Pollutant Migration through Clay

D.W. Airey
M.A., Ph.D.
Lecturer, University of Sydney

SUMMARY Tests have been performed on a remoulded clay to establish the variation of its hydraulic conductivity with stress history and void ratio. The hydraulic conductivity has been determined by both direct and indirect methods, and a comparison of these methods is presented. Finally, a pollutant, kerosene, was forced through the samples and the hydraulic conductivity determined. No flow of the kerosene was detected until a breakthrough pressure, dependent on the void ratio, was exceeded. Thereafter, the hydraulic conductivity was almost linearly related to the flow rate.

1. INTRODUCTION

Clayey soil barriers are widely used to contain water, chemicals and potentially harmful pollutants from municipal and toxic waste facilities because of their low hydraulic conductivities. However, their suitability for this purpose is the subject of increasing research as it has been demonstrated (eg. Mesri and Olsen, 1971, Fernandez and Quigley, 1985) that dramatic increases in hydraulic conductivity are possible when clayey soils are mixed with some pollutants, and in particular with liquid hydrocarbons. These increases occur because the thickness of the diffuse double layer that surrounds the clay particles is reduced, a consequence of the relatively low dielectric constants of the organic chemicals compared with water, effectively increasing the pore space between the particles. The reduction in double layer thickness can also be accompanied by volume reduction so that there is a possibility that cracks and preferential flow paths may develop in the soil. The risks of chemical processes affecting the integrity of clay barriers has led in many parts of the world to their prohibition as the principal barrier for containing toxic wastes.

Tests conducted in different apparatus to investigate the effects of liquid hydrocarbons on the hydraulic conductivity have given apparently conflicting results (Foreman and Daniel, 1986); in a rigid walled permeameter the hydraulic conductivity increased 1000 times when heptane, an immiscible liquid hydrocarbon, replaced water as the permeant, whereas in a flexible wall permeameter no flow was observed up to hydraulic gradients of 300. As neither apparatus represents the field conditions exactly there has been considerable debate as to which type of permeameter apparatus should be used to investigate the chemical effects. The tests described in this paper have been performed in an hydraulic oedometer apparatus. Apparatus of this type have been concluded by some authors (eg. Mitchell and Madsen, 1987) to be the most flexible and useful for estimating the chemical effects on the soil structure.

This paper describes the results of a series of tests to investigate the hydraulic conductivity of a natural clay from Sydney. A comparison is made of direct and indirect determinations of the hydraulic conductivity, using water as the permeant, from conventional and hydraulic oedometer cells. In addition the hydraulic oedometer has been used to determine the hydraulic conductivity of the soil using kerosene as the permeant.

2. NOTATION

In this paper the constant, k, relating the average flow velocity, v, to the hydraulic gradient, i, in Darcy's law

$$v = k\, i$$

will be referred to as the hydraulic conductivity. This constant is dependent on both the density, γ, and the viscosity, μ, of the flowing fluid. The intrinsic permeability, K, where

$$K = k\, \mu/\gamma$$

is independent of the fluid properties, that is K is a property of the porous medium alone. In this paper K will be referred to as the permeability of the soil.

3. SOIL AND FLUID PROPERTIES

Only remoulded soil samples have been used in the tests described below. The natural soil was first air dried and pulverised before being passed, dry, through a 425 micron sieve. Only the soil (about 75% of the total) that passed through this sieve has been used in the tests and in determining the soil properties described below. It is believed that if the soil had been wet sieved more than 90% would have passed 425 microns. The particle size distribution, obtained using a laser particle size analyser, is shown in Figure 1. It can be seen that the largest particle size was only 20 microns. X-ray diffraction analysis indicated that the soil was composed predominantly of kaolin, with approximately 20% quartz, and trace amounts of montmorillionite and illite. When dried the soil had a ubiquitous brown stain that is believed to be amorphous iron oxi-hydrate. This iron compound was precipitated on exposure to air during drying, as when the natural soil was freshly cut it had a greyish colour.

The soil had the following properties: Liquid Limit = 53, Plastic Limit = 26 and Plasticity Index = 27.

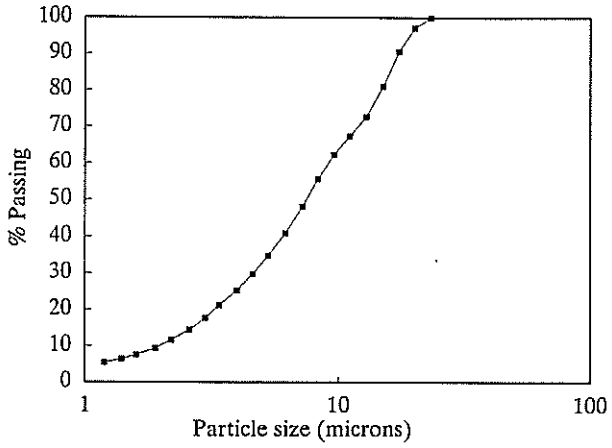

Figure 1. Particle size distribution

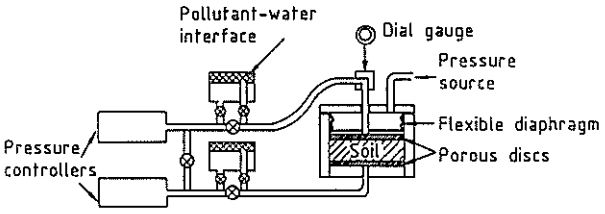

Figure 2. Hydraulic oedometer apparatus

The kerosene had the following properties: Specific gravity approximately 0.8, and Viscosity = 3.6 centipoise.

4. APPARATUS

Two types of oedometer have been used in these experiments:
(a) a small conventional oedometer cell with diameter 34 mm and initial sample height 12 mm. In this oedometer no control of the drainage was possible and only load and settlement readings could be obtained.
(b) an hydraulic oedometer with sample dimensions 75 mm diameter by 19 mm high. In this oedometer a back pressure could be applied to the sample and direct measurements of the hydraulic conductivity could be obtained. Two GDS pressure controllers (Menzies, 1988) were used to provide the back pressure and measure the volume changes during consolidation and hydraulic conductivity testing. The arrangement of the apparatus is shown in Figure 2. The settlement was measured by a dial gauge, and the vertical stress was provided by a separate pressure source. Also shown in Figure 2 are two pollutant-water interfaces that were used only when pumping kerosene through the soil. In each interface a layer of the less dense kerosene was placed above a layer of water. As the two fluids were immiscible this arrangement prevented any kerosene from entering the pressure controllers. Attempts to use membranes between the water and the kerosene were unsuccessful because the kerosene rapidly attacked the rubber compounds used and resulted in leaks from the interfaces.

5. EXPERIMENTAL PROCEDURE

The air dried and pulverised soil passing the 425 μm sieve was mixed with distilled de-aired water at a moisture content close to the liquid limit, and placed in a compaction mould. A vertical stress was applied, and increased in stages up to a maximum of 40 kN/m^2 for the hydraulic oedometer samples and 96 kN/m^2 for the conventional oedometer samples. Samples were removed from the compaction moulds and trimmed to fit the required oedometer.

For the conventional oedometer the vertical stress was increased in stages up to 580 kN/m^2 and then reduced to zero. For each stage the variation of settlement with time was recorded.

For the hydraulic oedometer a small vertical stress of 20 kN/m^2 was applied and then, keeping the vertical effective stress constant, the back pressure was increased to approximately 500 kN/m^2 to ensure full saturation. The vertical effective stress was increased in stages (see Table 1), with for each stage the changes of settlement and volume change with time recorded. For each stage after consolidation was complete, water was pumped through the samples to directly determine the hydraulic conductivity. The final operation in the first three tests was to pump kerosene through the soil. In test AR2 kerosene was pumped through the soil after the fifth stage at an effective vertical stress of 50 kN/m^2. This was followed by consolidation to a vertical effective stress of 400 kN/m^2 and further kerosene permeation.

Table I. Stress histories used for hydraulic oedometer tests

Test No.	Vertical Effective Stress (kN/m^2)
DG1	20-50-480-50
DG2	20-50-200-50
AR1	20-50-200-600-200-50
AR2	20-50-100-200-100-50-400

Two methods were used to determine the hydraulic conductivity: either one pressure controller could be set to provide a constant flow rate, the other being held at a constant pressure, and the pressure difference recorded, or each controller could be set at a different pressure and the flow rate recorded, with in both cases the tests continuing until equal flow rates were measured by the two controllers. No significant differences could be detected between the two methods.

The hydraulic conductivity could then be determined from

$$k = \frac{Q\,H\,\gamma_w}{A\,\Delta p} \qquad (1)$$

where Q = flow rate
A = Area of the sample
H = Sample height
Δp = pressure difference
γ_w = Unit weight of water

Before each conductivity measurement the two pressure controllers were connected together so that any difference in their readings could be determined and corrected for, and the accuracy of the pressure readings was of the same order as their resolution of 1 kN/m^2. The accuracy of the volume change readings (resolution 1 mm^3) was affected by small temperature variations (+/-1°C) in the temperature controlled laboratory. The resulting volume changes were significant at low flow rates, especially when using the kerosene-water interfaces because of their additional fluid volume, and have been allowed for by running the tests over more than 24 hours to get a reliable average. For a few stages different flow rates (between 0.5 and 7 mm^3/min) were used to check on the validity of Darcy's law, and when water was the permeant only very slight differences in hydraulic conductivity were measured.

In addition to direct measurements of the hydraulic conductivity indirect measurements were obtained from the consolidation responses. In conventional analyses of one-dimensional consolidation the coefficient of consolidation, c_v is deduced from a plot of settlement against either the square root or the logarithm of time. For both of these methods a curve fitting procedure is used (eg.Lambe, 1951) to determine t_{90} and t_{50} respectively. This allows c_v to be deduced as

$$c_v = \frac{0.848\,H^2}{t_{90}} \qquad (2)$$

and then the hydraulic conductivity can be calculated from

$$k = c_v\,m_v\,\gamma_w \qquad (3)$$

where m_v = volume compressibility
γ_w = unit weight of water

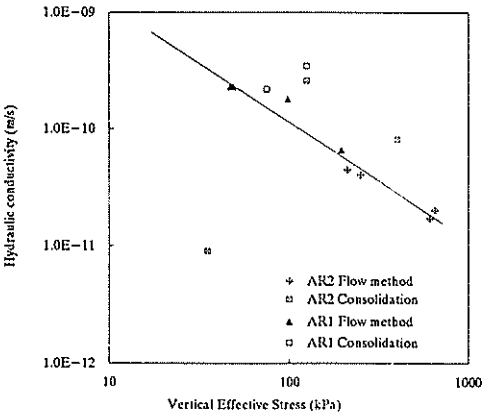

Figure 3. Relation between hydraulic conductivity and vertical effective stress during loading

6. RESULTS

Figure 3 shows a comparison of the hydraulic conductivities determined by the direct flow method and from the consolidation data plotted against vertical stress for the loading stages of tests AR1 and AR2. Results from the consolidation phases have been plotted against the mean effective vertical stress for that phase. The hydraulic conductivities determined by the direct flow method in both tests are in good agreement and can be described by a linear relation in this log-log plot. Values of the hydraulic conductivity determined from the consolidation responses appeared to be slightly higher, in these and other tests, for all but the initial stages. In all the tests the consolidation method gave a low estimate of the hydraulic conductivity for the first load increment. This occurred because the shape of the consolidation curve did not match the expected theoretical response, a result of the sample disturbance on installation into the oedometer. For test AR1 the direct flow method gave a conductivity of 3×10^{-8} m/s at a vertical effective stress of 50 kN/m^2, two orders of magnitude higher than the other values shown in Figure 3. This is believed to have been caused by leakage along the sidewalls of the oedometer, again the result of disturbance during sampling. During unloading the consolidation responses did not correspond closely with the expected theoretical response and the estimated hydraulic conductivities were up to two orders of magnitude lower than those obtained from the direct flow method. Based on these results it appears that the consolidation method can only be relied on to provide good estimates of the hydraulic conductivity for normally consolidated soils.

Figure 4 shows the hydraulic conductivities determined by the direct flow method (loading and unloading) and the indirect consolidation method (loading only) plotted against void ratio for all the tests. Although there is some scatter in this figure there is generally good agreement between the hydraulic conductivities estimated by the different methods. The scatter is not unexpected because in both methods differences in void ratio and hence hydraulic conductivity occur across the samples as a result of effective stress variations. The simple consolidation theory assumes that the hydraulic conductivity and compressibility remain constant during a load increment, whereas in practise they will be changing continuously.

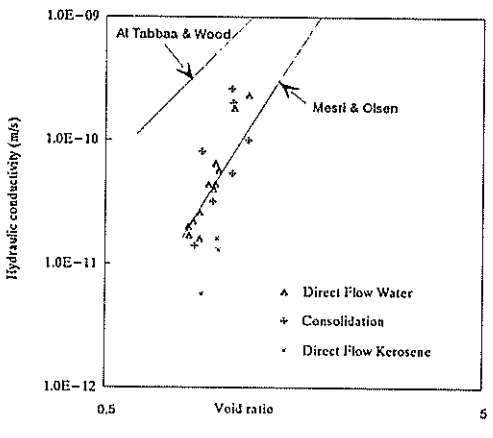

Figure 4. Relations between void ratio and hydraulic conductivity for kaolinitic clays

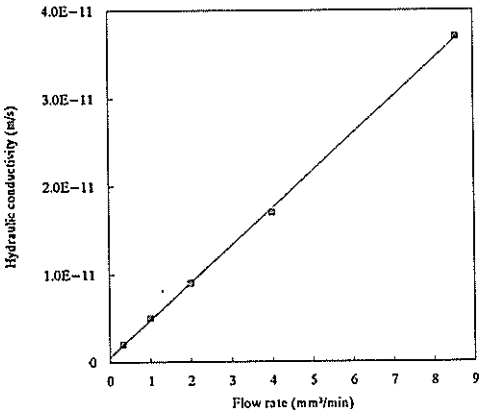

Figure 5. Relation between hydraulic conductivity and flow rate during kerosene permeation

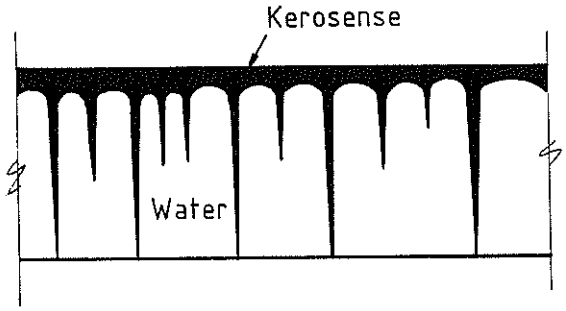

Figure 7. Sample section showing kerosene distribution

As expected the data show that the relation between hydraulic conductivity and void ratio is unaffected by the stress history of the soil, and can be described by a linear relation in this log-log plot. Similar results, also shown on Figure 4, have been obtained for other kaolinitic clays (Al Tabbaa and Wood, 1990, Mesri and Olsen, 1971). It can be seen that the results are practically identical to those reported by Mesri and Olsen.

Also shown on Figure 4 are values of the hydraulic conductivity obtained when pumping kerosene through the samples at a flow rate of 1 mm^3/min. These limited data appear to show that when kerosene is pumped through the water saturated soil the hydraulic conductivity is reduced. However, the hydraulic conductivity has been found to depend on the flow rate. For test AR2 at an effective vertical stress of 400 kN/m^2 the hydraulic conductivity increased almost linearly with the flow rate, for flow rates between 0.33 and 8.6 mm^3/min as shown in Figure 5. At this void ratio with water as the permeant an hydraulic conductivity of 3×10^{-11}m/s was expected, slightly lower than the value at the maximum flow rate using kerosene.

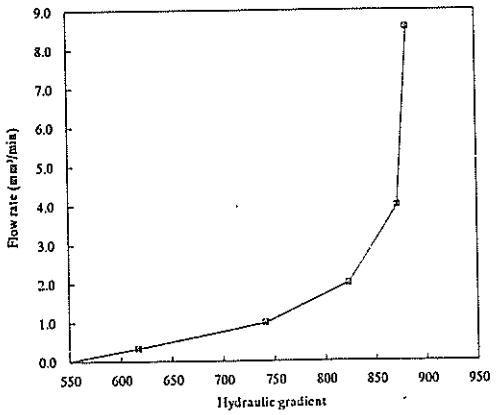

Figure 6. Relation between hydraulic gradient and flow rate during kerosene permeation

An important difference between the flows of kerosene and water was that no flow occurred for the kerosene until a breakthrough pressure was reached when the hydraulic gradient was sufficient to overcome the capillary pressures in the pores. Figure 6 shows the relation between the hydraulic gradient and flow rate which can be extrapolated to estimate the breakthrough gradient as approximately 550 for this 17 mm thick sample.

In the first test, DG1, the kerosene was pumped from the top to the bottom of the sample, and was able to pick up some black material from the flexible rubber membrane used to apply the vertical pressure. When the sample was sectioned at the end of the test a black stain in the soil enabled the presence of kerosene to be detected. It was observed that the kerosene had fingered its way through the soil (see Figure 7) and that the kerosene had only entered a small portion of the soil pores. Tests in which several pore volumes of two other immiscible liquid hydrocarbons were pumped through clays (Fernandez and Quigley, 1985) also resulted in only 5 to 10% of the pore water being replaced. In the subsequent tests the kerosene was pumped from the bottom of the oedometer so that the reaction with the rubber membrane would not affect the hydraulic conductivity, however, it was found that the hydraulic conductivity with respect to the kerosene was practically identical, that is the presence of the black material had no apparent affect.

In test AR2 after passing more than one pore volume of kerosene through the sample at an effective vertical stress of 50 kN/m^2 the vertical stress was increased by 350 kN/m^2 and the sample allowed to consolidate. The hydraulic conductivity determined indirectly from the consolidation response was 7×10^{-11} m/s. This was close to the value expected for the water saturated soil, and was further evidence that only a small amount of the pore space had been filled with kerosene. Further permeation with kerosene followed with significant reductions in hydraulic conductivity, and increases in the breakthrough pressure. The hydraulic gradient required for flow increased from approximately 150 to 550 as the void ratio decreased from 1.02 to 0.91.

7. DISCUSSION

Attempts were made to investigate the hydraulic conductivity of the natural soil, but these were unsuccessful because the samples could not be trimmed to fit the oedometers without cracking. Because the cracks would have a significant effect on the hydraulic conductivity it was decided to use only remoulded samples. However, when the soil was dried out iron that was originally in the ground water precipitated and turned the initially grey soil a reddish brown colour. On remoulding the samples the iron did not return to solution and this may have affected the hydraulic conductivities, and the values may therefore not be representative of the natural soil.

The flow of two or more immiscible fluids through porous media (rocks) is of considerable interest to petroleum engineers and has received much study (eg. Dullien, 1979). It has been shown that as an immiscible fluid such as kerosene enters a water saturated soil it has to overcome the capillary pressures at the fluid-water interface in the pores. As the capillary pressure is dependent on the pore size the fluid moves preferentially into larger pores creating the fingering type pattern shown in Figure 7. As the pressure of the fluid increases eventually breakthrough will occur and thereafter the fluid will flow only through the channels that have broken through. Some of the fluid will be left in non-conducting channels that do not influence the flow behaviour. By increasing the pressure head further, more water will be replaced by the fluid and the number of flow channels will be increased. Similar processes appear to have occurred as the kerosene was pumped through the clay samples used in these tests.

It has been found (eg. Dullien, 1979) that under conditions of steady flow the immiscible fluids flow independently of each other, with neither fluid influencing the flow behaviour of the other, and that this behaviour can be described by an extended form of Darcy's Law. For each fluid phase j:

$$v_j = \left(\frac{K_{ej}\, \gamma_j}{\mu_j} \right) i \qquad (4)$$

where v_j is defined by Q_j/A, and K_{cj} is the effective permeability of the fluid j. The effective permeabilities will thus depend on the saturation, or proportion of the pore space occupied by each phase. It is customary to express these effective permeabilities as fractions of the permeability K using:

$$K_e = K\, K_r$$

where K_r is called the relative permeability. This should take values between 0 and 1 depending on the saturation of the particular phase of interest.

The amount of kerosene pumped into the sample before breakthrough was approximately 5% of the pore fluid, so that a low relative permeability might be expected. However, the relative permeabilities varied between 0.43 at the lowest flow rate and 7.8 at the highest. High relative permeabilities, up to 2.5, have also been reported (Dullien, 1979) when viscous oils have been pumped through water saturated clayey rocks. It has been suggested that the water which is bound to the clay particles is not easily displaced by the oil (this has been demonstrated by Fernandez and Quigley, 1985), and has a "lubricating" effect, so that the kerosene behaves as if it has a viscosity lower than its bulk value. Since the bulk viscosity is used in the extended Darcy's law this will predict much smaller flows than are actually observed.

The hydraulic conductivity has been found to depend on the flow rate. A similar flow rate dependence has been reported (Quigley, 1989) from tests in which cyclohexane was pumped through a clayey soil. These observations suggest that the extended Darcy's law may not be valid when kerosene is pumped through the clay. However, the relative permeability can be expected to increase as the flow rate increases because the increased pressure head will force more kerosene into the soil increasing the kerosene saturation. The relative permeability in the extended Darcy's law is only constant for a fixed saturation.

It has been suggested (Fernandez and Quigley,1985) that the changes to the double layer thickness dominate the effects of viscosity and bulk density. If this is true then the extended Darcy's law will not be applicable as the permeability will be changing as kerosene displaces the water. The high relative permeabilities measured in these tests are indicative of chemical effects, but further tests are required to investigate the applicability of the extended Darcy's law. Alternatively, as suggested by Quigley, 1989 the kerosene may be forced along macropore channels or micro-fissures that expand or contract in proportion to the pressure head.

The results presented are in general agreement with those of other investigators who have forced insoluble liquid hydrocarbons through clayey soils. The hydraulic conductivity does not increase much beyond that of water, and significant breakthrough pressures are required to overcome the capillary pressures in the pores. However, it has been shown (Quigley, 1989) that the presence of small amounts of surfactant, as would be likely in any waste dump, can reduce the breakthrough pressure significantly so that this can not be relied on to contain insoluble liquid hydrocarbons.

7. CONCLUSIONS

The hydraulic conductivities determined from the direct flow method and indirectly from the consolidation response showed good agreement for the normally consolidated clay. The consolidation response gave poor predictions of the hydraulic conductivity on unloading and for the initial loading stage. Sidewall leakage can cause the direct flow method to indicate high hydraulic conductivities in the initial stages of the tests.

For clays the relation between hydraulic conductivity and void ratio can best be determined from tests on normally consolidated soil.

Significant hydraulic gradients were required to make the kerosene flow through the clay, and when flow did occur the hydraulic conductivity was less than for water. The hydraulic conductivity increased almost linearly with the flow rate.

ACKNOWLEDGMENT

Some of the tests were performed by David Duff and Paul Gorman as part of an undergraduate thesis. The assistance of Antonio Reyno and Dennis Nobbs is also gratefully acknowledged.

REFERENCES

Al-Tabbaa,A. and Wood,D.M.(1986) Some measurements of the permeability of kaolin. Geotechnique.

Dullien,F.A.L.(1979) Porous Media, Fluid Transport and Pore Structure. Academic Press

Fernandez,F. and Quigley,R.M.(1985) Hydraulic conductivity of natural clays permeated with simple liquid hydrocarbons. Can. Geotech. J. 22, 205-214.

Foreman,D.E. and Daniel,D.E.(1986) Permeation of compacted clay with organic chemicals. Proc ASCE, Vol 112, No 7, 669-681

Lambe,T.W.(1951) Soil Testing for Engineers. John Wiley & Sons

Mesri,G. and Olsen,R.E.(1971) Mechanisms controlling the permeability of clays. Clay and Clay Minerals, 19, 151-158.

Menzies,B.K.(1988) A computer controlled hydraulic triaxial testing system. Advanced triaxial testing of soil and rock. ASTM STP 977, 82-94.

Mitchell,J.K. and Madsen,F.T.(1987) Chemical effects on clay hydraulic conductivity. In Geotechnical Practice for Waste Disposal, ASCE Geot.Spec.Publ. No 13, 87-116.

Quigley,R.M.(1989) Pollution Migration, Waste Dumps & Groundwater. Part 1, Centre for Geotechnical Research, Sydney University.