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Gas migration through bentonite clay

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GAS MIGRATION THROUGH BENTONITE CLAY

LULEÅ MAY 31

DIV. OF SOIL MECHANICS
UNIVERSITY OF LULEÅ

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SUMMARY

Hydrogen gas produced by irradiation of pore water in the highly compacted bentonite that surrounds the copper canisters according to the KBS 2 and 3 concepts, may escape from the clay/copper interface if the gas pressure is higher than the groundwater pressure. A reasonable physical model predicts that gas may penetrate wider "capillary" passages that actually exist in the very dense clay, although these passages are still of microscopic size. In the large majority of the clay voids, the capillary action is sufficient, however, to resist gas penetration, and this suggests that a possible mechanism of gas migration is that of a "finger-like" pattern of tortuous gas passages extending from the canisters if radiolysis takes place at all. Two series of experiments have been run at gas pressures up to about 10 MPa. Nitrogen as well as hydrogen were used in these tests which seem to confirm, in principle, the validity of the physical model.

INTRODUCTION

Irradiation of groundwater produces hydrogen gas and oxidizing compounds like oxygen gas and oxygen peroxide. In the case of the KBS 2 and 3 concepts, gamma radiation is effectively shielded by the thick copper canister and produces very little radiolysis. α and β radiation have a very small penetration capacity in solid materials and are of no significance until the water is in direct contact with the uranium dioxide matrix (1). The decomposition of water through radiolysis that may take place at that stage needs to be considered, however, since pressurized gas may affect the physical status of the bentonite clay. This report deals with gas migration through highly compacted bentonite clay in general, and with hydrogen gas percolation in particular.

GAS MIGRATION THROUGH WATER SATURATED SOIL

General

Gas bubbles emerging from deeply sited organic strata and making their way upwards through the overlying soil, are commonly observed in soft Quaternary sediments. They are released through steeply oriented, continuous but narrow gas passages, which are frequently observed in samples taken from relatively shallow postglacial as well as glacial deposits. Such soils usually have a density of less than 1.5 t/m^3 and are characterized by an aggregated microstructure with fairly large voids (Fig.1). Highly compacted bentonite is much more homogeneous and less porous once it has become water saturated and matured, but it still contains numerous continuous passages which are responsible for water permeation under hydraulic gradients, and anion migration under concentration gradients (2), and through which pressurized gas may also penetrate (Fig.2).

Fig.1. Electron micrograph of ultrathin section of acrylate-bedded glacial, illitic clay. G is a truncated gas pore.

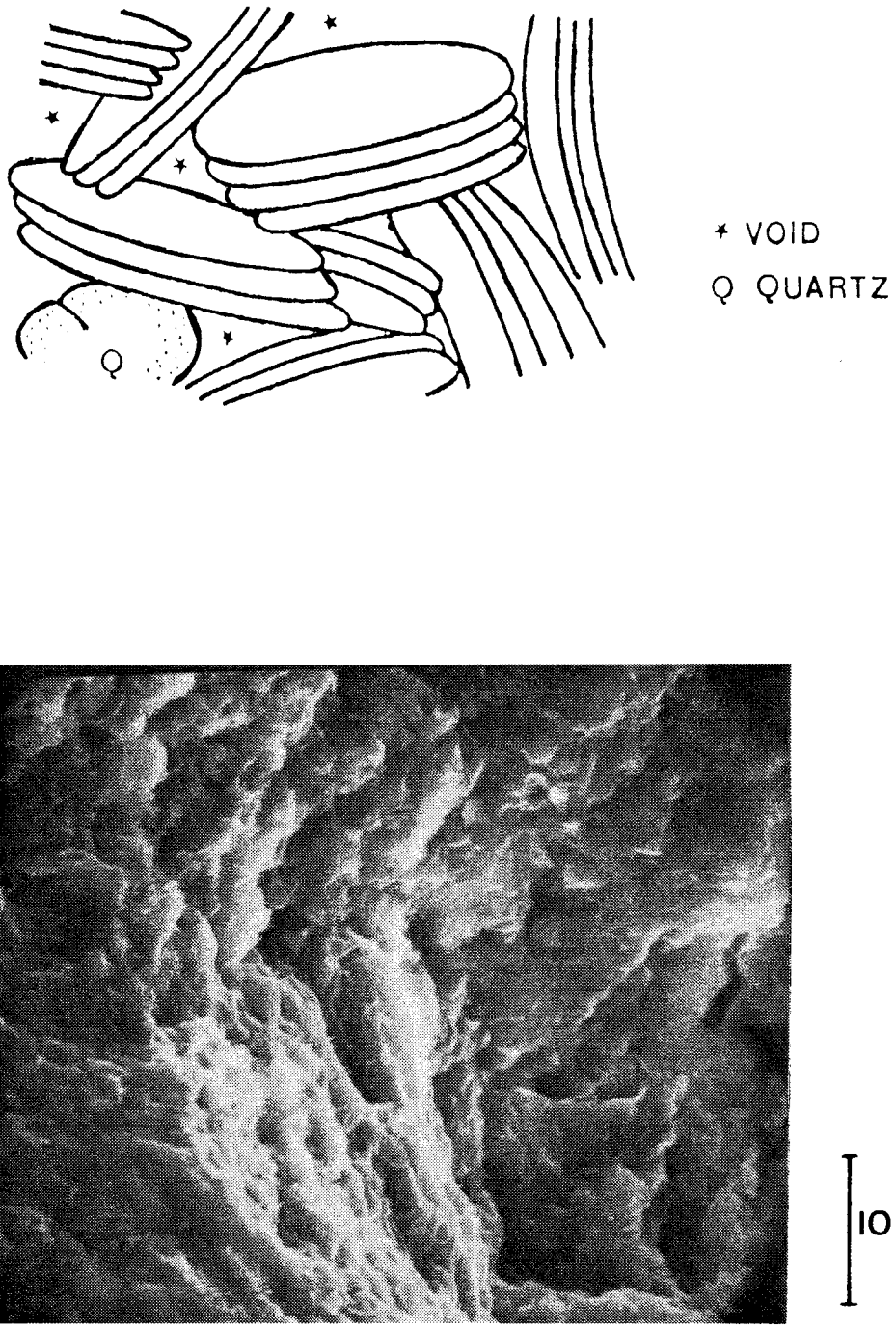


Fig.2. Microstructural characteristics of highly compacted bentonite.

Upper picture: Schematic arrangement of flake domains; stars represent continuous void passages.

Lower picture: Scanning electron micrograph of dense sodium bentonite.

Physical model

From a physical point of view, any soil, including highly compacted bentonite, can be regarded as a system of irregularly shaped capillaries (Fig.3). In clays, their width is very small and varying which leads to a largely varying capillary rise in nature. In saturated, highly compacted and matured bentonite the average "capillary" diameter can be roughly estimated at $5 \cdot 10^{-8}$ m, the statistical spread being illustrated by a 90-percentile of 10^{-6} and a 10-percentile of $5 \cdot 10^{-9}$ m. If we take the often used quartz capillary analogue as a basis of an estimation of the capillary forces (Fig.4) we arrive at the relationships:

$$\pi r^2 u + 2\pi r \sigma_s \cos\theta = 0 \qquad (1)$$

and

$$u = -\frac{2\sigma_s \cos\theta}{r} \qquad (2)$$

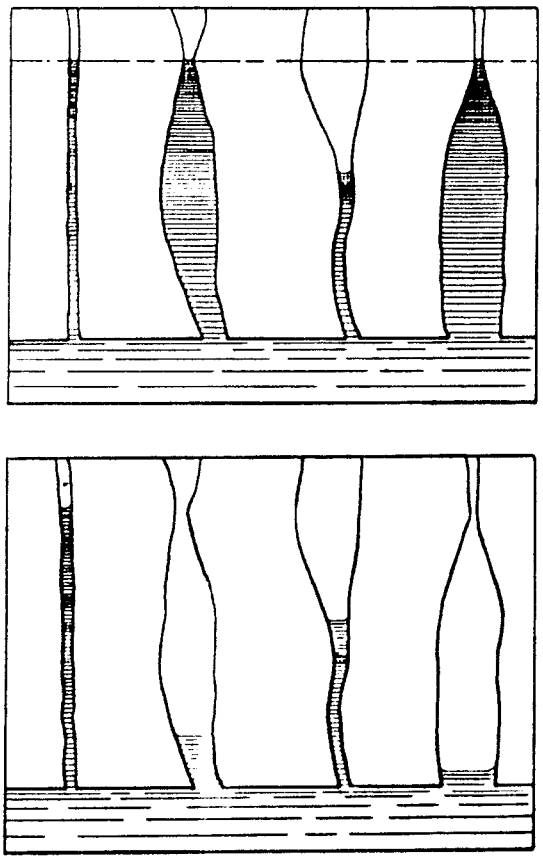


Fig.3. Capillary rise in pores of variable cross section.

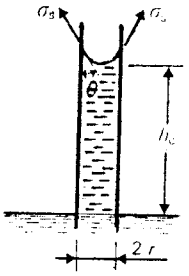


Fig.4. Schematic capillary

Since σ_s = 0.07 N/m and θ can be taken as 0°, we find the underpressure (tension) of the water at the meniscus to be about 0.3 MPa in the coarsest capillaries, while this pressure may be at least 60 MPa in very narrow capillaries. The average value is estimated at about 6 MPa.

The capillary effect is still there if the gas pressure is increased although this tends to displace the meniscus. However, gas cannot penetrate into capillaries unless the pushing force caused by the gas pressure exceeds the counteracting capillary force. If so, gas makes its way through individual continuous passages and continues to do so at a rate which is determined by the flow resistance, and by the gas production rate that is required to keep up the pressure. In addition, the gas pressure must exceed the water pressure, which amounts to about 5 MPa at the 500 m depth where KBS HLW repositories will be located. The minimum required gas pressure to produce gas migration is thus 5 MPa, and if all the capillaries were finer than $5 \cdot 10^{-8}$ m, this pressure would have to be higher than about 11 MPa. At an early stage of the investigation (3), this was assumed to be a critical pressure on purely theoretical grounds. Gas pressures in the range of 5-11 MPa were therefore assumed to create a gas bubble at the clay/canister interface and no penetration of gas into individual capillaries.

Recent microstructural and diffusion studies show that wider apertures do exist in highly compacted bentonite, and it is therefore probable that gas pressures exceeding 5 MPa will actually yield gas migration. The gas flow at pressures of this order of magnitude is expected to be insignificant, but pressures of about 10 Mpa will probably cause gas migration which is not negligible.

Experimental

Test program

Gas permeation tests can preferably be run by use of the swelling pressure oedometer since the sample first has to be water saturated and then kept confined without volume change throughout the long period of time that is required for attaining steady state conditions. There are practical difficulties, however, particularly with respect to the sealing of the test device, and pilot tests were therefore run with nitrogen gas at low and high pressures before the main test series with hydrogen gas were started. The first nitrogen gas test ran for 1 month, while a second test with high pressure was in operation for almost 8 months. Only one hydrogen gas test was conducted and it ran for almost 4 weeks. The main test data are specified in Table 1, the clay material being MX-80 Na bentonite, and distilled water being used for its saturation prior to the gas permeation.

Table 1 Main test data

Test no	Gas	Bulk density* t/m^3	Gas pressure MPa	Duration days
1	Nitrogen	1.86	1.0	31
2	Nitrogen	2.05	~10	217
3	Hydrogen	1.99	~3.5 - 10	25

* Water satured

Experimental set-up

The swelling pressure oedometer (4) was used for the preparation of clay samples of 5 cm diameter and approximately 1 cm thickness for the two densest samples and about 2 cm for the softest one. After saturation, which lasted for 10 to 30 days to guarantee complete filling of the voids, a gas collector in the form of a burette was attached to the outflow end of the oedometer (Fig.5). Before the application of gas pressure, the filter at the inflow end was dried so that the gas would not have to drive out water from the pores of the filter. Fig.6 shows the appearance of the test arrangement.

The nitrogen gas tests did not offer difficulties in using the standard swelling pressure oedometer, while the hydrogen gas experiment required replacement of the standard sealings with copper. Still, slight leakage took place, possibly along the interface between the clay and the oedometer ring.

Results

The nitrogen gas experiments yielded the gas conductivities that are specified in Table 2. The evaluation was made by applying Darcy's law, i.e. assuming direct proportionality between flow rate and the product of pressure gradient and conductivity. Since gas penetration is associated with the extrusion of water and this process is far from uniform over the pressurized cross section, the derived gas conductivities can only be taken as a rough measure of how gas penetrates the clay. The matter will be further discussed later in the text.

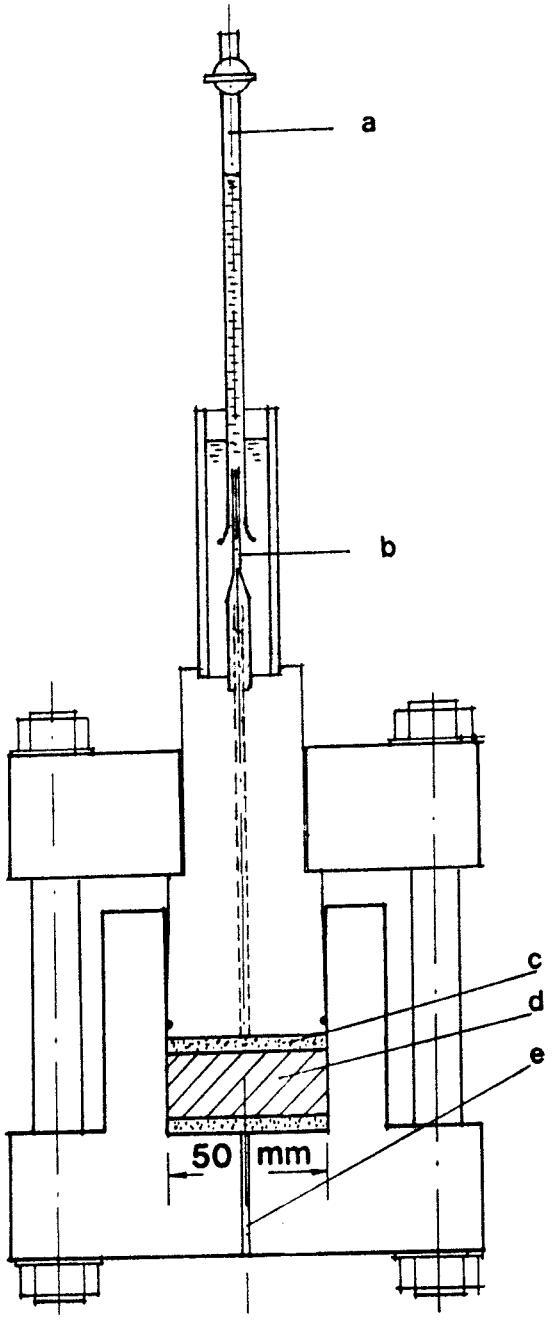
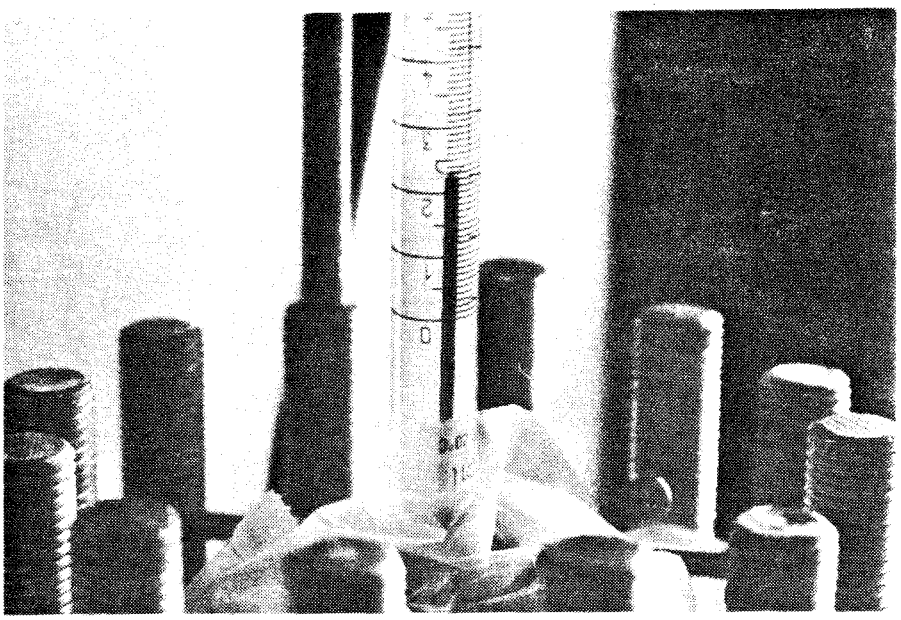


Fig.5. Device for determination of the gas conductivity of highly compacted bentonite.

a) Burette for gas collection, b) Syringe, c) Filter, d) clay sample, e) gas inlet.



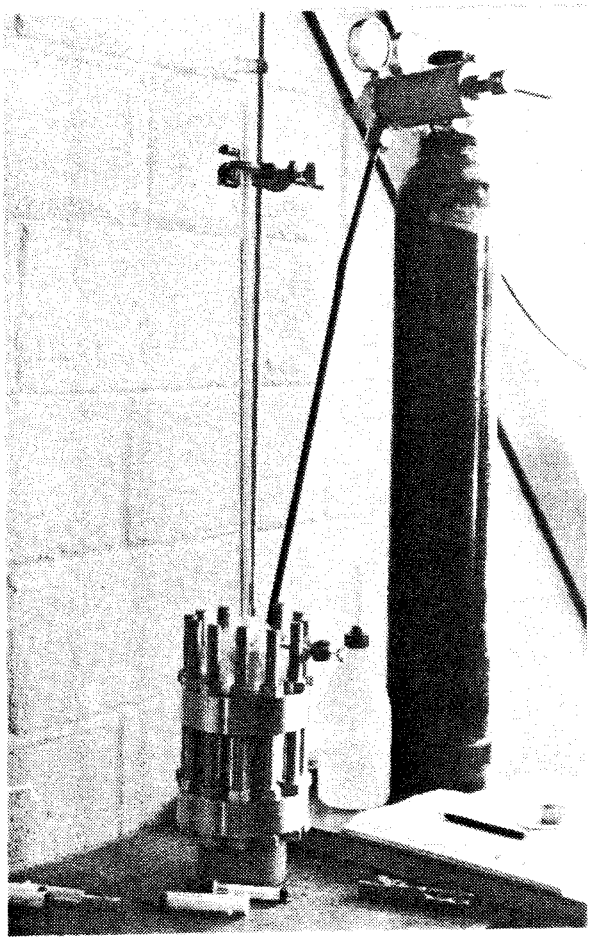


Fig.6 Experimental set-up.

Table 2. Nitrogen gas conductivity

Test no	Bulk density t/m^3	Gas pressure MPa	Time days	Gas conductivity m/s
1	1.86	1.0	31	$8.0 \cdot 10^{-14}$
2	2.05	~10	1-7	$2.8 \cdot 10^{-14}$
			7-37	$3.0 \cdot 10^{-14}$
			37-54	$2.6 \cdot 10^{-14}$
			54-79	$3.3 \cdot 10^{-14}$
			79-132	$2.8 \cdot 10^{-14}$
			132-182	$2.9 \cdot 10^{-14}$
			182-217	$2.8 \cdot 10^{-14}$

After the tests, the samples were extruded and examined with respect to their water content and density, as well as to their appearance. A careful determination of these data showed that the degree of water saturation was 100 %, the estimated accuracy of this calculation being of the order of 1 to 2 percent units. This means that the gas has not replaced much of the water in the samples and, therefore, that it must have migrated through a small fraction of the cross section only.

The hydrogen gas experiment was run under controlled, and successively increased gas pressures for 25 days, after which an accidental pressure drop happened. The pressure could not be raised again until one month later, and after a few days the test was stopped and the sample extruded for examination. It showed that the clay was no longer water saturated; certain zones extending from below right through the sample being considerably dryer than the rest of the sample. The most probable explanation of this phenomenon is that some drying took place during the period when no gas pressure prevailed, and that further removal of water was facilited when the pressure was raised again in the latest phase of the test (Fig.7).

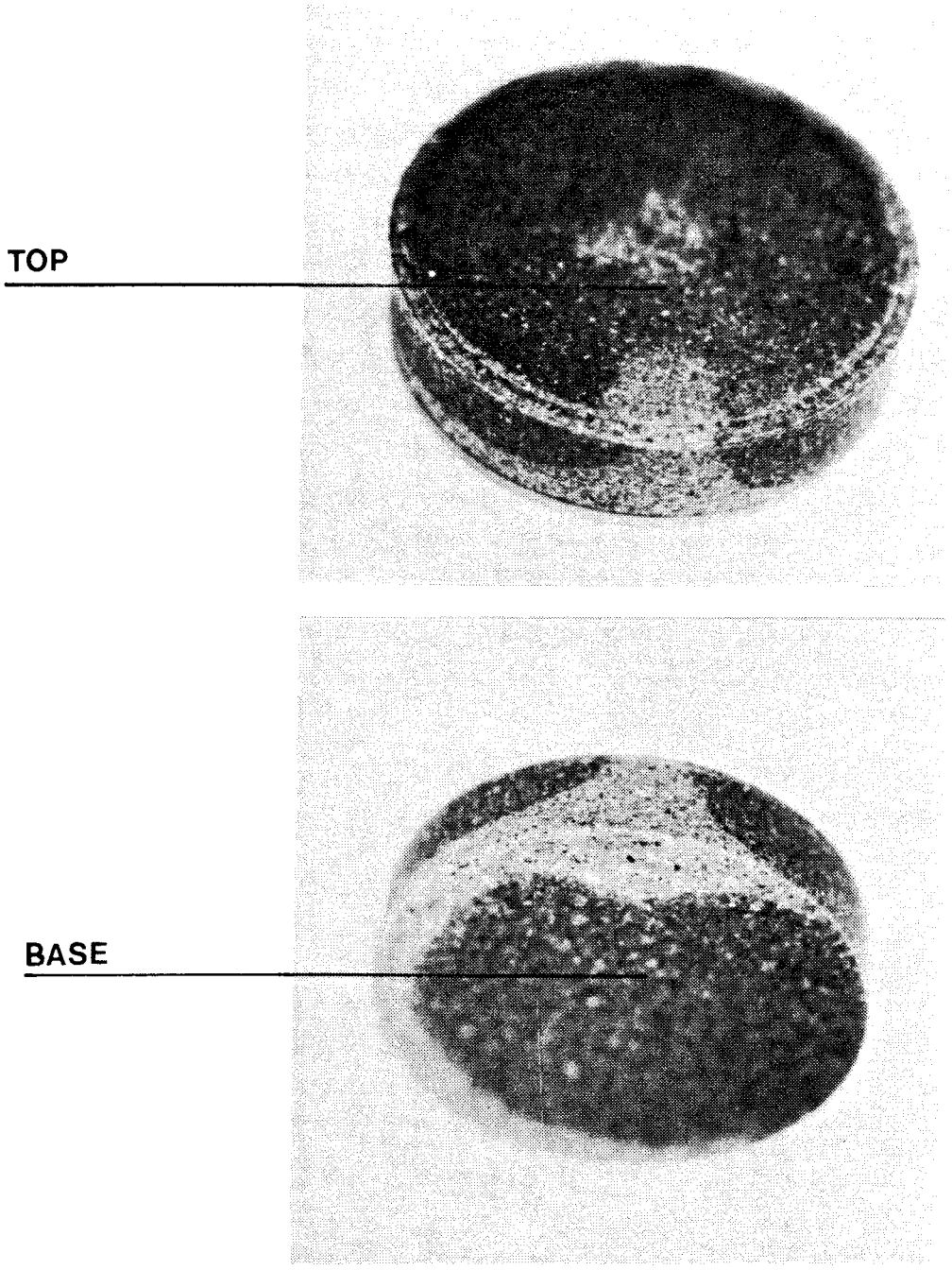


Fig.7. Appearance of the extracted sample percolated by hydrogen gas. The drier zones, which are bright, were probably partly formed during a period of pressure drop.

In contrast with the samples percolated by nitrogen gas, the presently discussed clay sample had a very clear yellow/brownish color of the entire basal plane at the outflow end. Its uniform distribution over this plane and its uniform extension inwards, i.e. about 0.1 mm, suggests that the hydrogen gas, which is assumed to have caused the phenomenon, had actually penetrated narrow, capillary passages that were uniformly distributed over the cross section in the initial 25 day period when the gas was pressurized. Thus, the gas migration processes are probably similar for both gas species from a physical point of view, while there seems to be some sort of chemical reaction between the hydrogen gas and the bentonite. This does not necessarily have to do with the smectite; it may well concern other mineral constituents, like oxides, or the organic content. It should be pointed out that the plastic nature of the miscolored clay was the same as that of ordinary MX-80 clay.

The evaluated hydrogen gas conductivity is specified in Table 3, with particular reference to the influence of time.

Table 3. Hydrogen gas conductivity

Test no	Bulk density t/m^3	Gas pressure MPa	Time days	Gas conductivity m/s
3	1.99	3.5-10.5	0-6	$1.2 \cdot 10^{-13}$
		5.0- 5.5	7-9	$1.8 \cdot 10^{-13}$
		3.5- 7.3	10-15	$1.4 \cdot 10^{-13}$
		3.5- 3.9	16-25	$1.6 \cdot 10^{-13}$

The uniform percolation rate supports the belief that the gas did not extrude water to form growing zones of low water content during the initial 25 day period.

Conclusions

If the nitrogen gas conductivity is evaluated in the same way as the hydraulic conductivity as we have been doing in the present study, it turns out that they are of a similar order of magnitude. Thus, we find the gas conductivity to range from about $8 \cdot 10^{-14}$ to $3 \cdot 10^{-14}$ m/s for the bulk density interval 1.86 to 2.05 t/m^3, while the corresponding hydraulic conductivity range is $4 \cdot 10^{-14}$ to $2 \cdot 10^{-14}$ m/s. Still, gas and water percolation must represent very different mechanisms, at least when continuous gas passages have been formed at a certain time after the onset of flow. Then, it is reasonable to believe that practically all the gas passes through the clay without displacing pore water, while water flow through water saturated clay under hydraulic gradients is expected to involve displacement of the major part of the pore water in all continuous passages.

The fact that nitrogen gas actually passed through bentonite without replacing water to a significant extent can only be explained by assuming that the gas flow took place through a very small fraction of the cross section, which can be taken as an evidence of the validity of our physical model. Thus, a small number of the coarser capillaries - they are still microscopic in size - offer only slight capillary resistance to the penetrating gas, and therefore let it through very soon after the application of a gas pressure. Actually, the nitrogen gas flow in our 10 Mpa experiment amounted to about $5 \cdot 10^{-12}$ m^3 of gas at atmospheric pressure per second in less than one hour, and this flow was then only slightly changed with time.

There are reasons to believe that hydrogen gas behaves similar to nitrogen gas with respect to the flow properties. In the present test the conductivity of the first-mentioned was found to be about 3 times higher, but our experience is that leakage can hardly be avoided with the authors' experimental set-up and that the conductivities probably are of the same order of magnitude. As to the miscoloring, its nature remains to be explained.

The main conclusion with respect to the practical consequence of gas production at the clay/canister interface, is that hydrogen gas will start to migrate in a "finger-like" manner through the clay as soon as the gas pressure exceeds the ground water pressure. When the excess gas pressure is low, the gas flow will be insignificant, while higher pressures will produce stronger flow, which will probably still be confined to very narrow, tortuous passages. As long as its pressure is maintained, the gas will continue to migrate upwards from the close vicinity of the canisters, through the more easily penetrated tunnel and shaft backfills, and towards the ground surface. It is required, of course, that inflow of water through the highly compacted bentonite takes place towards the canisters to maintain the radiolysis. This inward migration of water occurs through finer passages and is governed by pressure gradients, capillary suction and, above all, by the strong hydration power of the smectite minerals.

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