ESTIMATION OF THE MIGRATION PARAMETERS FOR THE BOOM CLAY FORMATION BY PERCOLATION EXPERIMENTS ON UNDISTURBED CLAY CORES

MARTIN J. PUT*, MARCEL MONSECOEUR*, ALFONS FONTYNE**, HIDEKAZU YOSHIDA**
* SCK/CEN, Boeretang 200, 2400 Mol, Belgium
** PNC, 9-13 1-Chome, Akasaka, Minato-ku, Tokyo, Japan

ABSTRACT

The safety assessment of the repository for high level radioactive waste in the Boom clay formation requires reliable data for the migration parameters. The experimental set-up and the interpretation method is briefly described for percolation experiments on undisturbed clay cores drilled from the formation and results are reported. The undisturbed clay cores are drilled perpendicular and parallel to the stratification of the formation to study the isotropy of the formation. To represent the real situation as close as possible, in-situ interstitial clay pore water is used as percolating liquid.

Parameter values determined in the percolation experiments are presented for bromine, iodine and tritiated water. Darcy velocity and effective stress are used as variables. Anisotropy of the formation is demonstrated for the hydraulic conductivity, but is found to be trivial for the dispersion parameters. A general relation between the apparent dispersion constant, the diffusion accessible porosity and the Darcy velocity is also given.

INTRODUCTION

The migration characteristics of the Boom clay are studied within the Belgian research and development program for the disposal of radioactive waste in a deep geological formation.

Different types of migration experiments are performed on samples taken from the Boom clay formation during excavation for the construction of the underground research facility. One of the methods used for the study of the migration characteristics are percolation experiments with impulse injection on clay cores. To reproduce the in-situ conditions as nearly as possible, interstitial clay pore water is used as percolating liquid for the experiments. This water is collected through filters installed in the underground facility.

The reported work is a joint effort between the SCK/CEN (Belgium) and PNC (Japan).

DESCRIPTION OF THE EXPERIMENT

Clay cores are drilled horizontal and perpendicular to the bedding plane of the Boom clay formation during excavation of the underground facility constructed at the Mol site. They are immediately loaded in metal cans, sealed to prevent oxidation and subsequently transferred to an inert atmosphere glove-box. Two vertically and one horizontally drilled cores are mounted in stainless steel cells (with a cross sectional area of 11.34 cm²), sandwiched between sintered metal filters of 3 mm thickness, and transferred to a consolidation apparatus. One end face of the cylindrical samples is connected.
with a N₂-pressurized vessel containing porewater abstracted from a piezometer in the underground laboratory. After stabilization of the flowrate, a 20 µl tracer solution is injected via the loop of a high-pressure valve and allowed to percolate through the sample. The percolating porewater is collected in discrete samples at the effluent end of the cell until almost the entire injected activity is migrated through the clay cores.

A series of tests are successively run on the same clay cores without and with consolidation. The tracers used are Br-82 and a mixture of I-131 and HTO.

During the first experiment with Br-82, the tracer is injected at the top of the cells with a downwards water flow direction. This resulted in a rather irregular flowrate, probably due to the formation of gas bubbles. In all subsequent experiments the tracer is injected at the bottom of the cells with an upwards water flow direction. This resulted in a very stable flowrate.

MATHMATICS

The transport equation is solved for the appropriate boundary conditions in order to interpret the experiments.

The experimental results show that the influence of the dead volume $V_o$ (figure 1), composed of the volume of the distribution chamber at the inlet filter and the filter itself is not negligible. The experiment has been modelled with the dead volume taken into account.

Suppose the impulse with quantity $Q_o$ injected at time zero arrives at time $t_o$ at the inlet. The concentration $C_o(t)$ in the liquid in the dead volume $V_o$ is given as the solution of the mass balance equation (well-mixed liquid)

$$\frac{dC_o(t)}{dt} = -\lambda C_o(t)$$  \hspace{1cm} (1)

![Figure 1: Outline of the percolation experiment with impulse injection.](image)
The time constant of the dead volume $\lambda = S V_d / V_o$ is a parameter of the experimental set-up and is determined by the dead volume $V_o$, the cross sectional area $S$ of the clay core and the Darcy velocity $V_d$ in the core.

Solution of equation (1) with the initial conditions $C_o(t<t_o) = 0$ and $t = t-t_o$ is

$$C_o(t) = Q_o / V_o \exp (-\lambda t)$$  \hspace{1cm} (2)

$C_o(t)$ is the source term for the solution of the transport equation in the clay core, which may be written as [1] [2]

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2} - V \frac{\partial C}{\partial x}$$  \hspace{1cm} (3)

where $D$ is the apparent dispersion constant in the clay core and $V$ the apparent convection rate given by $V_d / \eta R$, with $\eta$ the diffusion accessible porosity and $R$ the retardation constant. The concentration $C$ in (3) is the concentration in the liquid accessible by diffusion.

The total quantity $Q(t)$ passed through the plane $x = L$ after time $t$ may be calculated as

$$Q(t) = \int_{-}^{L} \eta R C(x, t) \, dx$$  \hspace{1cm} (4)

The transport equation (3) may be solved by the Laplace transform method. The solution is [1]

$$Q(t) = 0.5 \, Q_o \left[ \text{erfc} (\alpha - \beta) + \exp (4\alpha \beta) \, \text{erfc} (\alpha + \beta) \right. $$  

$$- \exp (2\alpha (\beta - \phi) - \lambda t) \, \text{erfc} (\alpha - \phi) $$  

$$\left. - \exp (2\alpha (\beta + \phi) - \lambda t) \, \text{erfc} (\alpha + \phi) \right]$$  \hspace{1cm} (5)

with $\alpha = L / 2\sqrt{D \tau}$

$$\beta = V / 2\sqrt{D \tau}$$  

$$\phi = x\sqrt{V^2 - 4 D\lambda / 2\sqrt{D \tau}}$$

Equation (5) is used for the interpretation of the percolation experiments with impulse injection. A curve fitting program is written to fit the parameters $D$ and $V$ to the percolated quantity through the clay core. The value $\eta R$ then may be calculated as $\eta R = V_d / V$. 
The advantage of this type of percolation experiment with impulse injection over a pure diffusion experiment with impulse injection is the simultaneous determination of $D$ and $nR$. When the experiments are repeated at different values of the Darcy velocity $V_d$, the dispersion length $a$ may also be estimated.

DISCUSSION OF THE RESULTS

The results of the percolation experiments are given in Table I. The pore water pressure is given by $P_{pore}$ and the consolidation pressure by $P_{cons}$. Where the consolidation pressure is not given, the sample is only confined and the consolidation pressure is unknown. The hydraulic conductivity $K$ has been calculated as $K = V_d \cdot L/P_{pore}$.

An example of the percolated fraction of the considered species, given in % of the total injected quantity $Q_0$, as a function of time since injection, is given in Figure 2 for the experiments 15 and 21. The line through the experimental points is equation (5) fitted to the experiment by adjusting $V$ and $D$. A very good agreement between the experimental points and equation (5) is obtained. The results on the figure are from a simultaneous injection of iodine and HTO in the same clay core and show that iodine comes in advance of

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(*) $V$ means clay sample cored perpendicular to the stratification of the formation and $H$ means parallel.
Figure 2: Percolated quantity as a function of time for simultaneous injection of iodine and HTO.

Figure 3: Concentration peaks in the percolated liquid for the simultaneous injection of iodine and HTO.
tritiated water (HTO) despite the fact that both are non-retarded species with about the same mobility in pure water \((\sim 2 \times 10^{-5} \text{ cm}^2/\text{s})\). The phenomenon is even better illustrated in figure 3 which gives the concentration in each collected sample normalized to the total injected quantity \(Q_0\) as a function of time since injection. The figure shows clearly that iodine moves about twice as fast as HTO. This is in good agreement with the concept of the diffusion accessible porosity which is 0.20 for iodine (ion exclusion) and 0.46 for HTO for the considered experiment (see table I). The apparent convection velocity may be calculated as \(V = V_d/\eta R\) which gives a value of 6.2 \times 10^{-6} \text{ cm/s} for iodine and 2.7 \times 10^{-6} \text{ cm/s} for HTO for the example.

Table I reveals a systematic difference of the hydraulic conductivity \(K\) for horizontally and vertically cored clay cores. The hydraulic conductivity for flows parallel to the stratification of the formation is about double those for flow perpendicular to the stratification.

The dispersion constant \(D\) is mostly written as \(D = D_a + a |V|\), with \(D_a\) the molecular diffusion constant and "\(a\" the dispersion length. For Boom clay the molecular diffusion constant \(D_a\) obeys the Bruggeman equation [3]. Therefore the general relation

\[ \eta RD = D_a \eta^{1+k} + a |V_d| \]  

(6)

has been fitted on the results of the percolation experiments. The agreement is quite good. For the dispersion length "\(a\" a value of 0.1 cm is obtained and for \(D_a\) a value of 2.7 \times 10^{-6} \text{ cm}^2/s. A value of 1.5 is obtained for the exponent \(1+k\) suggesting that the Bruggeman equation is also valid for the diffusion accessible porosity \(\eta\) for percolation experiments in the Boom clay.

Equation (6) has no term on the right side that depends on the retardation constant \(R\), suggesting that it is also valid for retarded species, i.e. the dispersion conductivity constant \(\eta RD\) is independent of the retardation constant \(R\).

The influence of the stratification on the value of \(\eta R\) is trivial and the differences are insignificant. The influence on \(D\) is indirect and given by the influence on \(V_d\). For the dispersion length "\(a\" no influence of the stratification is revealed.

CONCLUSIONS

Percolation experiments with impulse injection have been done on undisturbed Boom clay cores that were drilled perpendicular or parallel to the stratification.

The experimental results revealed an anisotropy for the hydraulic conductivity that was within a factor of 2 for the main directions. The influence of the stratification on the other migration parameters \(\eta R\), \(D\) and "\(a\" is trivial. A general relation between the dispersion conductivity constant \(\eta RD\), the diffusion accessible porosity \(\eta\) and the Darcy velocity \(V_d\) is proposed. It is a useful relation and is inspiring for further research. An estimated value of 0.1 cm for the dispersion length for the Boom clay has been obtained. Experiments with simultaneous injection of iodine and HTO revealed the usefulness of the concept of the diffusion accessible porosity \(\eta\) and its influence on the percolation velocity.
REFERENCES

