



# Applying the squeezing technique to highly consolidated clayrocks for pore water characterisation: Lessons learned from experiments at the Mont Terri Rock Laboratory



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## ABSTRACT

Knowledge of the pore water chemistry in clayrock formations plays an important role in determining radionuclide chemical speciation and migration in the context of nuclear waste disposal. Among the different *in situ* and *ex situ* techniques for pore water sampling in clay sediments and soils, the squeezing technique dates back 115 years. Although different studies have been conducted on the reliability and representativeness of squeezed pore waters, most of these involved high-porosity, high water content and unconsolidated clay sediments. Very few of them analysed squeezed pore water from low-porosity, low water content and highly consolidated clayrocks.

In this paper, a specially designed and fabricated one-dimensional compression, two-directional fluid flow cell was used to extract and analyse the pore water composition of Opalinus Clay core samples from Mont Terri (Switzerland) with water contents between 6.2 and 7.8 wt.%. A study of the influence of the squeezing pressure on the chemistry of the pore water was performed at pressures of up to 200 MPa. To validate and demonstrate the validity of the squeezing technique for obtaining reliable pore water, different tests were performed at various pressures and the squeezed waters were compared with those obtained by other methods, such as *in situ* collected borehole waters. The reproducibility and quality of the squeezing method was also checked, as well as different artifacts which could affect the reliability of the pore water data.

The results show that the reproducibility of the technique is good and no membrane effect (ionic ultrafiltration, chemical fractionation, anion exclusion) was found in Opalinus Clay at Mont Terri in the range of pressures analysed (70–200 MPa). Pore waters extracted in this range of pressures do not decrease in concentration as a function of pressure, which indicates that a dilution of water by mixing of the free pore water and the outer layers of diffuse layer water does not occur and a threshold (safety) squeezing pressure of 175 MPa could be established to avoid membrane effects. Furthermore, a direct comparison against *in situ* collected borehole waters shows that the pore waters extracted at these pressures are representative of the Opalinus Clay Formation at Mont Terri. Extraction artifacts such as temperature effect, oxidation and degassing can be avoided or minimised by taking special precautions when preparing and handling the core sample and extracting the pore water. Finally, we show that it is possible to obtain insightful information on anion-accessible porosity by combining squeezing data with aqueous leaching data to characterise the pore water composition and solute distribution for different rock porosities.

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## 1. Introduction

Argillaceous rocks with low permeability have been selected in several countries, such as Belgium, Switzerland and France

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(Ondraf-Niras, 2001; Nagra, 2002; Andra, 2005), as potential geological host rocks for the disposal of high-level radioactive waste (HLRW). The host rock represents the most important barrier to the migration of radionuclides to the surface environment, and clay minerals have favourable properties, such a large sorption capacity (ion exchange and surface complexation) and low permeability, which reduce water movement. Clayrock pore water is generally reducing with near neutral pH values, ensuring that

the solubilities of a large number of radioelements will be very low. Besides, the swelling capacity of some clay particles will cause the sealing of potential fractures and, as a consequence, significant retardation in the transport of most radionuclides (Horseman and Volckaert, 1996).

In order to determine the suitability of these clayrocks for waste disposal, various evaluations of the hydrogeochemistry and transport mechanisms from these geological formations to the biosphere must be conducted. The migration of radionuclides through the geosphere will occur predominantly in the aqueous phase, and hence the pore water chemistry plays an important role in determining the radionuclide migration characteristics in clay–rock formations, especially with respect to actinides and other redox sensitive elements (Bradbury and Baeyens, 1998; Altmann, 2008; Mazurek et al., 2011). Furthermore, a knowledge of the chemical composition of the clayrock pore water is essential to the design of safety scenarios because it establishes the base line chemistry of the undisturbed system to estimate the evolution of structural components (waste matrix dissolution, canister corrosion, sorption of mineral surfaces, dissolution of packaging and containers, engineered barrier degradation: bentonite, concrete, etc.), contributing to the engineering design and the chemical modelling of radionuclides in the near- and far-field environments.

Therefore, collecting representative pore water solutions from clayrocks is one of the main objectives of any hydrogeochemical research programme in argillaceous formations. A large effort has been undertaken to characterise the pore water chemistry in selected clayrock formations in Europe (Boom Clay (Belgium), Opalinus Clay (Switzerland) and Callovo-Oxfordian (France)) in order to understand the underlying geochemical processes and to establish a reliable model for the main water–rock interactions which control the physicochemical parameters and the chemistry of the main elements in the clay–water–solute system.

One of the most important problems in the study of the pore water chemistry in clayrocks is the validity of the sampling procedures. Different reviews of pore fluids extraction techniques can be found in Kriukov and Manheim (1982), Adams (1994), Reeder et al. (1998), Sacchi and Michelot (2000), Sacchi et al. (2001) and Bath et al. (2001). Recovering the pore water solution representative of *in situ* conditions from low permeable and low water content systems is very difficult, seems sometimes impossible and research efforts over the last 15 years have focused on developing techniques for determining the pore water composition of clayrocks in the context of waste disposal, including both direct methods (from borehole core samples or dedicated experiments carried out in underground research laboratories, URL) and indirect methods (from geochemical modelling):

1. *In situ* pore water sampling from sealed boreholes drilled from the rock laboratory, which offers direct measurements of seepage water chemistry (Pearson et al., 2003a, 2003b; De Craen et al., 2004a; Vinsot et al., 2008a, 2008b).
2. Laboratory pore water sampling from unaltered core samples by the squeezing technique at high pressure (Entwisle et al., 1989; Entwisle and Reeder, 1993; Fernández et al., 2003; De Craen et al., 2004b) or advective displacement experiments (Mäder et al., 2004).
3. Characterisation of water chemistry by geochemical modelling based on: (a) measured laboratory properties of the rock, such as mineralogy, physical properties (water content, dry density, porosity), physicochemical properties (aqueous leaching, cation exchange capacity, cation exchange population, surfaces areas, cation exchange selectivity coefficients, surface complexation properties, diffusion coefficients, etc.); (b) fixed concentration of non-reactive solutes (anion inventories); (c) constrains

related to mineral, cation exchange and redox equilibria; (d) understanding of the water–rock reactions and transport processes involved in the clayey system; and (e) a good selection of the thermodynamic database (Bradbury and Baeyens, 1998; Beaucaire et al., 2000; Arcos et al., 2001; Pearson et al., 2003a, 2003b; De Craen et al., 2004b; Gaucher et al., 2009; Appelo et al., 2008; Pearson et al., 2011).

Modelling work seeks to understand the processes for pore water composition controls and helps to extrapolate these results to other locations from which water samples are not available. However, confidence in modelling work results ultimately relies on a good agreement between predictive models and *in situ* or squeezing measurements (Vinsot et al. 2008b; Gaucher et al., 2009; Pearson et al., 2011).

Measuring the *in situ* composition of clayrock pore water from packed-off boreholes drilled in a rock laboratory is expensive compared to laboratory tests, and the results are often affected by experimental artifacts: oxidation, degassing, microbial growth, etc. (Wersin et al., 2011; Vinsot et al., 2008a,b). Moreover, *ex situ* water collection methods are often necessary when *in situ* water collection is not viable or requires too long sampling times. The aqueous leaching technique has been used to obtain pore water composition. However, as we will discuss below, this method influences the solubility of certain mineral phases and the cation exchange concentrations, modifying the real pore water chemical composition. Therefore, the squeezing technique can offer a good alternative method for measuring pore water composition, especially for chloride, bromide and stable isotopes ( $\delta^{18}\text{O}$  and  $\delta^2\text{H}$ ).

In this paper, a study of the pore water chemistry of the Opalinus Clay Formation in Mont Terri (Switzerland) was conducted by using the squeezing technique. The squeezing studies of the Opalinus Clay core samples carried out in the context of the “Geochemical Modelling Task” from the Mont Terri Project over more than 15 years have been used to validate the representativeness and the reproducibility of the squeezing technique for analysing the ion and isotope data in pore waters. First, a complete summary of the squeezing studies performed over time is outlined. Then, the influence of the squeezing pressure up to 200 MPa is examined, evaluating all the possible causes and suggesting a threshold squeezing pressure for this clayey formation. Different factors that may influence the reliability of pore water chemical composition extracted by squeezing are then examined. Finally, anion-accessible porosity values for chloride are determined, i.e., the average porosity water volume in which chloride is present at the same concentration as in bulk water far from any electrostatic field created by the negatively charged clay surfaces.

## 2. State of the art of the squeezing technique

### 2.1. What is squeezing?

Squeezing is analogous to the natural process of consolidation, caused by the deposition of material over geological time, but at a greatly accelerated rate. The squeezing process involves the expulsion of interstitial fluid from a saturated porous medium being compressed if the pore space can be sufficiently reduced by compaction (Entwisle and Reeder, 1993). When a stress or load applied to a soil or sediment is suddenly changed, this change is absorbed jointly by the interstitial fluid and the mineral skeleton as a consequence of the discontinuous nature of the sediment (composed of solid particles and spaces or voids filled with water). The change in pore pressure will cause water to move through the soil, hence the properties of the soil will change with time (Terzaghi, 1925).

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