Unsaturated and Saturated Flow in Swelling Clay

Courant Non-Saturé et Saturé dans Argile Gonflante

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SYNOPSIS Water uptake and migration in saturated, highly compacted bentonite as well as water flow after saturation, are largely dependent on physico/chemical and microstructural features. The rate of water uptake is a complex function of a number of variables but it can be predicted with sufficient accuracy for practical purposes by considering it as a diffusion process. Water flow in saturated dense bentonite is gradient-dependent which probably results from water viscosity anomalies. Tortuosity is largely responsible for the very obvious reduction in permeability when the bulk density approaches high values.

INTRODUCTION

Swedish work on finding a suitable technique to isolate unreprocessed nuclear reactor wastes from the biosphere has resulted in a proposal to use bentonite as one barrier (KBS, 1978). Metal canisters with waste material will be embedded in highly precompacted bentonite and stored in deposition holes bored from the floors of a number of tunnels at large depth in crystalline rock (Fig.1). Since the bentonite is not fully saturated at the deposition, the water uptake resulting from the access of ground water in the surrounding rock has the character of unsaturated flow. The rate of this uptake is of great practical significance since it affects the longevity of the canisters as well as the development of swelling pressures which affect the stress situation in the confining rock. When the saturation is completed, and when normal temperature conditions finally prevail, water flow through the bentonite is caused by regional hydraulic gradients only. Naturally, the rate of flow in the latter phase is equally important because of the long periods of time that have to be considered ($\geq 10^4$ years). This article presents some fundamental features of the unsaturated, and saturated flow phases.

DEFINITION OF CASE

The soil material is commercially available, granulated bentonite of type MX-80 (Na Wyoming bentonite). Such material is rich in montmorillonite (≥ 80%) and clay-sized particles (clay content 85-90%) although the industrial processing results in a rather coarse-grained character with an average aggregate size corresponding to the silt and sand fractions. The natural water content is of the order of 10%.

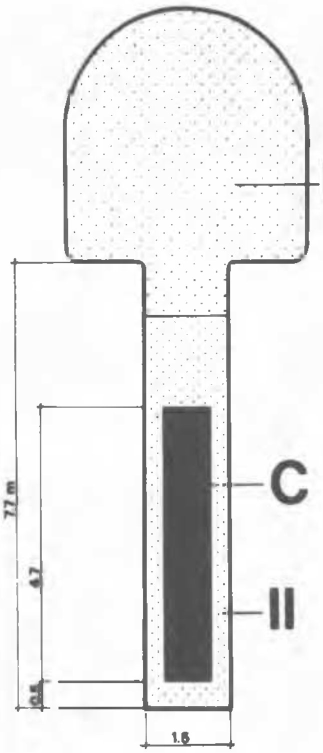


Fig.1. Schematic view of tunnel and deposition hole with canister for unreprocessed reactor wastes (C). I represents in situ-compacted sand/bentonite backfill, II represents closely fitting blocks of highly compacted bentonite.

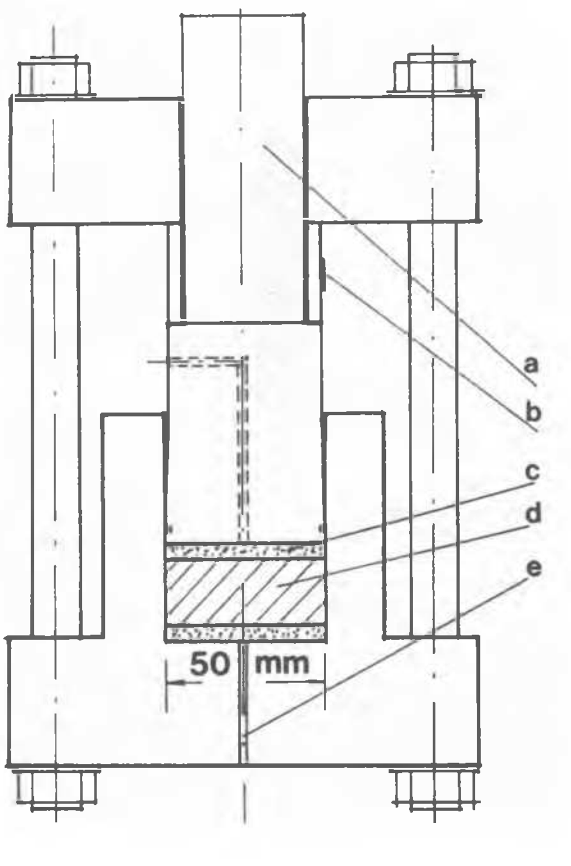
Precompaction under a pressure of the order of 50 MPa yields blocks with a bulk density of about 2.1 t/m^3 and approximately 50% water saturation. The density of confined samples increases to about 2.3 t/m^3 at water saturation, but in practice a certain amount of swelling cannot be avoided so that the final bulk density may be in the range of 1.9-2.2 t/m^3.

UNSATURATED FLOW

The main case in the study was uniaxial flow in confined samples. Unsaturated flow took place in the course of water uptake to yield complete saturation, while saturated flow was produced by applying an external gradient after the saturation.

The driving force of the unsaturated flow was primarily the affinity of the mineral surfaces to water and to osmotic forces, e.g. the hydration of surface-adsorbed ions.

Laboratory experiments at 20°C were made on samples compacted in oedometers used for careful determination of swelling pressures. Air-dry bentonite was filled in the oedometer (Fig. 2) and compacted to various densities. "Allard water", which is an artificial solution with a composition characteristic of deep ground water in Swedish crystalline rock (cf, KBS, 1978), was then let into the samples through one or both filter stones, while keeping them in fixed positions. The time required to obtain complete saturation was determined by measuring the swelling pressure at regular intervals. This procedure involved loading of the upper piston in a press, while recording the successively lowered compressive stress in the ring which locked the sample in position. The swelling force exerted by the sample on the lower piston, and thus on the ring, was accurately determined as the force required to make the ring stress-free.



While the initial water content of the unsaturated samples was practically the same in all the tests, the sample height and water inlet geometry were varied in a number of ways. This yielded a range of bulk densities; from 1.75 to 2.19 t/m^3 after water saturation. The state of complete saturation was reached when the swelling pressure became constant, so the determination of this pressure at regular intervals gave a very safe determination of the time required to obtain complete saturation.

The rate of the unsaturated flow, or rather water molecule migration was a function of the water content, which is known to govern the suction potential, and of the water-content gradients, as well as of the permeability. Disregarding the latter restriction and the possible influence of a number of physical effects (cf. "saturated flow"), the migration should have the character of a diffusion process governed by water-content gradients. Initially, the water content was approximately 10% throughout the sample while, eventually, a certain maximum value was reached in the entire, confined sample. In the case of bentonite initially compacted to approximately 2.1 t/m^3, this latter value was 20%.

The isotropic character of the highly compacted bentonite suggests a simple form of the diffusion equation:

$$\frac{\partial w}{\partial t} = D \frac{\partial}{\partial x}\left(\frac{\partial w}{\partial x}\right) \quad (1)$$

Where w = water content

x = distance from water inlet (one flow direction)

t = time

D = diffusion coefficient

Resonable agreement between theory and experiments was in fact obtained for $D = 10^{-9}\ m^2/s$. Thus, for 1 cm sample height and one water inlet, as well as for 2 cm sample height and two opposite inlets in the oedometer, an average water content of 19% - corresponding to 95% water saturation - was obtained experimentally as well as by applying Eq. (1) for $t \sim 5$ days.

The diffusion-type character of the migration was very clearly illustrated in an additional experiment with a large cylindrical sample of highly compacted bentonite, 30 cm in diameter and 130 cm in height, which was confined in a strong steel cylinder and exposed to "Allard water" at its base. 8 months after start, the test was stopped and the cylinder emptied and investigated with respect to the water content (Table 1). The water distribution, which tells that there is no actual "water front" proceeding through the mass, was found to be in reasonable agreement with predictions based on Eq. (1).

Fig.2. The LuH swelling pressure oedometer.
a) Free piston for loading the specimen for releasing the stress in the ring,
b) is the strain gauge glued to the ring,
c) filter stone of stainless steel,
d) bentonite specimen,
e) lower water inlet.

TABLE I

Water content of highly compacted Na bentonite (MX-80) in cylinder test

Water content interval, %	Distance from base surface, cm
10-12	23-30
12-14	19-23
14-16	16-19
16-18	12-16
>18	<12

SATURATED FLOW

Saturated flow under constant hydraulic gradients is characterized by the coefficient of permeability, which reflects the influence of various physical reactions, such as electrokinetic coupling, viscosity anomalies, tortuosity, and structural heterogeneity.

Laboratory experiments at 20°C were performed in which compacted samples were first saturated and then percolated under confined conditions. The compaction and saturation were made by using the swelling pressure oedometer as described previously. "Allard water" was again used for the saturation of the approximately 5 mm thick samples. After 3 days, which is known to be more than sufficient to obtain complete water saturation, the percolation was started. For this purpose the lower filter stone was connected to a cylindrical vessel in which de-aired "Allard water" was compressed by using a piston operated by means of high air pressure. This device has been frequently used for producing back pressures in triaxial tests of various soils and is known to transfer the pressure to the liquid with practically no loss through friction or leakage. In most tests, the air pressure was varied systematically in the order 500 kPa, 250 kPa, 100 kPa, and 50 kPa to find out whether the hydraulic gradient affects the permeability or not. Each resulting gradient (10^4, $5\cdot10^3$, $2\cdot10^3$, and 10^3, respectively) was held constant until stationary flow conditions were approached. The total testing time required for each sample was therefore rather long, 3-4 weeks as an average.

The percolated water quantities were extremely small as a consequence of the very low permeability, and the flow rate was therefore determined by observing the rate of displacement of a water meniscus in a calibrated capillary connected to the filter stone at the water exit. The accuracy of the evaluated permeability was estimated at approximately $5\cdot10^{-15}$ m/s. The technique is similar to the one used by Hansbo (1960).

While the air-dry bentonite powders had an initial water content of approximately 10% in all the tests, the saturated samples were given different bulk densities and water contents in order to investigate the relationship between these properties and the permeability.

The evaluated permeability was found to be a function of the hydraulic gradient as illustrated by the example in Fig.3. This confirms the observations of Hansbo and others that the rate of water flow through fine-grained clays often deviates from Darcy' s law. The trend is obvious as shown by Table 2.

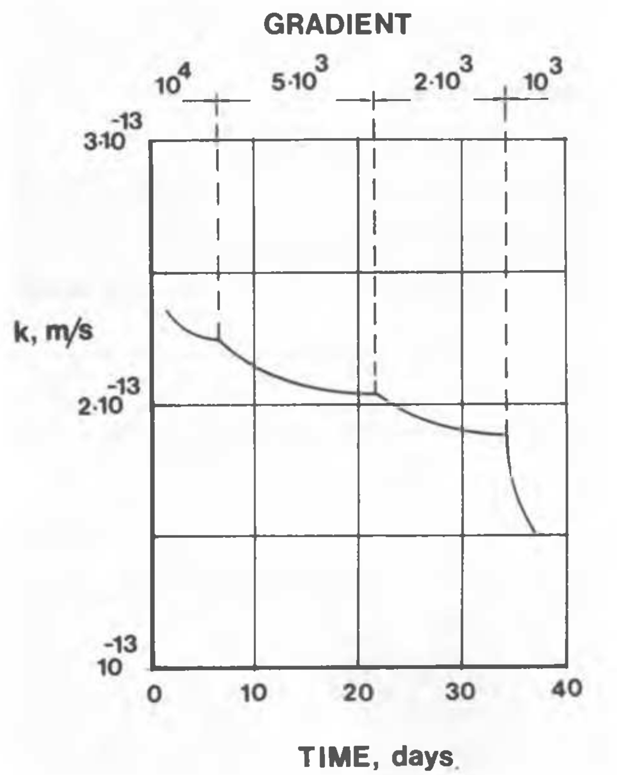


Fig 3. Example of permeability test with smoothened curve for the coefficient of permeability k versus time after the test start.

These observations suggest a physical model which requires the structural constitution to be considered. The microstructure of granulated bentonite powders, compacted at their natural water contents, can be illustrated by Fig.4. In the course of water saturation, the aggregates expand and, if given sufficient time, most of the larger interparticle voids will be filled by a clay gel formed by the expanding aggregates. Eventually, a condition of considerable isotropy and homogeneity with respect to particle orientation and interparticle distance will be reached at moderate and high densities. The average particle spacing, without special reference to inter- and intraparticle (lamellar) distance, is schematically illustrated by Fig.5.

TABLE 2

The coefficient of permeability, k, evaluated from the percolation tests

Test No.	Bulk density ρ_m, t/m^3	Water content %	Porosity n,%	Gradient i	k m/s
1	1.90	~35	~49	10^4	$2.2\cdot10^{-13}$
				$5\cdot10^3$	$2.0\cdot10^{-13}$
				$2\cdot10^3$	$1.9\cdot10^{-13}$
				10^3	$1.3\cdot10^{-13}$
2	1.95	~31	~46	10^4	$2.3\cdot10^{-13}$
				$5\cdot10^3$	$2.0\cdot10^{-13}$
				$2\cdot10^3$	$1.9\cdot10^{-13}$
				10^3	$1.5\cdot10^{-13}$
3	1.97	~31	~45	10^4	$1.2\cdot10^{-13}$
				$5\cdot10^3$	$1.0\cdot10^{-13}$
				$2\cdot10^3$	$9\cdot10^{-14}$
				10^3	$6\cdot10^{-14}$
4	2.15	~19	~34	10^4	$2\cdot10^{-14}$
				$2\cdot10^3$	$\sim2\cdot10^{-14}$

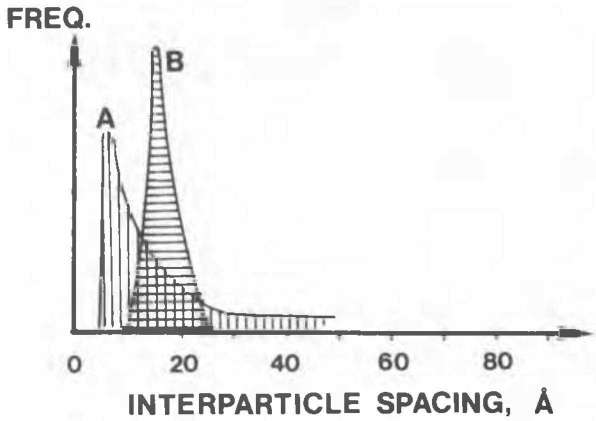


Fig 5. Schematic distributions of the interparticle spacing. A) original spectrum for "air-dry" bentonite, B) narrow peak for homogeneous, swollen sample with high density.

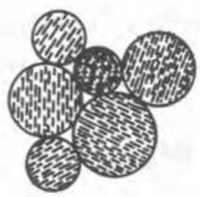


Fig.4.
Schematic picture of the particle arrangement in the granulated, "air-dry" bentonite.

The rates of water uptake and saturation depend on the sample size, while the processes leading to a final structural equilibrium with a high degree of homogeneity are not functions of this size. The latter require weeks or months to be completed and this may have produced a slight successive reduction of the average width of the flow passages in the tests, and of the permeability as well. The effect is minor, and the observed permeability drop at decreasing hydraulic gradients requires some other physical explanation, such as:

1. Electrokinetic coupling, e.g. the production of an opposite osmotic flow when a liquid is forced through clay.

2. The high viscosity of surface-near water makes it flow slower than surface-distant water.

3. A heterogeneous microstructure of the aggregated type implies that a number of particles at the periphery of the aggregates are very weakly bound and therefore free to move even at very low flow rates of the pore water.

The first effect can be assumed to yield a successive retardation of the flow rate. It is fairly unimportant compared with some of the other effects (Olsen, 1961).

The second effect implies that the surface-near water molecules are rather strongly bond to the crystal lattice, a practical consequence being that such water does not start flowing until the activation energy for bond rupture is exceeded. This yields a gradient-dependent, non-Darcy behaviour. The matter of water molecule bonding to clay lattices and the viscosity of "vicinal water", has been discussed for many years and completely different opinions have been expressed by various investigators. Recent analyses of creep (Pusch & Feltham, 1980), indicate that very thin inter- and intralamellar water lattices are firmly attached to the crystal lattice and that they possess a certain strength. The influence on permeability, in terms of lower flow rates at low gradients, should naturally be most obvious for dense clays.

The third effect should be insignificant in the final state of microstructural homogeneity, at least at high bulk densities. At lower densities than the ones of interest in this study, as well as in freshly compacted dense granulated bentonites, small particles may easily be transported through void passages by flowing pore water also at very low gradients.

Summing up the various contributions it can be concluded that gradient-dependence of the permeability at the investigated high bulk densities probably originates from different viscosity properties of mineral-adsorbed and free water.

The strong impact of increased densities on the permeability, as illustrated by Fig.6., is due to the largely reduced widths of the passages, and to viscosity anomalies, as well as to tortuosity, which is a measure of the actual flow path length in the structural network.

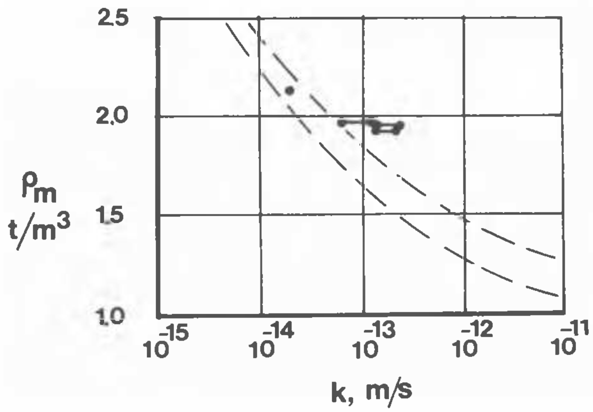


Fig 6. The coefficient of permeability versus bulk density. Where two values are plotted for one and the same density, the left represents $i=10^3$, and the right $i=10^4$. Broken curves are upper and lower boundaries of literature-derived values.

Tortuosity is conveniently expressed by means of the model in Fig.7, which shows a soil consisting of equally spaced, anisometric particles arranged in a brick-like way. The angle, Θ, between the principal axes of the particles and the direction of permeation is assumed tc correspond to the average degree of particle orientation. Starting from the Darcy and Kozeny Carman equations, Olsen (1961) derived a mathematical expression of the tortuosity T of flow which yields approximately twice as high T:s for n = 20 than for n = 80%, where n is the porosity. This ratio is representative of soil systems with the rather extreme anisometric particle shape, which is characteristic of montmorillonite. It suggests that tortuosity is an important flow-retarding factor at high bulk densities.

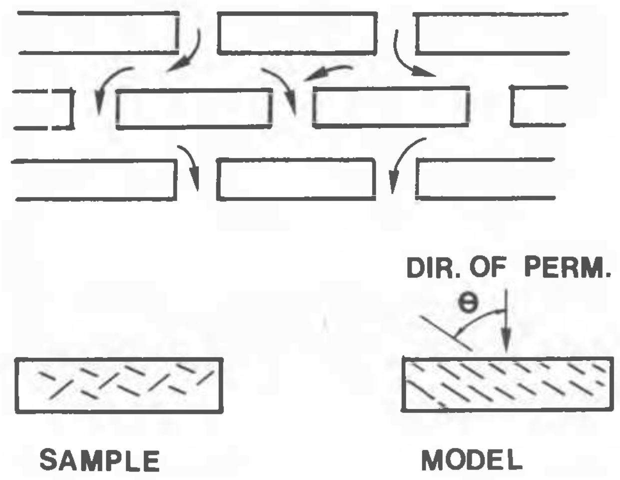


Fig 7. Olsen's physical model for the derivation of a mathematical expression of tortuosity.

CONCLUSIONS

The water uptake and migration in unsaturated highly compacted bentonite as well as the water flow after saturation are largely dependent on physico/chemical and microstructural features. Although the rate of water uptake is a complex function of a number of variables, such as tortuosity and permeability, it can be roughly predicted by simulating the process to be one of diffusion. Flow under saturated conditions is largely affected by the bulk density. Gradient-dependence is obvious at high as well as low gradients. At high densities, viscosity anomalies of mineral-adsorbed water may contribute to yield a low and gradient-dependent permeability. At low gradients this results in practically impervious conditions. Thus, with a regional gradient of 10^{-2} and a permeability of 10^{-13} m/s, the flow rate will not be higher than approximately 1 mm in 30 000 years.

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