34°).

The boundary conditions are as follows:

- On the surface around the injection chamber screen: a concentration equal to the one in the chamber.
- On the other part of the surface around the borehole equipment, on the external surface of the domain and on the vertical plane of symmetry: zero flux.

The concentration in the injection chamber is a function of time t that depends on the outgoing flux. The transport equation is solved with CASTEM 2000 [6]. The Fick Eq. (1) is discretised within a Mixed Hybrid Finite Elements (MHFE) formulation and a fully implicit time discretisation [7, 8]. The tracer concentration in the injection chamber is computed with an explicit method as described in [2]. The 19 300 elements mesh is refined in the area of the chamber. A 1-day time step is adopted. The parameters identified are the diffusion coefficients (D_L , D_T) and the porosity accessible to diffusion (ω).

2.3 Results

HTO

The experimental data for the tritium-concentration evolution in the injection chamber over time is considered by Palut *et al.* [2]. These authors conclude that the porosity accessible to diffusion seems to over-parameterise the problem and indicate a weak monitoring sensitivity to the transverse-diffusion coefficient. Moreover, Palut *et al.* [2] plotted the tritium content in the injection chamber with $(D_L, D_T, \omega) = (18 \times 10^{-11} \text{ m}^2/\text{s}, 1.2 \times 10^{-11} \text{ m}^2/\text{s}, 12\%)$ and compared it with experimental data. They observed that the last group of points (corresponding to the fifth readjustment of tritium concentration) is well fitted, compared to the first groups of points. They explained it by the fact that during the short times the tracer plume is located around the injection chamber in a higher diffusion coefficient zone, namely a disturbed zone, and for longer time periods, when the plume has diffused further, the apparent diffusion coefficient decreases and is closer to that of the undisturbed rock. Palut *et al.* [2] suggest that the site-diffusion coefficient is better identified by taking into account only the long-term experimental data, those measured after 113 days (after the last readjustment). In that case, the error minimisation (the one taking into account only the last group of points) gives a value of $16 \times 10^{-11} \text{ m}^2/\text{s}$ for D_L and therefore of $2.28 \times 10^{-11} \text{ m}^2/\text{s}$ for D_T as the identified diffusion coefficients.

The entire set of computed tritium-content profiles are compared with the experimental profiles in [2]. In Fig. 1, three computed tritium concentration are plotted with $(D_L, D_T, \omega) = (10 \times 10^{-11} \text{ m}^2/\text{s}, 1.43 \times 10^{-11} \text{ m}^2/\text{s}, 15\%)$. It is worthwhile noticing the good fit with the experimental data. More precisely, it is possible to observe that the main position of the profiles are well described, but that calculated curves systematically overestimate the tritium concentrations for the longest distances from the injection borehole. That may be interpreted again by the effect of a disturbed zone. As a matter of fact, Palut *et al.* [2] noticed that a lower value ($D_L = 5 \times 10^{-11} \text{ m}^2/\text{s}$) seems to fit better the measured concentrations for the longest distances. That value is likely to be the best estimate for the undisturbed rock.

Fig. 1. Tritium content in 3 profiles parallel to bedding plane vs. space, comparison between experiment and numerical results (D_L , D_T , ω) = ($10 \times 10^{-11} \text{ m}^2/\text{s}$, $1.43 \times 10^{-11} \text{ m}^2/\text{s}$, 15%) from [2].

Iodide

In order to describe the decrease of iodide concentration in the injection system, Montarnal *et al.* [4] first considered an initial iodide concentration of 1.9 g/L, corresponding to the iodide concentration in the water of the injection system, measured immediately after injection. The numerical process selected to minimize the quadratic error on concentration data over time generates very high diffusion coefficient values for iodide: $D_L = 3 \times 10^{-10} \text{ m}^2/\text{s}$ for a 10% porosity, for example. The D_L/D_T ratio is arbitrarily set at 4.4, based on the diffusion-coefficient data obtained in the laboratory, with the D_T parameter influencing only slightly the results of the adjustments, as in the case of HTO. Such diffusion coefficient values are not very realistic, because they correspond to a diffusion coefficient for iodide in free water higher than $2 \times 10^{-9} \text{ m}^2/\text{s}$. Furthermore, the description of the injection signal is far from being satisfactory. Based on that observation, additional numerical interpretations were carried out by Montarnal *et al.* [4]. The first task consisted in simulations using an initial iodide concentration of 1.6 g/L in the injection system. That initial concentration is supposed to take into account the effect of a fast initial disappearance of iodide in the injection system that may result from chemical or biochemical retention in the injection system or in the rock in contact with the injection chamber. The D_L/D_T ratio remained at 4.4. Those models show that the minimum quadratic error corresponds to a coefficient, D_L , ranging from 1.14×10^{-10} to $1.54 \times 10^{-10} \text{ m}^2/\text{s}$ for a porosity of 15 to 7%, although that parameter remains difficult to determine. Even if those values are consistent with a diffusive process and provide a satisfactory description of the concentration signal in the injection system, they do not allow for a description of the iodide-concentration profiles observed in the rock.

Because it is impossible to describe the full system data by just using the iodide quantities in the injection system diffusing gradually into the rock, the attention is focused on the interpretation of the concentration profiles in the rock. Consequently, new calculations only consider the rock

according to the same previous geometry. The iodide concentration in the injection system is considered as a time-dependent boundary condition at the rock interface. That boundary condition is described by an exponential function of a degree-2 polynomial. The D_L/D_T ratio remains at 4.4. The D_L and ω parameters are adjusted in such a way that the quadratic error on the concentration profiles was minimised. It is shown that an effective diffusion coefficient of $D_L = 1.5 \times 10^{-11} \text{ m}^2/\text{s}$ and a porosity of 12.5% are sufficient to describe correctly the entire set of tracer profiles in the rock (cf. Fig. 2). Fig. 3 shows the sensitivity of the adjustment to the values of D_L and ω . The porosity value is mostly sensitive to the concentration in the rock close to the injection borehole, while the concentration calculated in the middle of the profile does not depend very much on porosity. The conclusion should therefore be that the porosity value provided by the model corresponds most likely to a zone where the rock is disturbed by the injection borehole.

The simplest hypothesis to explain the behavior of iodide is to consider a chemical transformation. As a matter of fact, several authors describe the chemical fixation of iodide, either associated or not with biological activity. A brief examination of the literature shows that: (1) organic matter often plays an essential role in the chemical retention of iodide in natural porous environments [9] with a strong influence of the biological or enzymatic activity [10]; (2) the fixation of iodide on organic matter seems to result from covalent bonds [11]; (3) mineral phases, such as illite, metallic oxides and sulphides, are liable to fix significant quantities of iodide [12, 13]; (4) the fixation of iodide is affected by redox and pH conditions. Consequently, many chemical and biochemical mechanisms may have had an influence during the DI test and may have fixed iodide either reversibly or irreversibly. All those facts provide various potential explanations for the lack of mass balance for iodide that was not observed for HTO. One particular observation is that the conditions in the injection borehole are theoretically more favourable to iodide retention (non-sterile conditions, probable existence of iron oxides, gradually more positive redox potential) than those of the rock, although iodide retention in the rock cannot be excluded. At that stage of knowledge, two hypotheses may be formulated:

- According to the first hypothesis, iodide retention takes place almost entirely in the injection borehole or in its immediate vicinity (a few millimetres into the rock at the most). In that case, the last simulations carried out, on the basis of the interpretation of the iodide-concentration profiles, are valid and the D_L and ω parameters are correct.
- The second hypothesis assumes the existence of a major iodide retention mechanism in the rock, thus slowing down or partly stopping iodide migration in the rock. Parameter estimations by a model considering only diffusion are therefore biased, and the diffusion rate of iodide is underestimated by the D_L and ω parameters proposed here. The D_L coefficient would then be underestimated.

Presently, no distinction between those two assumptions can be made. However, it is noteworthy that a diffusive approach very well describes the concentration profiles of iodide in the rock.

Fig. 2. Upper: entire set of rock iodide contents along the different profiles vs. distance, comparison between experimental data and numerical results with $D_L = 1.5 \times 10^{-11} \text{ m}^2/\text{s}$, $D_T = 0.34 \times 10^{-11} \text{ m}^2/\text{s}$ and $\omega = 0.125$. Down: 3 profiles parallel to bedding plane (out of the entire set).

Fig. 3. Rock Iodide content along profile 10 (parallel to bedding plane) vs. distance. Comparison between experimental data and numerical results with various D_L and ω .

3. Laboratory measurements on centimetric core samples

A series of diffusion experiments were performed on Opalinus Clay samples. Here we focus on the most relevant data regarding the *in situ* DI test: diffusion parallel to bedding and effect of confining pressure for HTO, $^{125}\text{I}^-$ and $^{36}\text{Cl}^-$.

Early through-diffusion experiments using HTO and iodide ($^{125}\text{I}^-$) as tracers were performed on two samples from the SHGN niche (sandy facies). Deionised water was used as test solution for experiments with HTO. A NaI 0.2 M solution was used in the experiments. This NaI solution acted as carrier for the $^{125}\text{I}^-$ tracer [14]. Rock samples were sliced parallel and perpendicular to bedding into 1-cm-thick discs. Separate experiments for diffusion parallel and perpendicular to bedding were performed. In all cases, a uniaxial confining pressure of 9.5 MPa was applied parallel to the tracer concentration gradient. Melkior [14] used a three-dimensional numerical model to account for the effect of the end-plates and filters in the diffusion cells on the results of the experiment. According to the model calculations, the end-plates had a significant effect on diffusion. The fact of having only a limited number of openings through those end-plates caused tracers to be introduced from a few single point sources into the samples, instead of being introduced homogeneously over the sample surface. The calculated

values are the same for both samples: $D_L = 8.5 \times 10^{-11} \text{ m}^2/\text{s}$ and $\omega = 0.10$ for HTO and $D_L = 1.4 \times 10^{-11} \text{ m}^2/\text{s}$ and $\omega = 0.04$ for $^{125}\text{I}^-$.

Diffusion coefficients and accessible porosities for HTO, $^{36}\text{Cl}^-$ and $^{125}\text{I}^-$ were measured on Opalinus Clay samples (shaly facies) using the through diffusion technique perpendicular to bedding [15]. The confining pressures ranged from 1 to 5 MPa, the latter value simulating the *in situ* overburden pressure. The test solution was as close as possible to the Opalinus Clay pore water. The measured values of D_T are: $D_T = 1.2\text{--}1.5 \times 10^{-11} \text{ m}^2/\text{s}$ and $\omega = 0.009\text{--}0.11$ for HTO, $D_T = 4.0\text{--}5.5 \times 10^{-12} \text{ m}^2/\text{s}$ and $\omega = 0.05$ for $^{36}\text{Cl}^-$ and $D_T = 3.2\text{--}4.6 \times 10^{-12} \text{ m}^2/\text{s}$ and $\omega = 0.07\text{--}0.10$ for $^{125}\text{I}^-$. It is observed that increasing the pressure from 1 to 5 MPa resulted in a decrease of the diffusion coefficient of 17% for HTO, 27% for $^{36}\text{Cl}^-$ and 30% for $^{125}\text{I}^-$ [15]. One may mention the relatively high accessible diffusion porosity for $^{125}\text{I}^-$ relatively to the value for $^{36}\text{Cl}^-$, although their respective values of diffusion coefficients are very similar. This might be explained again by an effect of chemical retention of this tracer in the rock.

More recently, radial through diffusion experiments parallel to bedding using HTO and $^{36}\text{Cl}^-$ as tracers have also been performed at PSI [16]. A confining pressure of 7 MPa was applied perpendicular to bedding for all tests. The results for the tests parallel to bedding are as follows: $D_L = 5.42 \times 10^{-11} \text{ m}^2/\text{s}$ and $\omega = 0.17$ for HTO and $D_L = 1.59 \times 10^{-11} \text{ m}^2/\text{s}$ and $\omega = 0.082$ for $^{36}\text{Cl}^-$.

All the results indicate that (1) diffusion parallel to bedding is larger than diffusion perpendicular to bedding, (2) diffusion of halides is reduced compared to diffusion of HTO, which is consistent with an effect of anionic exclusion (3) D_L values for $^{125}\text{I}^-$ and $^{36}\text{Cl}^-$ are in good agreement.

4. Data comparison and conclusion

A set of diffusion data on the same media (Opalinus Clay) was obtained by the *in situ* DI experiment and by tests carried out in the laboratory. Fig. 4 summarises the values of

Fig. 4. Values of effective diffusion coefficient of HTO parallel to bedding plane.

the effective HTO coefficients obtained for diffusion parallel to bedding. The figure shows first that all of the results obtained with the through diffusion method on small-scale samples are within the same range as those obtained from the *in situ* profiles. This indicates that the disturbances of the rock tested in the laboratory have a moderate effect on the value of the diffusion coefficient. For the porosity accessible to diffusion for HTO, the value resulting from the interpretation of diffusion profiles is in line with the values obtained with the through diffusion method.

The values for effective diffusion coefficients parallel to bedding obtained by laboratory tests for iodide and the value from the *in situ* profiles are very close: 1.4×10^{-11} and 1.5×10^{-11} m²/s, respectively. The value for $^{36}\text{Cl}^-$ is also very similar (1.6×10^{-11} m²/s). The similarity between the through-diffusion results and the values from the *in situ* experiment must not give the false impression of great precision in the estimation of that parameter. In fact, the values for $^{125}\text{I}^-$ were obtained with chemical conditions that were non-representative of the site. It is recognised today that water salinity affects the anionic-exclusion process. Moreover, the estimation of the effective diffusion coefficient of iodide on the basis of the *in situ* concentration profiles is marred with uncertainties, notably in relationship to the possibility of chemical retention. Compared to the porosity accessible to iodide diffusion, observations must be limited to the fact that the value determined on the basis of *in situ* diffusion profiles is located in the value interval estimated by the tests conducted in a diffusion cell.

Finally, a more recent *in situ* diffusion test (DI-A) at the Mont-Terri URL using HTO, $^{22}\text{Na}^+$, Cs^+ and stable iodide as tracers [17] has to be mentioned. As observed with DI, the iodide concentration profiles in the rock are in a very good agreement with diffusion parameters from laboratory tests. Nevertheless, in DI-A no initial loss of iodide was observed. This discrepancy of iodide behaviour in the DI and DI-A experiments remains difficult to explain and the question of chemical retention of iodide in Opalinus Clay is still open.

References

- Thury, M., Bossart, P.: Mont Terri Rock Laboratory – Results of the Hydrogeological, Geochemical and Geotechnical Experiments Performed in 1996 and 1997. Swiss National Hydrological and Geological Survey, Report No. 23 (1999).
- Palut, J. M., Montarnal, P., Gautschi, A., Tevissen, E., Mouche, E.: Characterisation of HTO diffusion by an *in situ* tracer experiment in Opalinus Clay at Mont-Terri. *J. Cont. Hydrol.* **61**, 203 (2003).
- Wersin, P.: Geochemical modelling of bentonite porewater in high-level waste repositories. *J. Cont. Hydrol.* **61**, 405 (2003).
- Montarnal, P., Lamoureux, M.: DI Experiment: Numerical modelling of the experiment II. CEA report: Publication DM2S SFME/MTMS/RT/01-018 (2001).
- Bear, J., Verruijt, A.: *Modelling Groundwater Flow and Pollution*. D. Reidel Publishing Company (1994).
- Verpeaux, P., Millard, A., Charras, T., Combescure, A.: A modern approach of computer codes for structural analysis. In Proc. of the 10th Conf. on Structural Mechanics in Reactor Technology, Anaheim, USA (1989).
- Chavent, G., Roberts, J. E.: A unified physical presentation of mixed, mixed-hybrid finite elements and standard finite difference approximations for the determination of velocities in waterflow problems. *Adv. Water Res.* **14**(6), 329 (1991).
- Dabbene, F.: Mixed-Hybrid Finite Elements for Transport of Pollutants by Underground Water. Proc. of the 10th Int. Conf. on Finite Elements in Fluids, Tucson, USA (1998).
- Yoshida, S., Muramatsu, Y., Uchida, S.: Soil-solution distribution coefficients, K_d s, of I^- and IO_3^- for 68 Japanese soils. *Radiochim. Acta* **82**, 293 (1998).
- Christiansen, J., Carlsen, L.: Enzymatically controlled iodination reactions in the terrestrial environment. *Radiochim. Acta* **52/53**, 327 (1991).
- Mercier, F., Moulin, V., Guittet, J. M., Barré, N., Toulhoat, N., Gautier-Soyer, M., Toulhoat, P.: Application of NAA, PIXE and XPS for the quantification and characterisation of the humic substances/iodine association. *Radiochim. Acta* **88**, 779 (2000).
- Kaplan, D. I., Serne, R. J., Parker, K. E., Kutnyakov, I. V.: Radioiodide sorption to sediment minerals Scientific basis for nuclear waste management XXII. Mat. Res. Soc. Symp. Proc. **556**, 1059 (1999).
- Lefevre, G.: Mécanismes de sorption des ions iodures sur les oxydes et les sulfures de cuivre divisés – Perspectives environnementales. PhD Thesis. Université Henri Poincaré, Nancy I. 12th October (1999).
- Melkior, T.: Etude méthodologique de la diffusion de cations interagissants dans les argiles. Application à la mise en œuvre expérimentale et modélisation du couplage chimie-diffusion d'alcalins dans une bentonite synthétique. Publication CEA-DTA CEA-R-5890 (2000).
- Van Loon, L., Soler, J. M., Bradbury, M. H.: Diffusion of HTO, $^{36}\text{Cl}^-$ and $^{125}\text{I}^-$ in Opalinus Clay samples from Mont-Terri: effect of confining pressure. *J. Contam. Hydrol.* **61**, 73 (2003).
- Van Loon, L., Soler, J. M., Müller, W., Bradbury, M.: Anisotropic diffusion of HTO, $^{36}\text{Cl}^-$ and $^{22}\text{Na}^+$ in Opalinus Clay. Migration'03 conference (2003). Submitted to *Radiochim. Acta*.
- Van Loon, L. R., Wersin, P., Soler, J. M., Eikenberg, J., Gimmi, Th., Hernán, P., Dewonck, S., Savoye, S.: *In situ* diffusion of HTO, $^{22}\text{Na}^+$, Cs^+ and I^- in Opalinus Clay at the Mont Terri underground rock laboratory. Migration'03 conference. *Radiochim. Acta* **92**, 757 (2004).