Physics and Chemistry of the Earth 33 (2008) 1019–1025

Contents lists available at ScienceDirect

Physics and Chemistry of the Earth

journal homepage: www.elsevier.com/locate/pce

Overview and consistency of migration experiments in clay

M. Aertsens *, P. De Cannière, K. Lemmens, N. Maes, H. Moors

SCK-CEN, Boeretang 200, B-2400 Mol, Belgium

ARTICLE INFO

Article history:
Available online 18 June 2008

Keywords:
Migration experiment
Diffusion accessible porosity
Apparent diffusion coefficient
Dispersion coefficient
Boom clay
Silica

ABSTRACT

An overview is given about the types of migration experiments in clay and their corresponding mathematical models aimed to determine: (1) the product ηR of the diffusion accessible porosity η and the retardation factor R, and (2) the apparent dispersion coefficient D. An example shows that not all set-ups allow to estimate values for these parameters. An important concern is consistency.

Consistency requires a good correlation between the diffusion accessible porosity η (fitted from migration experiments) of tritiated water and the independently measured water content. Column migration experiments (pulse injection type) on compact clay cores taken at different depths in the Ypres Clay Formation, confirm this correlation and lead to the expected value of the grain density, showing that nearly the entire porosity of the clay is accessible for water diffusion.

Another requirement for consistency is that different experimental set-ups lead to the same value for the migration parameters. The average values of the apparent diffusion coefficient of dissolved silica obtained with different types of tests in Boom Clay are, respectively, 6×10^{-13} m^2/s (in-diffusion, 25 °C), 2×10^{-13} m^2/s (percolation, 25 °C) and 9×10^{-13} m^2/s (combined glass-dissolution/diffusion, 30 °C). The values of the product ηR are around 60–70 (in-diffusion) and between 100 and 150 (percolation). Using fit results from the combined glass-dissolution/diffusion experiments with other results, leads to ηR values ranging from 40 to 90. The values of the migration parameters obtained from different types of experiments and the corresponding models are consistent.

1. Introduction

In Belgium, Boom Clay and Ypres Clay are considered as reference host formations for research studies on the disposal of high-level radioactive waste. A safety analysis of the disposal system requires the values of the migration parameters of radionuclides in the clay: (1) the product ηR of the diffusion accessible porosity η and the retardation factor R, and (2) the apparent dispersion coefficient D (m^2/s). Several types of migration experiments are used to determine these migration parameters in clay. An important concern deals with the consistency of the different types of experiments: do they lead to the same values for the migration parameters and are the predicted parameter values consistent with observations from independent measurements?

Two types of experiments are applied: (1) pure diffusion tests, with a concentration gradient only, and (2) column migration tests with an additional hydraulic pressure gradient. Pure diffusion tests allow to determine the apparent diffusion coefficient D_{app} (m^2/s), while the second type of experiments provides the apparent dispersion coefficient D (m^2/s). Both are related by $D = D_{app} + \alpha_D V$ with α_D the dispersion length (m) and V the apparent velocity of the tracer (m/s). For low permeability cores and low pressure gradients, the assumption $D_{app} \gg \alpha_D V$ is acceptable, and the value of the apparent diffusion coefficient D_{app} is approximated by the apparent dispersion coefficient D.

For water saturated clays, the total porosity η_{tot} is the volume fraction of the clay occupied by water. Part of the total porosity is not available for transport due to: (a) electrochemically bound water at the clay/water interface, (b) the Donnan effect excluding negatively charged ions from accessing narrow interparticle space in the clay, and (c) possible effects of size exclusion and dead end pores. Water flow predominantly occurs through large pores between the clay grains. Although water hardly flows between clay platelets, diffusion causes transport there. Put and Henrion (1992) distinguish 'effective porosity', the porosity available for water flow, and 'diffusion accessible porosity' η, which has a higher value than the effective porosity. Kato et al. (1995) use a different terminology and define for pure diffusion experiments 'effective porosity' as 'the porosity where the species can diffuse through'. Clearly, this is the 'diffusion accessible porosity' η of Put and Henrion (1992) and Henrion et al. (1991), simply called 'accessible porosity' by Garcia-Gutiérrez et al. (2004) (concerning pure diffusion experiments). Using the terminology of Put and Henrion

* Corresponding author.
E-mail address: marc.aertsens@sckcen.be (M. Aertsens).

1474-7065/$ - see front matter © 2008 Elsevier Ltd. All rights reserved.
doi:10.1016/j.pce.2008.05.019

(1992), the diffusion accessible porosity η includes the effective porosity, and describes the space where transport occurs in a pure diffusion experiment as well as in a combined dispersion/advection experiment. In the latter case, no distinction will be made between porosity available for (water) flow and porosity accessible for diffusion.

This paper describes experimental results for two tracers: tritiated water (HTO) and dissolved silica ^{32}Si ($Si(OH)_4$ or $SiO(OH)_3^-$). The silica migration parameters in Boom Clay are determined because silica is the major component of nuclear waste glasses and the long term glass-dissolution rate could be controlled by diffusion and sorption of silica in the clay (Pescatore, 1994). For HTO, the diffusion accessible porosity η, determined by dispersion/advection experiments, is compared with independent measurements of the water content (Aertsens et al., 2003a). The migration parameters of silica in Boom Clay have been determined using different types of experiments and are compared to check their consistency. It is examined that similar set-ups allow to obtain values for all migration parameters or not.

After describing the different types of experiments used for both tracers, the corresponding models are explained, followed by a discussion of the experimental results with respect to their consistency.

2. Experimental

2.1. Experimental set-ups

The tracer source can be placed at one side of the clay core or sandwiched between two clay cores. For each type of experiment it is possible: (a) to measure as a function of time the tracer concentration in water at one or both sides of the clay core, and (b) to determine the tracer profile in the clay core at the end of the experiment. Fig. 1 shows a schematic overview of the set-ups.

In the simplest set-up, termed 'percolation experiment', an aliquot of tracer spiked either directly on the clay surface or on a filter paper, is sandwiched between two back-to-back clay cores. The whole is confined between two filters in a permeation cell and continuously percolated with clay water. A pulse injection experiment is a variant of the previous set-up, where the permeation cell contains only one clay core confined between two porous filters. An aliquot of tracer is injected at the inlet filter at zero time. In both types of experiment, the water flowing out of the system is collected and the tracer concentration in the water is measured as a function of time. Dispersion/advection experiments allow the simultaneous determination of the hydraulic conductivity and the migration parameters of the tracer in the clay core.

Two variants without advection are through-diffusion and in-diffusion tests. In a through-diffusion experiment, the clay core is 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. The tracer quantity is measured at the outlet compartment as a function of time. It is recommended to measure the tracer concentration vs. time at the inlet as well. Traditionally, both inlet and outlet compartments are chosen sufficiently large, so that both the concentration decrease at the inlet and the concentration increase at the outlet are negligible. If during the measurements no tracer has been detected in the outlet compartment, a through-diffusion experiment can be considered as in-diffusion.

Apart from the just mentioned migration experiments, also combined glass-dissolution/diffusion experiments (Lemmens and Aertsens, 2006) have been performed. The set-up is similar to that of the percolation experiment (in this case without advection), but the source is a glass coupon, releasing ^{32}Si when slowly dissolving in the clay pore water. At the end, the mass loss of the glass (due to glass-dissolution) and the ^{32}Si profile in the clay are determined.

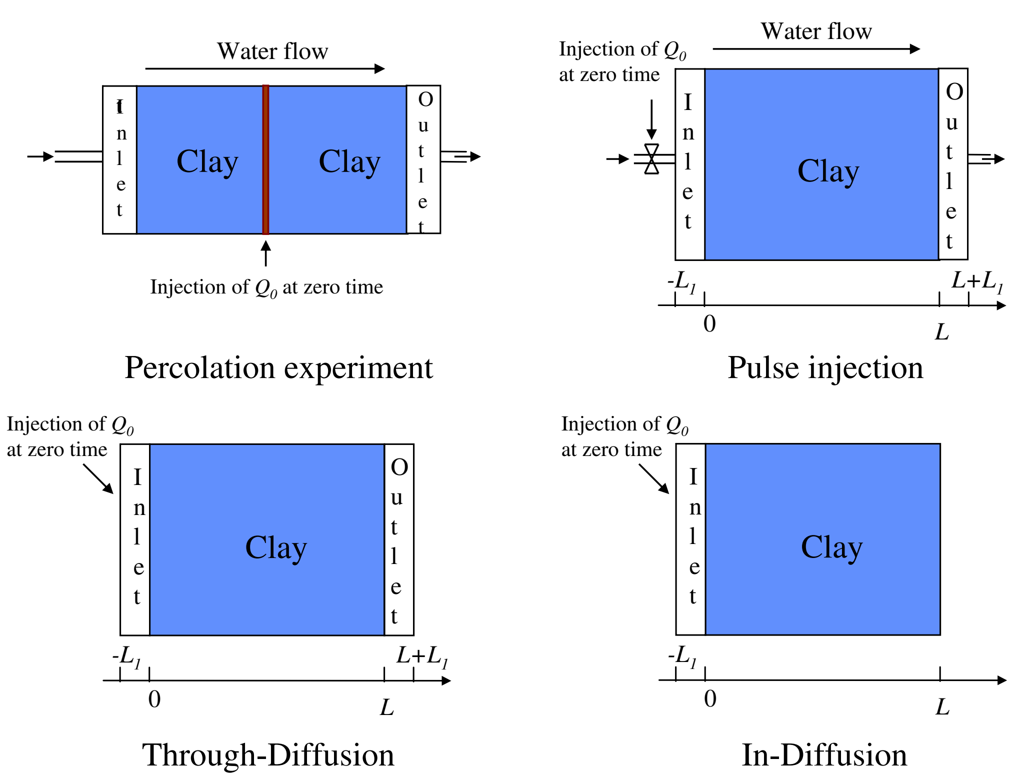

Fig. 1. Schematic overview of the different experimental set-ups in the migration tests.