Numerical Calculation of the Rate and Distribution of Porewater Flow in Clay used for Isolating Hazardous Waste

R. Pusch¹, S. Knutsson¹, L. Xiaodong² and R. Prikryl³

¹Luleå Technical University, Luleå, Sweden

²East China Institute of Technology, Fuzhuo, Jiangxi, China

³Charles University, Prague, Czech Republic

Abstract

The hydraulic performance of clay barriers for isolating hazardous waste products depends on the experimentally determined hydraulic conductivity. While the large majority of a cross section of dense smectite clay is nearly impermeable because of the immobility of the sorbed porewater, there are narrow, tortuous channels that are more or less continuous. They contain less dense clay gels that let water through under hydraulic gradients. Applying the 3Dchan code one finds fair agreement between theoretical and experimental hydraulic conductivities for smectite-rich clay if hydraulic gradients of 30-50 are used, while flow experiments using very high gradients give much lower conductivities. The discrepancy can be explained by the high flow rate in the channels by which particles are moved, causing clogging. The process is illustrated by using FEM analysis of the flow rate distribution and by applying Computational Fluid Dynamics (CFD).

Keywords: clay, erosion, flow, hydraulic conductivity, microstructure, porewater, smectite.

1 Scope of study

Water transport in soils is an issue of fundamental importance to earth dam construction and foundation engineering. Dams leak and concentrated flow may cause erosion, piping and malfunction. In recent time the matter of effective isolation of radioactive waste by use of low-permeable smectite clay has become an important task and the hydraulic conductivity of such clays is presently in focus of the organizations that are responsible for final disposal of waste emanating from the nuclear industry. The common way to determine the conductivity is to run oedometer tests for measuring the through-flow and evaluate the conductivity by applying Darcy's law. The very long time needed for reaching steady state flow has led laboratory staffs to apply very high hydraulic gradients, which turns out to have

an impact on the accuracy of the evaluated conductivity. This is because high flow rates have an impact on the microstructural constitution of the percolated clay by causing erosion and clogging.

Compression of the entire test sample can also take place and the processes mentioned can lead to underestimation of the evaluated hydraulic conductivity by more than two orders of magnitude.

2 Hydraulic conductivity of clay in a repository

2.1 Basic

The most important waste-isolating property of clay barriers is the hydraulic conductivity. It should be lower than that of the surrounding rock, which means that smectite clay with appreciable density is a favourite candidate. The particles of such clays consist of stacked lamellae with highly hydrophilic potential [1]. A large part of the porewater in smectite is in interlamellar positions and effectively bound to the crystal lattices. Because of the hydrophilic nature smectite has strong expandability and determination of the hydraulic conductivity K must be made under confined conditions using oedometers as shown in Figure 1. In general, data deduction, from tests with the oedometer, uses the Darcy's model $v = Q/A = Ki$, where Q represents the quantity of fluid Q flowing through the sample in a unit time period. Since the cross-sectional area A of the sample is known, the average permeating velocity $v = Q/A$ for the unit time period, can be related to the test-specific gradient $i = h/H$.

Figure 1. Schematic section of oedometer for determination of the hydraulic conductivity. The thickness of the sample is h , and the water pressure difference across it is H . The through-flow is Q .

The present techniques for determination of hydraulic conductivity provide one with a macroscopic measure of how permeable the soil is. One needs to bear in mind that the value of K is not measured but calculated from the flux-compliance. It follows that the test procedure and the data reduction technique do not determine the role of variations in pore sizes and interconnectivity of pore spaces. Whilst this may not be too important for coarse-grained soils and clays composed of non-expanding minerals, it is significantly important for smectites because of a number of practical implications that impact directly on the flow rate through the smectite clay.

The conductivity of smectite clay is determined by the volume fraction and continuity of the small permeable parts of the microstructure, and hence, the microstructural constitution is an important factor. Temperature is also an important factor since it affects the viscosity of the water. Determining the hydraulic conductivity of swelling clays is in fact far from straight forward because of a number of complicating facts:

- in natural and artificially prepared clay samples the presence of gas bubbles can affect percolation. The evaluated conductivity can be underestimated by bubbles occupying wider voids and channels, resulting in hindrance of the through-flow of water. Since the wider voids and channels are the most pervious parts of the void system, gas bubbles may exercise a significant role in hydraulic conductivity testing and evaluation.
- water can migrate by diffusion - Knudsen flow [2] - provided that there is a driving force, which can be a gradient in hydration potential, as manifested by differences in water content on the microstructural scale.
- the percolation rate can affect the stability of microstructures through erosion and particle transport if the percolation rate is high – caused by high hydraulic gradients.
- very high hydraulic gradients can in fact cause compression of the entire clay sample by which channels tend to be compressed and the evaluated conductivity to be too low.
- generation of electrical potentials by percolating water will tend to counteract flow and yield too low a hydraulic conductivity.

2.2 The microstructural constitution

We will use here the 3Dchan model [3] for predicting the hydraulic conductivity in 3D, simulating the actual variation in density that can be derived from microstructural analyses [4]. The major features of the model is the assumed network of channels such as the one shown in Figure 2, the size distribution of their cross section being statistically normal and the number being determined by the porosity of the clay. Assuming circular cross sections of the channels and using the Hagen-Poiseuille law, the flow rate and flux through the channel network can be calculated for given boundary conditions [5].

Figure 2. Channel network mapped as a cubic grid with channels intersecting at a node in the grid [3,4].

The channels contain numbers of bundles of capillaries with a diameter that match the porosity of the clay. After complete grain expansion, the voids filled with clay gels are assumed to have a normal size distribution with the following intervals for three reference clays, i.e. A) 1-5 μm for clay with 2130 kg/m^3 density at water saturation, B) 1-20 μm for clay with 1850 kg/m^3 density, and C) 1-50 μm for clay with 1570 kg/m^3 density. Table 1 gives the density, total porosity n , and resulting bulk hydraulic conductivity of the considered clay types, using the 3Dchan code. It also provides experimental data from oedometer tests with the high hydraulic gradients 3000-10000 meter water head per meter flow length (m/m) that are used by many investigators.

Reference MX-80 clay	Dry density, kg/m^3	Density at Saturation, kg m^3	n	Calculated K , m/s	Experiment K , m/s*
A	1800	2130	0.01	3E-12	2E-14
B	1350	1850	0.20	1.3E-11	3E-12
C	900	1570	0.47	2.4E-10	8E-11

* Hydraulic gradients 3000-10000 m/m (Clay Technology AB, Lund)

Table 1. Hydraulic conductivity (K) of three clay types prepared by compacting MX-80 powder and saturating them with electrolyte-poor water. n is the porosity.

One concludes that the model gives conductivity values that are higher than measured ones for hydraulic gradients ranging between 3000 and 10000. For a typical density, i.e. 2130 kg/m^3 , the experimental value is about 100 times higher than the calculated while the difference is much smaller for the softest clay. We will see that the discrepancy can be explained by underestimation of experimentally determined hydraulic conductivity for high hydraulic gradients.

2.3 Flow paths

The fact that a large part of the porewater in smectite clay is immobile [1] suggests that the flow rate in permeable parts at hydraulic testing can be considerable and

cause erosion. By using the theoretical microstructural model that gave the porosities in Table 1 and applying the Hagen-Poiseuille law for calculating the hydraulic performance of the flow channels one can estimate their frequency. The result is shown in Table 2 and visualized in the form of flow rate distribution in Figure 3 for a $250 \times 250 \mu\text{m}^2$ cross section, taken as relevant element area (REA), [5].

Reference MX-80 clay	Bulk density, kg/m^3	Number of 20-50 μm channels	Number of 5-20 μm channels	Number of 1-5 μm channels	Average ratio of channel cross section area and total REV area, %
A	2130	0	0	135	1.06
B	1850	0	10	385	4.8
C	1570	2	85	950	14.4

Table 2. Number of differently sized channels per $250 \times 250 \mu\text{m}^2$ cross section area of the reference smectite clay (MX-80).

Figure 3. Distribution of flow rates in $250 \times 250 \mu\text{m}^2$ cross sections of smectite clays with different densities. A) Clay with 2130 kg/m^3 density at saturation. B) Clay with 1850 kg/m^3 density at saturation, C) Clay with 1570 kg/m^3 density at saturation [5].

The actual distribution of channels representing flow paths has been estimated by examining samples of smectite-rich clay (MX-80) representing type A percolated for 10 days by 3.5% CaCl_2 solution to which Methylene Blue solution with 2 % concentration had been added. The outermost millimetre from the inflow side was cut off from the 30 mm diameter samples having 15 mm original height, and the rest dried for 24 hours at 105°C after which photographs were taken. The blue spots were of different size because of the variation in cross section of the channels, the size also being affected by diffusive migration of the organic dye molecules from them (Figure 4).

Figure 4. Micrograph of a major blue spot. The dye had diffused from the channel with an estimated diameter of a few micrometers and coloured the light-grey clay blue.

Digitalization of photographs of oedometer-tested samples after drying has given detailed information of the frequency and size of percolated channels by examining identified blue spots. The clays had densities resembling those of the reference clays A, B and C and are represented by Figures 5, 6 and 7, in which the automatically evaluated accumulated cross section areas of blue spots are also given. The percentages vary from about 1.75 to 3.3 %. These are in reasonable agreement with the theoretical values in Table 2 for the two densest clays but far less than the value for Clay C because the density at water saturation of this clay, 1720 kg/m^3 , was higher than the density 1570 kg/m^3 of the theoretical C-model clay. The blue spots are larger than the actual channel width because of the diffusive migration of the tracer, and the actual percentage of the permeable part can hence be less than the figures given in the respective picture.

Figure 5. Distribution of permeated channels in the dried MX-80 sample with density 1950 kg/m^3 at water saturation as indicated by the blue spots. The diameter of the sample is about 30 mm. The percentage of blue spots of the total cross section is 1.75 %. The white objects are desiccation fractures.

Figure 6. Distribution of permeated channels in the dried MX-80 sample with density 1820 kg/m^3 at water saturation. The diameter of the sample is about 30 mm. The percentage of blue spots of the total cross section is 2.66 %. The white objects are desiccation fractures.

Figure 7. Distribution of permeated channels in the dried MX-80 sample with density 1720 kg/m^3 at water saturation. The diameter of the sample is about 30 mm. The percentage of blue spots of the total cross section is 3.31 %. The white objects are desiccation fractures.

Referring to the graph in Figure 8, which is based on the data in Table 2, the experimentally determined ratio of the channel cross section area and the total sample area is of the same order of magnitude as the theoretically derived ratio - except for the softest clay C. For this clay, the experimental ratio is only about 50 % of the theoretical value. One finds from the digital maps that the distribution of the channels was relatively uniform and that leakage along the clay/cell contact was negligible.

Figure 8. Theoretical percentage of channel cross section area of total REA area.

2.4 Laboratory technique

The risk of internal erosion has caused considerable debate on what hydraulic gradients can be applied without affecting the microstructural constitution. Testing Standards Organizations, such as American Society of Testing Materials (ASTM), recommend a maximum gradient of 30 m/m. However, the desire to obtain results in one day or a few hours has led investigators to use much higher hydraulic gradients. Values representing steady state flow can be obtained in less than one hour making it possible to perform large series of measurements in limited time. However, high gradients produce high flow rates and observation of percolated clay using optical microscopy and numerical calculations have indicated that erosion of channels in fully water-saturated smectite clay can move particles to constrictions in the channels resulting in void-clogging and reduction of the evaluated bulk hydraulic conductivity [6,7]. The conclusion from these investigations is that the flow rate required for tearing off 0.5 μm particle aggregates is E-3 m/s, E-4 m/s for 1 μm aggregates, E-5 m/s for 10 μm aggregates, and E-7 for aggregates larger than 50 μm . The reason for the size dependence is that small particle aggregates have a larger number of cohesive bonds than bigger aggregates. The small ones hence require higher flow rates for being released and transported.

Using Darcy's law the average flow rate at percolation of water-saturated smectite-rich clay with a density of about 2000 kg/m^3 and a hydraulic conductivity E-13 m/s evaluated from oedometer tests, is about 1.5E-9 m/s for a hydraulic gradient of 10000 m/m, which would not cause erosion. However, considering that the permeable fraction of such clay is only about 2 % of the total section as illustrated by the microstructural model data in Table 2 and the section in Figure 5, the channel flow rate is about E-7 m/s, implying a small risk of release and migration of particle aggregates. Now, assuming instead a bulk hydraulic conductivity of E-11 m/s, the high gradient would give a channel flow rate of about E-5 m/s for a 2 % permeable cross section and hence an obvious risk of erosion and clogging that may well lead to an apparent bulk conductivity of E-13 m/s or less. The risk of erosion and clogging of permeable channels may in fact be even higher since the fraction of major flow paths is probably lower than 2 %.

The risk of underestimation of the hydraulic conductivity by using high hydraulic gradients has been validated by experiments as shown in Figure 9. It demonstrates that the evaluated hydraulic conductivity from tests performed with a hydraulic gradient of nearly 9000 m/m, is about 1% of that with the gradient 40. The conclusion is that ASTM's recommendation to use low hydraulic gradients in percolation tests for evaluating the hydraulic conductivity of clays should be followed.

The higher hydraulic conductivity of clay saturated with salt in comparison with salt-free porewater of the same density testifies to the importance of the microstructural control. For low-electrolyte concentrations in the porewater, the channels constituted by voids are filled with soft clay gels. With salt water, gel

coagulation and collapse occurs, which can make particle aggregates, driven by flowing water, clog the flow channels, which are wider than in clays saturated with and percolated by low-electrolyte water [1].

Figure 9. Change in evaluated hydraulic conductivity of MX-80 clay in oedometer percolation tests by changing the hydraulic gradient. The first 250 hours involved water saturation and recording of through-flow under a hydraulic gradient of 40 m/m. The period from 250 to 255 hours represented percolation under the gradient 9000 m/m, again followed by percolation under the gradient 40 m/m [8].

3 Numerical calculation of flow on the microstructural level

3.1 Finegrained smectite clay

The flow pattern in a water saturated element of smectite clay with the density 1600 kg/m³ at complete saturation with low-electrolyte water has been simulated by FEM technique in 2D for the gradient 1 m/m taking the 30x30 μm² clay element of smectite clay to have a typical microstructural constitution [1]. The outcome of the study is illustrated by Figure 10, from which one concludes that the flow rate in larger voids is of the order of 2E-8 m/s [9]. It would hence be about 2E-6 m/s for a gradient of 100 m/m, still causing no damage to the microstructure. However, a gradient of 10000 m/m would raise the flow rate to 2E-4 m/s, causing disruption of soft clay gels and migration of small and large particle aggregates causing clogging

of voids in the channels and risk of significant underestimation of the hydraulic conductivity in laboratory tests.

Figure 10. FEM calculation in 2D of flow through $30 \times 30 \mu\text{m}^2$ clay element of smectitic clay with 1600 kg/m^3 density. For a hydraulic gradient of 100 m/m the maximum flow rate is $2\text{E-}6 \text{ m/s}$ [9].

3.2 Numerical study of channel flow in fine-porous medium

As indicated by the just mentioned FEM study and the percolation experiment with different hydraulic gradients, the ambient flow conditions may change the evaluated hydraulic conductivity significantly, due to different flow conditions, characterized as laminar, inertia-dominated, and turbulent flow regimes. This is mainly because the inertia forces can become significant for the flow inside the voids of the media and, hence, affect the overall parameters with increasing Reynolds number that is directly correlated to the applied gradient of the experimental and/or computational setup via equations like Darcy's law.

These matters and the effect on material parameters have been investigated by applying a method implying combination of Computational Fluid Dynamics (CFD) and Monte-Carlo simulations [10]. The CFD-simulations were performed on small well defined structures, both 2D and 3D, and for a wide range of Reynolds numbers and porosities [11, 12, 13, 14], while the Monte-Carlo simulations were conducted on larger systems of particles with well or poorly ordered material configuration [15, 16]. It follows from these simulations and defined different flow regimes that one can determine under what flow conditions that an experiment should be conducted in order to avoid inertia-dominated or turbulent regimes. When high gradients are

applied to a porous particle system that is viable to deform under increasingly higher gradients, the risk of potential damage increases quite rapidly since flow-induced normal and shear forces increase with the Reynolds number. With high gradients there can be a risk of build-up of pressurized zones implying changes of the evaluated hydraulic conductivity with the applied gradient. An increase in Reynolds number generates higher shear and normal forces, which can deform the particle aggregates and cause local sealing. Figure 11 represents one of the CFD-simulations with a Reynolds number of 45 (particle Reynolds number), the vectors indicate the velocity distribution, the length corresponds to the magnitude of the velocity, and the contours on the lower particle indicate the pressure, the darker grey the lower pressure.

Figure 11. Velocity vectors and contour pressure plot, Reynolds number 45.

4 Discussion and conclusions

Natural expansive clays as well as those prepared by compacting granules of expansive clay minerals have a heterogeneous microstructure. While the large majority of a cross section is nearly impermeable because of the immobility of the porewater making up the interlamellar hydrates, there are narrow, tortuous channels that are more or less continuous. The channels contain less dense clay gels that let water through under hydraulic gradients. Applying the 3Dchan code one finds fair agreement between derived and experimentally determined hydraulic conductivities for smectite-rich clay if hydraulic gradients of less than 100 m/m are used, while flow experiments with high gradients give much lower conductivities than those obtained by using the code. The discrepancy can be explained by the high flow rate in the channels by which particles are torn off and transported to constrictions where they become accumulated and cause clogging. This can give too low values of the hydraulic conductivity evaluated from oedometer tests with very high hydraulic gradients as illustrated by the FEM analysis of the flow rate distribution. Use of

Computational Fluid Dynamics (CFD) and Monte-Carlo simulations supports this conclusion.

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