Session 8

Transport Processes & Modelling

Chair: Joseph Stucki / Christophe Poinssot
ANION EXCLUSION EFFECTS IN COMPACTED BENTONITES: TOWARDS A BETTER UNDERSTANDING OF ANION DIFFUSION

Luc R. Van Loon, Martin A. Glaus, Werner Müller

Paul Scherrer Institut, CH-5232 Villigen PSI

Diffusion of $^{36}$Cl in compacted bentonite was studied using the through-diffusion technique (Glaus et al. 2007). Both the bulk dry density of the bentonite and the composition of the external solution were varied. Increasing the bulk dry density of the bentonite resulted in a decrease of both the effective diffusion coefficient and the Cl-accessible porosity. Increasing the ionic strength of the external solutions resulted in an increase of both the effective diffusion coefficient and the Cl-accessible porosity. The latter can be explained by anion exclusion effects (Donnan exclusion). At high ionic strength values ($1 \geq 1$ M NaCl) the Cl-accessible porosity approaches the free water porosity, i.e. the interparticle porosity. This interparticle porosity is the difference between the total and the interlayer porosity of the bentonite. The interlayer porosity was shown to depend on the degree of compaction. Up to a bulk dry density of 1200 kg m$^{-3}$ the interlayer is build up of three water layers. Between 1200 and 1600 kg m$^{-3}$ the interlayer water is reduced from three to two layers of water. Above 1600 kg m$^{-3}$ evidences for a further decrease to one layer of water were found. These findings are in full agreement with X-ray data from Kozaki et al. (1997) showing a decrease of the basal spacing of Na-montmorillonite (the main clay mineral in bentonite) with increasing degree of compaction.

The relationship between the effective diffusion coefficient of Cl$^-$ and the diffusion accessible porosity can be described by an empirical relationship analogue to Archie’s law, independent of the degree of compaction and the ionic strength of the pore water (Figure 2). To predict the effective diffusion coefficient of Cl$^-$ (or other monovalent anions) in compacted bentonite, the diffusion coefficient of Cl$^-$ in water, the bulk dry density of the bentonite and the ionic strength of the pore water have to be known.

![Figure 1](image1.png)

**Figure 1:** Total porosity and free water porosity as a function of the bulk dry density of compacted Na-bentonite (Volclay KWK).

![Figure 2](image2.png)

**Figure 2:** Relationship between the effective diffusion coefficient and the diffusion accessible porosity for the diffusion of $^{36}$Cl$^-$ in compacted Na-bentonite (Volclay KWK).
References


HIGH PERFORMANCE FVFE MULTIDOMAIN NUMERICAL METHOD. APPLICATION TO RADIONUCLIDE TRANSPORT THROUGH GEOLOGICAL LAYERS

M. Dymitrowska, M. Bourgeois, and G. Mathieu

IRSN - Institut de Radioprotection et de Sûreté Nucléaire, Av. du Gen. Leclerc BP n°17, 92262 Fontenay-aux-Roses, France (magdalena.dymitrowska@irsn.fr)

INTRODUCTION
To assess confinement capabilities of LL-HLW geological repository, IRSN perform numerical simulations of the radionuclide migration from the radioactive waste repository to the ground surface with the MELODIE software. Such a computational modelling is uneasy due to the strong variability of the repository properties and of the geometrical scales (from cm to km) modelled. Therefore, IRSN has developed numerical approaches within the MELODIE software in order to take into account these heterogeneities. This paper presents the numerical finite volume/finite element method (FVFE) implemented in the MELODIE software and, as an example of the performance of this method, describes transport calculations performed throughout the NF-PRO project. The purpose of these calculations is to assess the influence of hydraulic, chemical, and activity release rate assumptions on the radionuclide migration from the canisters through the repository.

NUMERICAL METHODS USED IN THE MELODIE SOFTWARE
The initial design of the MELODIE software was based on finite element (FE) method. The FE method, used to solve parabolic and elliptical problems, is not satisfying for dealing with discontinuities in solutions and with convection terms (especially when the convection is dominant). To improve the accuracy and the reliability of calculations, the FVFE method is adapted using a Galerkin development for time and diffusion type term discretization. The convection term is computed by a FV method using a Godunov P1 scheme with a possible extension to the second order. The FVFE method operates on triangular or tetrahedral meshes and requires constructing a dual mesh based on gravity centres of elements. The elementary exchange contributions are calculated on the dual mesh included in a primal meshing element to guarantee local conservativity of the method. To deal with the huge amount of calculation nodes, a domain decomposition strategy with Dirichlet-Dirichlet coupling is used based on embedded meshes strategy.

MODEL ASSUMPTIONS
The calculation domain, modelled in 3D, represents the main components of the repository placed in the middle of the geological layer of the Callovo-Oxfordian (COX) clay. Six disposal cells closed by bentonite plugs are connected to an access shaft through drifts as shown in figure 1. Seals are placed in the drifts and at the basis of the shaft to limit the water flow. The excavation-damaged zone (EDZ) surrounds the repository components. General design is based on data provided in NF-Pro for the spent fuel concept (NF-Pro, 2005). The global mesh is composed of 4,052,538 tetrahedral elements and 732,864 nodes distributed in 10 embedded meshes (noted 1 to 10 in the figure 1).

Migration of I-129 (soluble and non-sorbed), Nb-94 (non soluble and sorbed) and Se-79 (non soluble and non-sorbed) is simulated for various evolution scenarios taking into account an upward flow in the COX (0.4 𝜇m/yr). The migration is first studied considering that the components perform as they have been specified. Then, three alternative scenarios are created in order to assess:

- the influence of design considering a failure of the narrow trench filled with bentonite (of the drift seals), which is supposed to interrupt the EDZ and limit the water flow in the drift,
**Figure 1:** Above view of the repository - 10 embedded meshes.

- the influence of source term (UO₂ matrix and IRF) on radionuclide release and transfer,
- the influence of chemical form (anionic vs cationic form for Nb-94 and pessimistic parameters for Se-79) on their transport characteristics.

Calculated indicators, to quantify the radionuclide migration, are molar fluxes at the interfaces delimiting the disposal cell zone and corresponding to two radionuclide transfer pathways:

- the drift (ϕ1) throughout the bentonite plug,
- the host rock (ϕ2) throughout the EDZ.
- In addition, molar flux indicators are also calculated at the boundaries of the domain:
  - at the top and bottom limits of the host rock (ϕ3) to quantify the confinement properties of the formation and transfer throughout the host rock,
  - at the exit of the shaft (at the top of the host rock) (ϕ4) to assess the relative importance of the shaft pathway with respect of host rock pathway.

**RESULTS**

Calculations performed showed that proposed FVFE method is efficient to solve the equations of flow and advection-diffusion transport in highly heterogeneous porous media. Contrary to the FE method, the FVFE method ensures that no negative concentration occurs, even close to the source of activity in spite of the coexistence of contrasted transport regimes. In the first scenario treated as a reference scenario, the major part of the activity is transferred through ϕ2 (90 % for I-129) because of the diffusive transport regime within the cell. I-129 fluxes are the highest essentially because the major part of Se-79 is precipitated in the vicinity of the canister and Nb-94 is strongly sorbed in clayey components. Then, the sensitivity calculations allow to highlight the following interesting results:

- Dead-end design ensures a diffusive transport regime in the disposal cell, even if the advection is the dominant transport regime in the access drifts. For I-129, 80 % of the released activity is still transferred through ϕ2.
- The presence of the IRF in the UOX canister limits the relevant role of UO₂ matrix degradation to delay activity release for I-129. In fact, the amount of activity instantaneously released (8 % of the total activity) fixes a lower limit for the calculated molar fluxes.
- When Se-79 are considered as soluble and Nb-94 as a less sorbed cation, molar fluxes of those radionuclides reach almost the levels of I-129 fluxes at interfaces in the vicinity of the cells. As a matter of fact, Se-79 and Nb-94 inventories (in mol) are more or less the same as I-129 one and radioactive decay doesn’t play any role because of the low transfer distances (for ϕ1 and ϕ2).

Complementary calculation results and a more detailed analysis will be given in the final paper.
IMPACT OF BENTONITE EXTRUSION
ON RADIONUCLIDE MIGRATION
IN A SATURATED PLANAR FRACTURE

R. A. Borrelli and J. Ahn

Department of Nuclear Engineering, University Of California – Berkeley
Berkeley, California, 94720 – 1730, USA

INTRODUCTION
Effective strategies for high level radioactive waste (HLW) management require long term isolation from the environment. Isolation of HLW is based on the utilization of a multibarrier concept; to achieve this objective assures that the failure of an individual component will not result in total failure of the entire system. In the engineered barrier system, (EBS), for the saturated repository, the function of the buffer is to isolate the waste package from near field processes. Compacted bentonite will be utilized for the buffer material. This study presents a coupled mathematical model for radionuclide transport in a saturated planar fracture that includes extrusion of bentonite. Previous modeling studies of radionuclide transport in a two phase system do not include impacts of bentonite swelling and extrusion into the fracture on the transport of radionuclides. In the present study, a model has been derived describing radionuclide migration through the saturated planar fracture and incorporating characteristics of bentonite: extrusion into the fracture, sorption, and effects of bentonite swelling on groundwater flow.

MODEL
In Figure 1, for this analysis, a planar fracture assumed to be initially filled with water intersects water saturated bulk bentonite. The bulk bentonite has a cylindrical shape, and extrusion occurs in a radial direction into the intersecting planar fracture. A radionuclide source is located at the intersection between the bulk bentonite and the fracture ($R_a$). A constant concentration is assumed at the source ($N'$). Beyond the tip of the extruding bentonite, it is assumed that only water fills the fracture.

Based on the definition of the domain, two distinct regions exist in the model space. One region is defined by the extruding bentonite, and the other region exists beyond the expanding bentonite, containing only groundwater. The transport model is valid for both regions; however, due to the extruding bentonite, parameters in the model will be different according to the specific region in the domain. Furthermore, because bentonite extrudes into the fracture, the region of extrusion will also vary with time. In the present model, existence of bentonite particles beyond the tip in the form of colloidal suspension is neglected.

A model for movement of water and bentonite in a planar fracture is coupled to another model for radionuclide transport in the same planar fracture by fluid phase and solid phase advection, diffusion, and sorption onto moving bentonite. Movement of water and bentonite is quantified through space – time

![Figure 1](image_url)

**Figure 1:** Domain for radionuclide transport modeling.
Figure 2: Radionuclide transport with fluid phase advection, solid phase advection, and sorption.

dependent porosity and tip location of extruding bentonite. The radionuclide transport model is unique in that a solid phase movement is included and in that water will flow in a direction countercurrent to the direction of bentonite extrusion. Finite element solutions have been derived for the porosity and for the radionuclide concentration.

RESULTS AND DISCUSSIONS

Results are presented in terms of the spatial distribution of the radionuclide in the water phase in the fracture as a function of time.

The results shown in Figure 2 indicate that the sorption distribution coefficient ($K_d$) and the solid phase advection provide a significant impact on the behavior of the system. For this sorption distribution coefficient, radionuclide concentration exceeds the constant concentration at the source. Furthermore, for this simulation radionuclides are contained completely within the region of bentonite extrusion at each time step. In the region near the continuous radionuclide source, concentration will consistently remain high in the fluid phase. Therefore, many radionuclides sorb to the solid particles in the system. These radionuclides are then transported in the fracture due to the solid phase advection. Radionuclides then desorb from the solid material into the fluid phase in a region where fluid phase concentration is low.

The results shown by Figure 2 suggest that if short lived radionuclides remain completely contained within the extruding bentonite region, then they could eventually vanish, due to decay. This added barrier to release could considerably affect assessments of repository performance. This possibility suggests importance of the region in the vicinity of buffer/rock interface. Validation of this model can allow for long term analyses and eventual inclusion into future repository performance assessments.
THE DI-B IN-SITU DIFFUSION EXPERIMENT
AT MONT TERRI: RESULTS AND MODELING

J. M. Soler¹, J. Samper², A. Yllera³, A. Hernández³, A. Quejido³, M. Fernández³,
C. Yang², A. Naves², P. Hernán³, P. Wersin²

1. ICTJA-CSIC, Lluís Solé i Sabarís s/n, 08028 Barcelona, Spain (jsoler@ija.csic.es)
2. Univ. de A Coruña, Campus de Elviña, 15192 A Coruña, Spain (jsamper@udc.es)
3. CIEMAT, Avda. Complutense 22, 28040 Madrid, Spain (abel.yllera@ciemat.es)
4. ENRESA, Emilio Vargas 7, 28043 Madrid, Spain (pher@enresa.es)
5. NAGRA, Hardstrasse 73, CH-5430 Wettingen, Switzerland (paul.wersin@nagra.ch)

INTRODUCTION

The DI-B experiment was a Spanish Project led by Enresa and financed by Enresa and Nagra. One of its main objectives was to study the transport and retention properties of selected tracers in the Opalinus Clay, but using only non radioactive species. The selected tracers (Yllera et al., 2004) were HDO (water), I⁻ (anionic species), ⁹Li⁺ (non- or weakly-sorbing cation) and ⁷Rb⁺ (strongly-sorbing cation).

EXPERIMENTAL CONCEPT

The diffusion experiment was carried out in the BDI-B3 borehole located at the entrance of the DI niche in the Gallery 98 of the Mont Terri underground rock laboratory. This zone of the Mont Terri tunnel corresponds to the shaly facies of the Opalinus Clay, a few meters away from the underlying formation (Jurassic Marl, Toarcian). The in-situ diffusion experiment was performed as a single point dilution test by injecting stable tracers into a packed-off section at the bottom of a vertical borehole 7.7 m deep. The dip of the bedding was 32° to the SE. The length of the borehole interval where tracers were injected was 0.6 m and the radius of the borehole was 0.038 m. The experiment included downhole and surface instrumentation. Downhole instrumentation consisted of a pneumatic single packer system with a porous screen made of sintered stainless steel mounted just below the packer at the bottom of the borehole. Surface instrumentation included a closed 316-L stainless steel circuit intended to circulate the synthetic porewater containing the tracers. Injection and monitoring of tracer concentrations were done from the main tank through a valve system. A volume of rock around the injection interval was overcored after the end of the experiment, allowing the measurement of tracer distribution profiles. The experiment lasted slightly over one year.

RESULTS AND INTERPRETATION

The results of the diffusion experiment included the evolution of tracer concentrations in the injection system and tracer distribution profiles in the rock. Two and three-dimensional modeling of the results has been performed using the CRUNCH (Steefel, 2006) and CORE³D (Yang et al., 2003) reactive transport codes, respectively. The experiment was designed so the length of the injection interval was larger than tracer transport distances. This experimental setup favors, in principle, a simple 2D interpretation of the results, since diffusion occurs mainly along bedding planes. Samper et al. (2006) performed a full 3D modeling of the HDO and I⁻ results. Their reported values for the effective diffusion coefficient parallel to bedding, anisotropy ratio and accessible porosity for both tracers are:

\[ \begin{align*}
\text{HDO:} & \quad D_{\parallel}=4.0\times10^{-11} \text{ m}^2/\text{s}, \quad D_{\perp}/D_{\parallel}=4, \quad \phi=0.15 \\
\text{I⁻:} & \quad D_{\parallel}=8.3\times10^{-12} \text{ m}^2/\text{s}, \quad D_{\perp}/D_{\parallel}=4, \quad \phi=0.08
\end{align*} \]

Samper et al. (2006) also performed a sensitivity analysis of the results with respect to different parameters. They showed that the effect of considering only diffusion along the bedding planes (\(D_{\parallel}/D_{\perp}=100\)) was
relatively minor, causing only a significant effect on tracer distribution in the rock near the top and bottom edges of the injection interval.

The results from 2D modeling using CRUNCH are:

\[
\text{HDO: } \text{De}_\text{HDO}=5.1\times10^{-11} \text{ m}^2/\text{s}, \phi=0.22 \\
\text{I: } \text{De}_\text{I}=1.2\times10^{-11} \text{ m}^2/\text{s}, \phi=0.09
\]

The difference in the results may be due to the different geometries (3D vs. 2D), to other aspects of the calculations or to how model and experiments are compared. At any rate, the differences between the two sets of results are small and confirm the smaller effective diffusion coefficient and accessible porosity of I with respect to HDO. Also, it is worth noting that the difference between the 2D and the 3D approaches should become smaller for sorbing tracers, since their transport distances are smaller.

The results corresponding to \(^{4}\text{Li}\) and \(^{87}\text{Rb}\) have also been modeled using both 2D and 3D approaches. For \(^{4}\text{Li}\) (C\(_0\)=3×10\(^{-3}\) mol/l), the best fit for both models was achieved with De\(_\text{L}\)= 7×10\(^{-11}\) m\(^2\)/s, Kd=0.24 l/kg and \(\phi=0.164\) (capacity factor \(\alpha=0.7\)). The De and Kd values are very similar to those obtained previously for Na\(^+\) at Mont Terri (Van Loon et al., 2004; Wersin et al., 2006). The Kd value is certainly larger than what was initially expected.

Regarding \(^{87}\text{Rb}\) (C\(_0\)=3×10\(^{-7}\) mol/l), only three profiles were finally measured, and they showed significant variability. This variability has also been observed for other strongly sorbing tracers (Cs\(^+\), Co\(^{2+}\)) in the DI-A and DI-A2 in situ experiments at Mont Terri. It has not been possible to obtain unique values for De\(_\text{L}\) and Kd for \(^{87}\text{Rb}\), although strong sorption has been confirmed.

References:


IN-SITU DIFFUSION TEST IN OPALINUS CLAY WITH TRITIUM, ANIONIC AND CATIONIC TRACERS: COMPARISON WITH LABORATORY EXPERIMENTS AND MODELLING

P. Wersin¹, J. Soler², L. Van Loon³, S. Dewonck¹, J. Eikenberg³, B. Baeyens³, D. Grolimund³, T. Gimmi³

1. NAGRA, Hardstrasse 73, CH-5430 Wettingen, Switzerland
2. CSIC-IJA, E-08028 Barcelona, Spain
3. PSI, CH-5232 Villigen, Switzerland
4. ANDRA, F-55290 Bure, France

INTRODUCTION

Argillaceous rocks are considered as ideal candidates for hosting waste repositories because of their very low permeability and good retention properties for contaminants. The transport and retention properties of such diffusion-dominated systems need to be tested and quantified at different scales by carefully designed laboratory and in-situ studies, and, if possible, also by tracer profiles at the formation scale. At the Mont Terri Rock Laboratory, several in-situ diffusion studies have been successfully carried out and consistent diffusion data for non-reactive tracers has been achieved (Tevissen & Soler, 2003). In addition, a first migration test, termed DI-A1, including the sorbing cationic tracers Na⁺ and Cs⁺ yielded consistent results (Wersin et al., 2004; Van Loon et al., 2004) with lab diffusion data.

Based on the promising outcome of the DI-A1 test, it was decided to perform a second test DI-A2 with the objective of (i) confirming the available in-situ lab diffusion data and (ii) focussing in more detail on the diffusion of reactive tracers. These included $^{85}$Sr²⁺, Cs⁺, $^{60}$Co²⁺ and Eu³⁺. In addition, the anionic tracers Br⁻ and I⁻ as well as HTO were applied. The results obtained from the in-situ test were interpreted by modelling and comparison with lab diffusion data on small-scale samples.

EXPERIMENTAL METHODS

The in-situ experiment consisted of a single borehole through which synthetic porewater including the tracers was continuously circulated. Diffusion into the rock occurred from the 1-m packed-off test interval. A hydrotest prior to tracer injection indicated a low hydraulic conductivity in the range of 1.3×10⁻¹³ m·s⁻¹ in the test interval zone. The tracer concentrations and main ion composition in the borehole were regularly monitored. After 12 months a number of tracer profiles of an overcored section was analysed. The equipment was removed and a leaching test was performed. This indicated that only minor sorption to equipment had occurred, except for Eu³⁺, where the major part had been taken up by the screen material. The results of the tracers, except for Eu³⁺, were interpreted with a 2D diffusion/sorption model using the CRUNCH code. The aim was to obtain values for diffusion coefficients, accessible porosities and sorption parameters and compare these with results from small-scale laboratory experiments.

RESULTS AND CONCLUSIONS

The composition of the synthetic porewater remained fairly constant with time, but some increase in sulphate and alkalinity was observed in the first months. This indicates that the geochemical disturbance induced by the borehole and the circulation system was limited to some pyrite oxidation resulting from the drilling procedure. The evolution of the tracers HTO, $^{85}$Sr²⁺ and $^{60}$Co²⁺ in the circulating fluid and for a typical rock profile is illustrated in Figure 1 (data shown by points). The evolution of the other tracers Br⁻,
Figure 1: Tracer concentrations for HTO, $^{85}$Sr$^{2+}$ and $^{60}$Co$^{2+}$ in circulating fluid (left) and in one representative rock profile (right). Points represent experimental and lines simulated data.

I$^-$ and Cs$^+$ is not shown. All these tracer data were analysed by 2D modelling, assuming that diffusion occurred predominantly in the bedding plane.

From the experimental and modelling data, the following main conclusions can be drawn:

- In general, the diffusion and sorption data obtained from the DI-A2 in-situ test is consistent both with previous in-situ data and with lab diffusion and batch sorption data.

- The diffusive flux of anions into the rock is lower than that of water tracers because of anion exclusion, that is, because only a fraction of the pore space is accessible for anions.

- The diffusive flux of cations into the rock tends to be higher than that of water tracers, which may be related to an additional driving force for diffusion in the diffuse double layer around the clay surfaces.

- The interpretation of the diffusion of strongly sorbing tracers which display inhomogeneous spreading into the formation remains a challenging task. This calls for the application of novel spectroscopic analytical methods.

References:
OSMOTIC EXPERIMENT
AT THE MEUSE/HAUtte-MARNE URL
(FRANCE)

M. Cruchaudet¹, J. Croiset², J.M. Lavanchy²

1. Andra, Laboratoire de Recherche Souterrain de Meuse / Haute-Marne, RD960 BP9 55290 Bure, e-mail: martin.cruchaudet@andra.fr
2. Colenco Power Engineering AG, Groundwater Protection and Waste Disposal, Täfernstr. 26, CH-5404 Baden- Switzerland, (jean.croise@colenco.ch)

INTRODUCTION
Andra is currently performing a detailed characterisation of the geochemical, geological, geomechanical and hydrogeological properties of the Callovo-Oxfordian argillaceous formation at the Meuse/Haute-Marne Underground Research Laboratory (France). Amongst the experiments carried out since 2004, the so-called PEP experiment deals with in situ measurements of pore pressure and permeability in the undisturbed rock throughout the Callovo-Oxfordian. One of the objectives for this experiment was to determine how water salinity could impact pore pressure measurements, in particular due to osmotic effects and a specific series of tests was carried out, which is named the “osmotic experiment” in the following.

EXPERIMENTAL CONCEPT
The osmotic experiment was carried out in a 20 m deep borehole (PEP1001) of 101 mm diameter, drilled from a drill located at 490 m depth. This borehole was air-drilled and equipped with a multi-packer system isolating the 4 intervals of 1 m length each. In each interval, a strainer was set to avoid excessive creeping of the borehole and to have a minimum water volume, in each interval of 2.5 L.

The intervals were first saturated with a water of chemical composition close to that of the porewater in the Callovo-Oxfordian. After an initial pressure recovery phase, permeability tests were carried out in each of the four intervals. The permeability values obtained in this borehole were ranging from 8.10⁻¹⁴ to 7.10⁻¹⁵ m/s.

Pressure stabilisation was monitored for more than four months, the lines to the intervals being kept closed. The concept for the osmotic experiment was then to replace water in two of the intervals by either water of higher salinity or lower salinity, maintaining the pressure as constant as possible during all the water exchanging phase. This concept was derived from that of a similar experiment at the Mont Terri URL in Switzerland (e.g. Noy et al., 2004).

In March 2006, two water exchanges in two different intervals were performed with respective salinities of 1 g/L and 7 g/L, in order to observe if such salinities could produce a measurable change in pressures of the intervals (due to the osmotic capacity of the clay).

In October 2006, three additional water exchanges in three different intervals were performed with respective salinities of 5 g/L (corresponding to the expected salinity in the interval), 25 g/L and 100 g/L.

RESULTS AND INTERPRETATION
In March 2006, pressure variations measured in the intervals were higher than expected from initial simulations. Furthermore, pressures were increasing in both intervals whereas the salinity for water injected was in one case higher than salinity in the interval and in the other case lower. The analysis of both the pressure and the temperature measurements indicated that the temperature difference between water injected and water in the intervals was most probably responsible for the pressure variations observed; the pressure response due to osmosis was masked.
In October 2006, the second test series was performed with careful monitoring and regulation of the temperature of the injected water. Temperature variations of less than 0.3 °C were measured during the tests. For the “blank” test performed with water of 5 g/L of salinity, the pressure was slightly perturbed and increased by 30 kPa but had recovered to its initial level within 24 hours. For a water exchange with a salinity of 25 g/L, a maximum pressures increase of 50 kPa was observed after 10 days, whereas in the same time, for the exchange with a salinity of 100 g/L, a maximum of 110 kPa was observed.

An interpretation of this chemical-osmotic experiment by means of numerical simulations was carried out (Croisé and Lavanchy, 2006), which followed the approach as described in Garivato et al., 2006. Results obtained from the simulations provided an effective diffusion coefficient ranging from 1.10^{-11} and 5.10^{-11} m²/s and a coefficient of osmotic efficiency ranging from 5 % to 18 % which are very consistent values with a mean distances between clay platelets of 20-50 Å.

References:
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