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Swelling pressure of highly compacted bentonite

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Luleå 1980-08-20

SWELLING PRESSURE OF HIGHLY COMPACTED BENTONITE

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This report concerns a study which was conducted for the KBS project. The conclusions and viewpoints presented in the report are those of the author(s) and do not necessarily coincide with those of the client.

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SWELLING PRESSURE
OF HIGHLY COMPACTED
BENTONITE

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CONTENTS

	Page
INTRODUCTION	1
DESCRIPTION OF MATERIALS	1
EXPERIMENTAL	3
The swelling pressure oedometer	3
Test program	5
Test results	7
MX-80 bentonite	12
Erbslöh bentonite	13
DISCUSSION	13
Low density conditions	13
Theoretical prediction of swelling pressures	15
Homogeneity as a function of time and temperature	17
CONCLUSIONS	17
REFERENCES	19
APPENDIX	

INTRODUCTION

The water uptake of swelling clays is due to the affinity of the mineral surfaces to water and to osmotic forces, e.g. the hydration of surface-adsorbed ions. If the clay is confined so that swelling is hindered, a "swelling pressure" will be exerted on the confining boundaries. This pressure ranges from a low value at low bulk densities to high values when the clay is as dense as suggested for the Swedish KBS 2 concept. This concept implies that highly compacted, dense Na bentonite is used to isolate metal canisters with highly radioactive waste products from the surrounding rock. The main object of the study reported here was to investigate the relationship between the swelling pressure and bulk density of bentonite with special reference to the influence of ion exchange and temperature. The matter has been dealt with in two preliminary reports by the author (PUSCH 1978 and 1979)

DESCRIPTION OF MATERIALS

The bentonites intended for the preparation of highly compacted blocks of clay should be granulated with a size of the aggregates mainly corresponding to the silt and fine sand fractions. Two suitable commercially available bentonites were investigated here; the American Colloid Co. type MX-80 (Na Wyoming bentonite) being the main KBS reference material, and the Erbslöh Ca bentonite, representing a natural calcium-saturated clay material. Both clays are characterized by a minus 2 μm content of approximately 85%, and a montmorillonite content of about 80-90% of this fraction.

From a chemical point of view it is important to notice that the two bentonites are not purely sodium- or calcium-saturated clay materials. Thus, preliminary analyses at LuH show that Erbslöh Ca bentonite contains 20-60 mg Ca, 15-30 mg Mg, and 20-40 mg Na per liter of pore water, while the Wyoming bentonite has a content of about 30 mg Ca, 15 mg Mg and 70 mg Na per liter of pore water. This relatively small difference points to fairly similar swelling properties as well, which is also verified by the test results.

The microstructural constitution turns out to be of great importance with respect to the swelling pressure. It is illustrated by Fig. 1, which shows a dominant anisotropy of the aggregates. At moderate pressures, the overall particle orientation is largely random, while at high pressures the aggregates are pressed against each other which yields considerable contact areas and aggregate deformation, as well as a tendency of the particles to be oriented perpendicularly to the applied pressure.

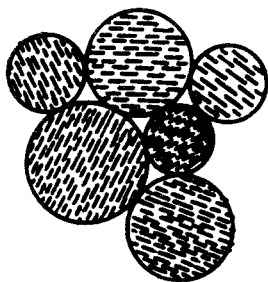


Fig. 1 Schematic microstructure of MX-80 and Erbslöh type bentonites

EXPERIMENTAL

The investigation comprised the development of suitable equipment and techniques for measuring the swelling pressure, as well as the performance of a large series of swelling pressure determinations.

The swelling pressure oedometer

More than 90 swelling pressure tests have been performed by using a new type of oedometer. Such a device had to be developed since commercially available oedometers were found to suffer from the drawback of not being sufficiently rigid to sustain neither the compaction of bentonite powder to high densities, nor the development of high swelling pressures without large strain of the oedometer itself.

In principle, the oedometer was operated in the following way (Fig. 2). Air-dry bentonite powder was uniaxially compacted to various densities by loading the slightly conical piston with the upper filter stone of stainless steel at its base. The piston was then locked in position by the steel ring equipped with a strain gauge shown on top of the piston. The ring, which rested against the top plate, was kept practically stress-free at the mounting of the device. This first operation yielded a sample of compacted bentonite with a water content (ratio of mass of water to that of solid, mineral substance) of approximately 10%, thus simulating the production of precompacted bentonite blocks, which are intended to be used in the deposition holes in future rock storages of the KBS 2 concept type.

Water of various compositions in the different tests was then let in from below or from above as well, while keeping the sample confined in its cylindrical space. The swelling pressure produced by the water uptake caused a compressive stress in

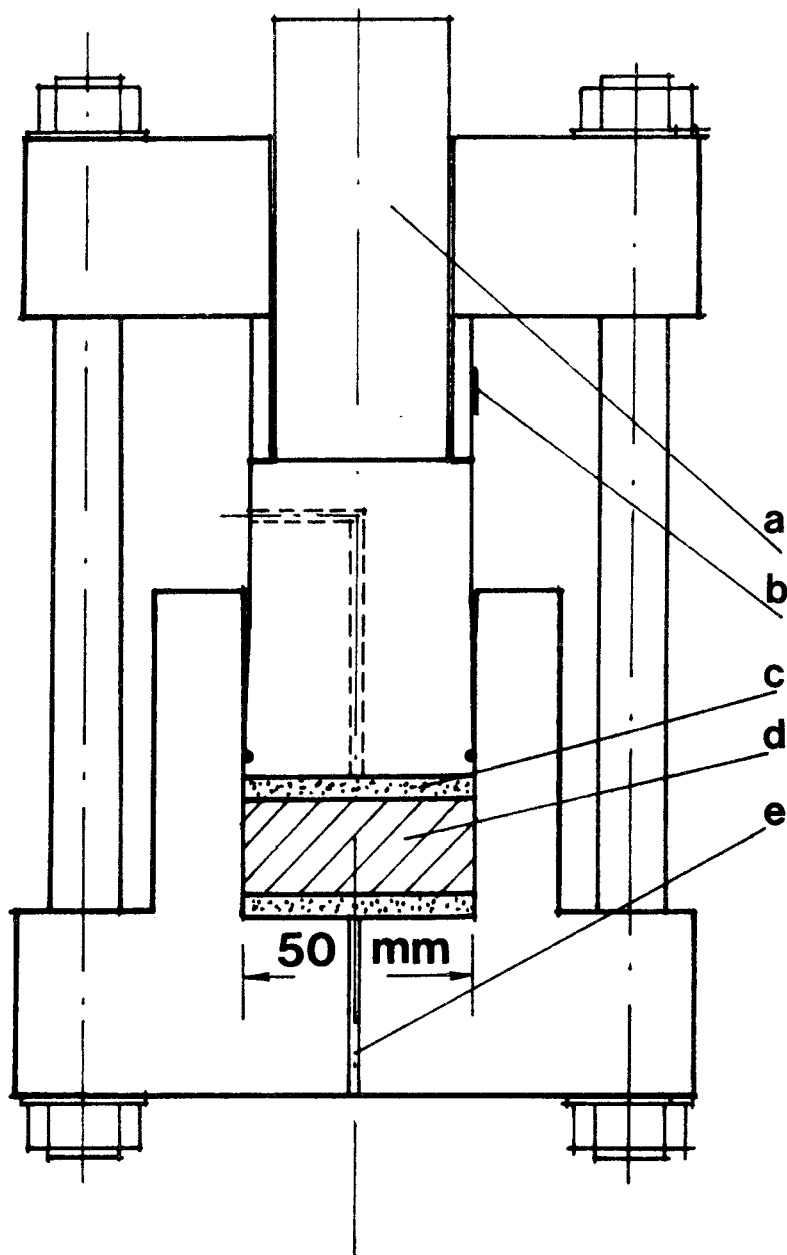


Fig. 2 The LuH swelling pressure oedometer
a) Loose piston for loading the sample.
b) Strain gauge glued to the ring.
c) Filter stone
d) Sample
e) Lower water entrance

the steel ring. It was recorded regularly by placing the device in a press and loading the upper loose piston until the ring became stress-free. The applied load thus corresponded to the swelling force under practically constant volume conditions of the bentonite sample.

A number of duplicate tests were made by using the same type of oedometer but with load cells to record the swelling force. They all gave lower values because of the rather large strain of the load cells (about 50-100 μm) at the measurements.

Test program

While the initial water content of the non-saturated bentonite powders was practically the same in all the tests, the sample height, water inlet geometry, and water composition 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, which is the state where the swelling pressure tends to stabilize.

The sample height was varied from 1 cm to 4 cm, with water inlet at one or both ends. Four different water solutions were used: 0.6 M NaCl solution, 0.3 M CaCl_2 solution, artificial ground water ("Allard water", cf. KBS Report 98), and distilled water. "Allard water" is an artificial solution with a composition similar to that of ground water at large depths in Swedish crystalline rock. The tests were run at 20°C and 90°C as well, to check the influence of temperature. In some tests the samples were allowed to swell during the first stages of the water uptake but were then confined to maintain a constant volume. These tests were made to study, qualitatively, the nature of the swelling pressure, e.g. the importance of elastic rebound with respect to that of colloidal origin.

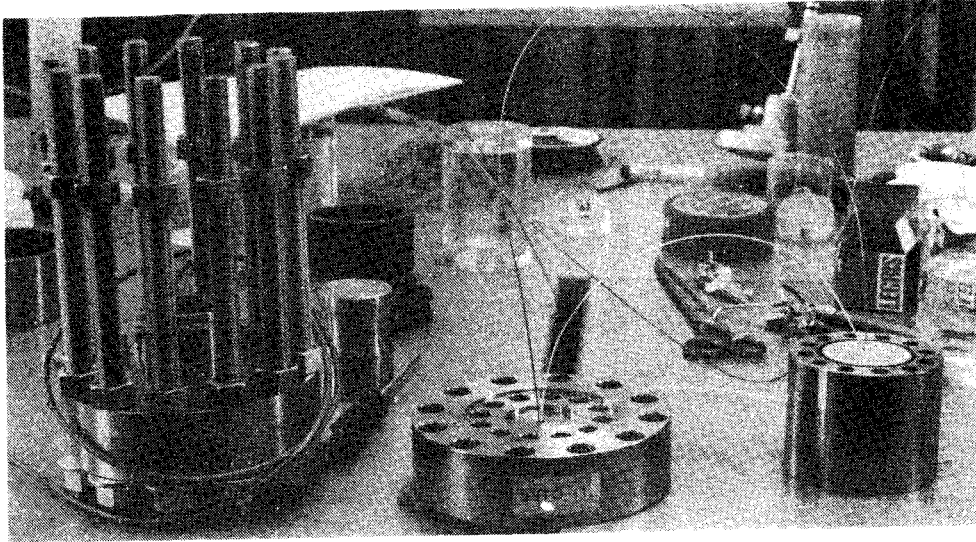


Fig. 3 Major parts of the swelling pressure oedometer.

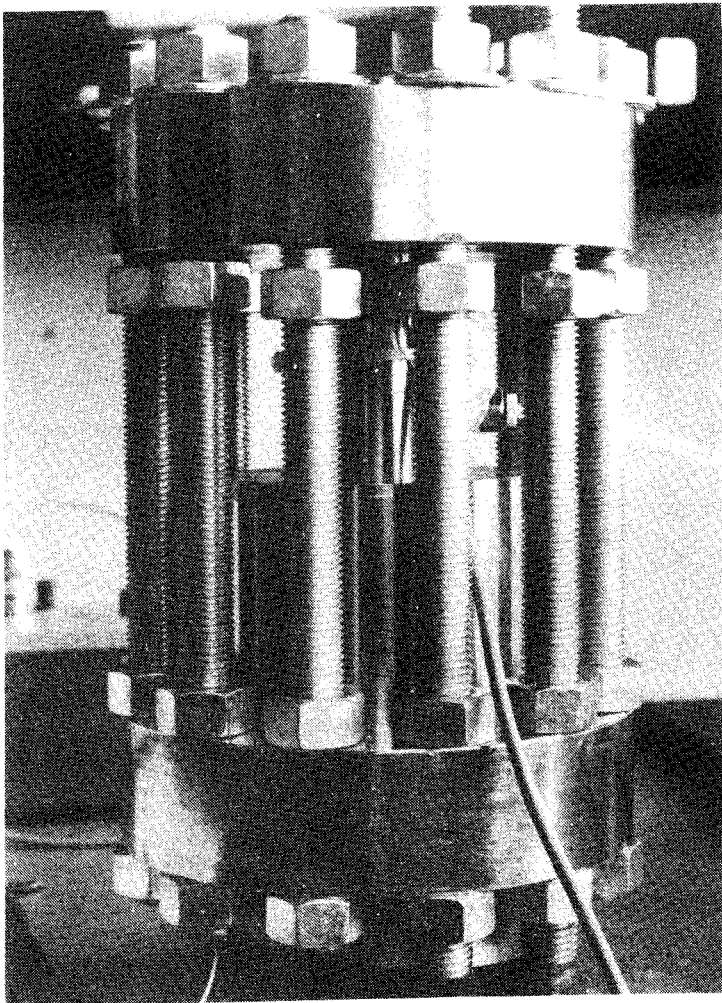


Fig. 4 Swelling pressure oedometer in operation.

Great care was taken to observe the time-dependence of the development of the swelling pressure since this was assumed to illustrate the rate of water uptake or migration as well. This problem is not considered in the present report.

Test results

Most of the tests gave the curve shape shown in Fig. 5, meaning that two maxima were observed. In some cases the first maximum is replaced by a shift¹⁾ but the general trend of the curve is still the same. A selection of representative records are presented in an appendix.

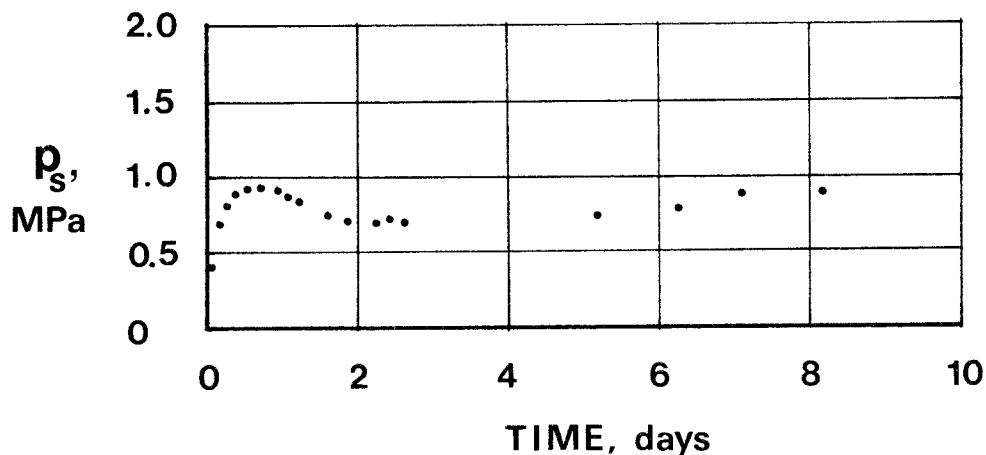


Fig. 5 Typical plot of swelling pressure versus time. MX-80 bentonite, sample height 2 cm, water uptake from both ends; distilled water. (Bulk density 1.8 t/m^3 of saturated sample).

The first peak value is probably mainly related to the energy of the elastic network of aggregates built in by the compaction, and to some extent to the pressure produced when the aggregates start to take up water while they are still strong structural units.

¹⁾ Inflexion or irregular

The successive water uptake produces an increased interparticle distance which leads to a lower swelling pressure and shear strength in the inter-aggregate contact regions. The aggregates are then easily displaced, and deformed by which some of the continuity and strength of the network is lost. This results in a decreased pressure after the first peak. Further water uptake is associated with a redistribution of particles by which the clay is transferred from an aggregated to a more homogeneous, dispersed state. A successively increased number of active inter-particle bridges, acting as springs, is then formed which increases the swelling pressure and eventually yields a second maximum.

Figs. 6 and 7 show the experimental results of the swelling pressure tests on MX-80 bentonite at $\sim 20^{\circ}\text{C}$ and 90°C respectively, while Fig. 8 gives the result of the Erbslöh tests. The plotted values represent the second maximum, which was fully developed after a time t_s in seconds approximately given by the expression¹⁾:

$$t_s = C \cdot \frac{H^2}{(-u) \cdot k} \quad (1)$$

where C = constant (approximately = 1)

H = height of sample at water uptake from one end (m)

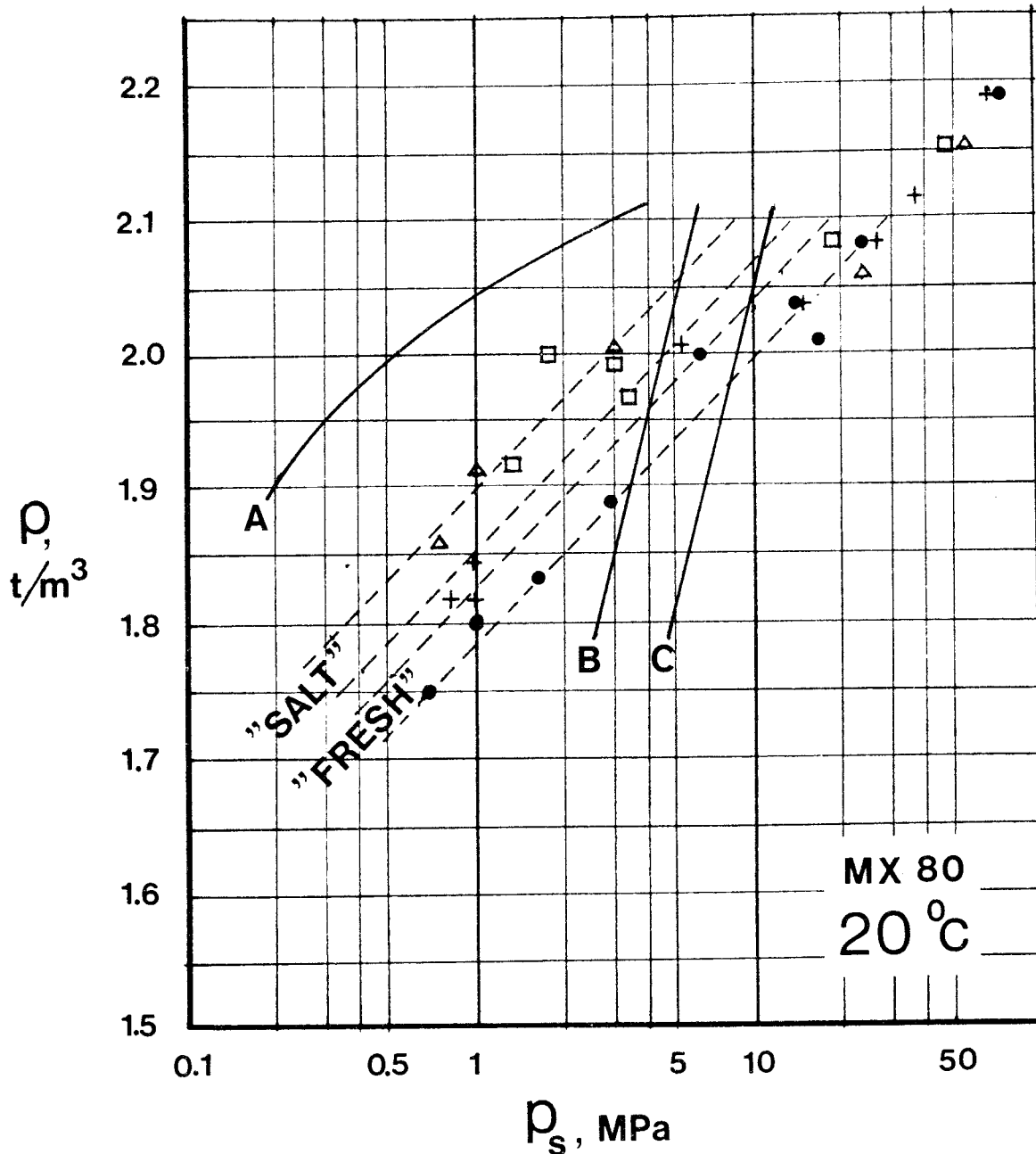
u = suction (equivalent piezometric head)²⁾ (m)

k = permeability (m/s)

The secondary maximum is developed when the water saturation is practically complete. The ρ -values in Figs. 6-8 refer to a water-saturated state.

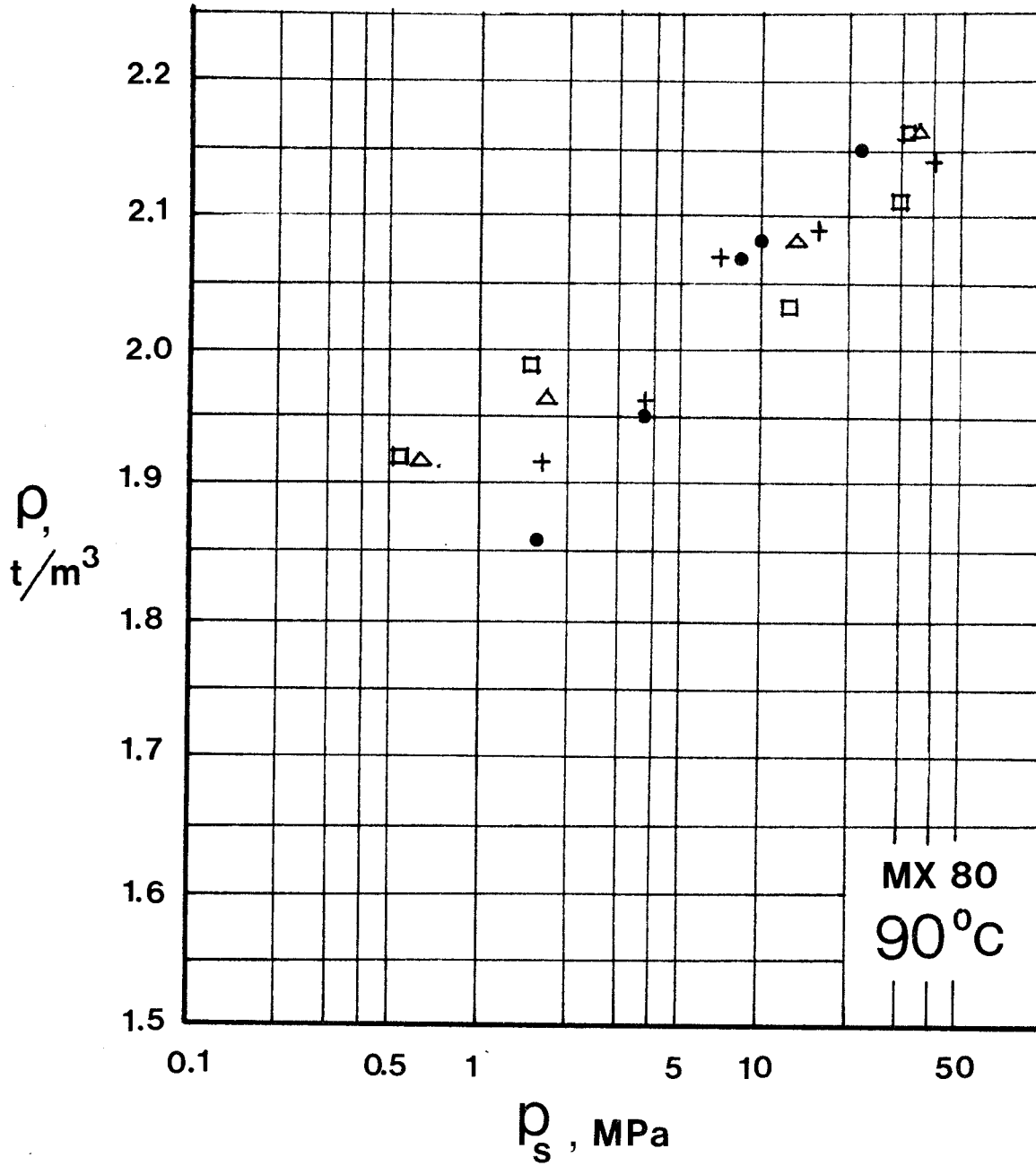
1) This empirical relationship was derived by Lennart Börgesson, Div. of Soil Mechanics, Univ. of Luleå, who was responsible for much of the design of the test equipment and for the supervision of the experiments.

2) $(-u)$ is approximately equal to the swelling pressure as concluded from current investigations.



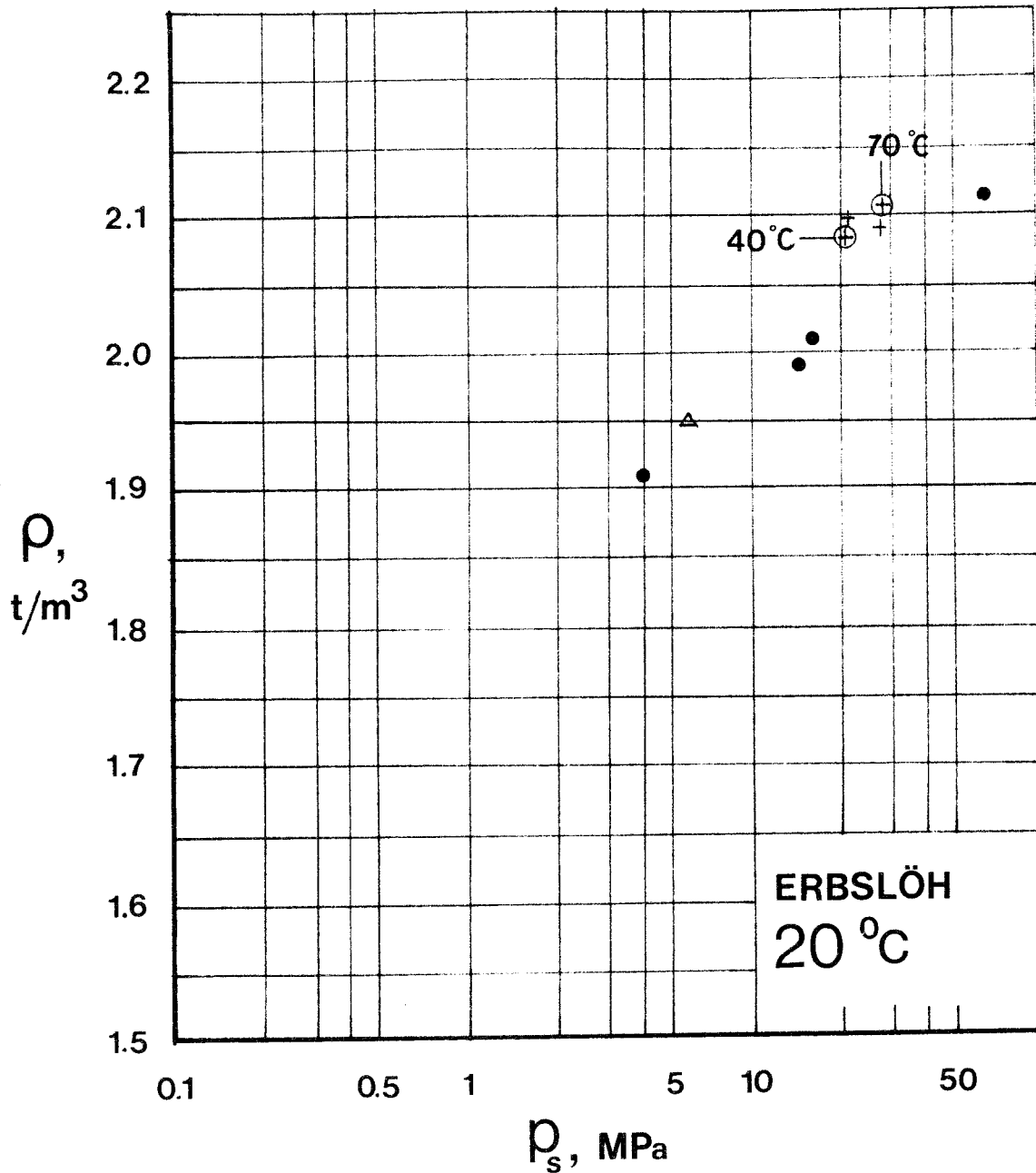
- + ART. GROUND WATER (ALLARD)
- DIST. WATER
- Δ 0.6 M NaCl SOL.
- 0.3 M CaCl₂ SOL.

Fig. 6 Recorded swelling pressure of MX-80 bentonite at 20°C. A, B, and C represent derived relationships between ρ and p_s using YONG & WARKENTIN's theory disregarding microstructural effects. A represents Ca-saturated montmorillonite in 0.3 M CaCl₂ solution while B, and C represent Na montmorillonite in $3 \cdot 10^{-2}$ M NaCl solution, and distilled water, respectively (cf. p. 16).



- + ART. GROUND WATER (ALLARD)
- DIST. WATER
- Δ 0.6 M NaCl SOL.
- 0.3 M CaCl₂ SOL.

Fig. 7 Recorded swelling pressure of MX-80 bentonite at 90°C.



- + ART. GROUND WATER (ALLARD)
- DIST. WATER
- △ 0.6 M NaCl SOL.

Fig. 8 Recorded swelling pressure of Erbslöh bentonite at 20°C. Two observations at higher temperature are included.

a. MX-80 bentonite

It is concluded from Fig. 6 that the pore water chemistry is not a determinant of the swelling pressure, p_s , when the bulk density exceeds about 2.05 t/m^3 . Below this value the observations can be roughly grouped into two zones, one representing the "salt" solutions, and the other representing distilled water as well as the artificial "Allard" ground water. It can be concluded that even if the salinity of the ground water presently observed at this depth is increased to that of the oceans, the swelling pressure will not be dramatically changed. The actual change will be a drop of p_s to $\frac{1}{3}$ to $\frac{1}{2}$, at maximum, of the original value.

It can be concluded also that there is a slight difference only between the p_s -values obtained when NaCl and CaCl_2 solutions were used for water saturation and percolation. The latter solutions yield values which are not less than about 50% of those obtained for NaCl solutions. This indicates that a complete ion exchange from Na to Ca, which can be expected to take place close to concrete supports, bulkheads etc., affects the swelling pressure of MX-80 bentonite to a moderate extent only.

A comparison of Figs. 6 and 7 shows that heating to 90°C has a moderate but obvious impact on the swelling pressure of Na bentonite, saturated and percolated with Allard's and distilled water, as well as with the "salt" solutions. A lower swelling pressure at 90°C is observed than at room temperature. The effect seems to be especially obvious for low densities and salt water, and for high densities and fresh water. The number of tests is too small to give any definite relationship between swelling pressure and temperature, but a 50% reduction when the temperature is raised from 20 to 90°C should be expected.

b. Erbslöh bentonite

Fig. 8 shows that the Erbslöh Ca bentonite behaves in principle as the MX-80 bentonite. Thus, at room temperature, Allard's, and distilled water give swelling pressures of practically the same order of magnitude as the MX-80 material. The single test with salt water indicates that a high concentration of Na does not produce a significant increase of the swelling pressure. A temperature raise to 40 and 70°C, respectively, seems to have only a very small effect on the swelling pressure.

These observations naturally mean that the Erbslöh bentonite is of great potential interest for practical use.

DISCUSSION

Low density conditions

The magnitude of the swelling pressure at densities lower than the ones investigated by the author is of interest in the practical use of bentonite as an isolation. Very valuable information in this respect is offered by a comprehensive study by LOW (1980) of pure Na montmorillonite gels. 35 different montmorillonites from various parts of the world were investigated with reference to the crystal lattice dimensions, structural formulae, specific surface area, and to the swelling pressure at various, high water contents. Fig. 9 presents the published values reinterpreted in terms of p_s versus ρ assuming ρ_s to be 2.70 t/m³¹⁾. They are all located in a slightly curved band (L) which definitely proves that also water-rich Na montmorillonite gels produce a considerable swelling pressure (more than 0.2 MPa at $\rho = 1.3$ t/m³). It should be observed that the L-band forms a somewhat skew continuation of the "fresh"-water band of MX-80 (cf. Fig. 6).

¹⁾ All the samples were prepared by mixing montmorillonite and distilled water.

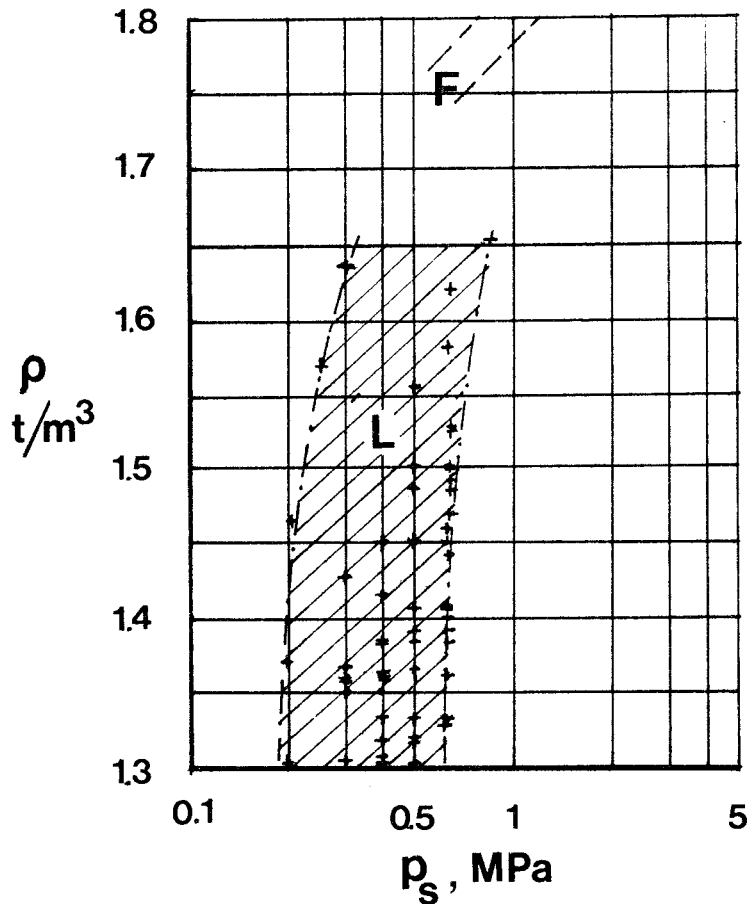


Fig. 9 Swelling pressure versus bulk density of saturated Na montmorillonite. (Reinterpreted from LOW, 1980). F represents the "FRESH"-zone in Fig. 6.

The non-perfect fit is explained by the fact that MX-80 is not a pure montmorillonite, and that it therefore yields a swelling pressure which should be only about 60-75% of that of pure Na montmorillonite, assuming the swelling pressure of the bentonite to be proportional to its montmorillonite content. It can be concluded, on the basis of Fig. 9, that fresh water-saturated MX-80 bentonite expanded to give $\rho = 1.3 \text{ t/m}^3$ gives a swelling pressure of not less than 0.1 MPa at room temperature.

It should be mentioned here that the tests performed with an initial free swelling to reduce the influence of built-in energy which produces a swelling pressure due to elastic

rebound, gave practically the same final p_s -values as the samples which were confined from the start of test. This confirms that a homogeneous state is arrived at irrespectively of what the internal aggregate and particle displacements may have been. Thus, there seems to be a unique relationship between the bulk density and the swelling pressure.

Theoretical prediction of swelling pressures

Much work was devoted throughout the investigation to find or develop a suitable method for predicting the swelling pressure on a theoretical basis. Although it did not turn out to be very fruitful, valuable information was gained concerning the physico/mechanical behaviour of this structurally complex clay.

The most promising approach is probably offered by the YONG & WARKENTIN (1975) double-layer theory. This theory is based on a model of charged, tabular particles in parallel arrangement with diffuse layers of exchangeable ions, which overlap, resulting in a higher ion concentration between particles than in bulk pore water. Also, the plausible assumption is made that water adsorbed to the clay surface accounts for the first few water layers taken up on swelling and that further swelling results from osmotic pressure. The properties required for making the calculation are the specific surface area, the kind of exchangeable cations, the concentration of ions in the pore water, and the surface density of the clay. The concentration C_c of cations at the midpoint between two interacting plates can be taken as:

$$C_c = \frac{\pi^2}{z^2 B (d+x_0)^2 10^{-16}} \quad (2)$$

where C_c = concentration in mol/l; z = valence of exchangeable cation; d = half-distance between two clay plates in Å; x_o = correction factor of 1-4 Å; and $B = 10^{15}$ cm/mmol.

The swelling pressure p_s can then be calculated from the van't Hoff equation, which has the following form for monovalent ions:

$$p_s = RT(C_c - 2C_o) \quad (3)$$

where R = gas constant; T = absolute temperature; and C_o = concentration of salt in the bulk pore water, mol/l.

The relationship between water content w and inter-particle spacing $2d$ is:

$$w = Sd/100 \quad (4)$$

where w = water content in weight percent; S = Surface area m^2/g ; and d = half-distance between parallel clay plates in Å.

YONG & WARKENTIN used the value $800 m^2/g$ for the specific surface area S when all the particles are parallel. This figure turns out to be somewhat high as can be concluded from LOW's study according to which S ranged between 485 and $800 m^2/g$ for 31 out of 36 natural montmorillonites. 16 out of 36 had values in the range of 700-800 m^2/g .

It follows from the preceding text that the application of this theory means that the swelling pressure is calculated as being produced by the repulsive forces, ignoring the influence of attractive van der WAALS forces. The calculated swelling pressure will therefore be too high but the error is known to be small at least at larger interparticle distances such as the ones corresponding to bulk densities lower than about $1.6 t/m^3$. As concerns the influence of anions,

their concentration between the clay particles can be neglected for densities higher than about 1.6 t/m^3 and pore water salinities lower than 0.001 M . This should confine the validity of the YONG & WARKENTIN theory to a bulk density interval of no particular interest as concerns the behaviour of highly compacted bentonite. As can be concluded from Fig. 6, however, the theory seems to fit the experimental results better at higher bulk densities, e.g. $1.9\text{-}2.1 \text{ t/m}^3$, and low salinities (curves B and C). High salinity and saturation with polyvalent cations (curve A) yield theoretical values which are much lower than those obtained in the experiments.

It was evident from the very start of the study that the derivation of theoretical expressions of the swelling pressure requires that the microstructural constitution is considered. At present, this represents great difficulties since the effect of a varying interparticle distance and orientation is not known and cannot yet be properly accounted for. The theory at small interparticle distances is of particular importance. Thus, it is obvious that when the average interparticle spacing ($2d$) is too small to allow for the formation of electrical double-layers, the theory cannot be valid. Theoretically, the critical value of $2d$ would be of the order of 5\AA , which means that neither this, nor any other theory based on electrical double layers is valid for water contents smaller than about 20%, e.g. when the bulk density exceeds about 2.05 t/m^3 . This is in fact proved by the observation that the pore water chemistry is not a determinant of the swelling pressure for $\rho > 2.05 \text{ t/m}^3$. The estimation of p_s for densities exceeding this value have to be made on an empirical basis, e.g. the diagrams of Figs. 6, 7, and 8. Very probably, higher p_s -values than the ones corresponding to $\rho = 2.1 \text{ t/m}^3$ will be reduced through stress relaxation, but there is no safe information as to the final, equilibrium stress or the involved time-dependence.

Homogeneity as a function of time and temperature

The successive redistribution of particles during and after the water uptake leading to complete water saturation, is a time-consuming process at room temperature. The major part of this redistribution, which leads to an increased degree of homogeneity, takes place within a few days or weeks. Complete equilibrium, however, may well require several weeks or months. During this period of time the permeability and diffusivity may be largely changed, while the swelling pressure may not be substantially affected. This is because the mean interparticle distance may stay practically constant after the development of the secondary swelling pressure maximum. Thus, a 10 months' test showed a practically constant swelling pressure after this event.

At 90°C a high degree of homogeneity is probably obtained much more rapidly than at room temperature. No actual proof of this is offered by any test but it is a reasonable conclusion based on the fact that elevated temperatures yield lower swelling pressures. This reduction may well be explained by less stable interlayer and interparticle water lattices when the temperature increases. This means that displacements of particle aggregates and individual particles take place more easily, and that the migration and flow of water associated with altered interparticle distances is more rapid.

CONCLUSIONS

A number of previous questions have been answered by the study and the swelling pressure characteristics of montmorillonite-rich bentonites can now be considered as sufficiently well-known for practical purposes.

The most important observation is that the pore water chemistry is hardly a determinant of the swelling pressure in the bulk density interval of primary interest for the KBS 2 concept ($\rho > 2 \text{ t/m}^3$). This eliminates the risk of a largely reduced pressure associated with pore water electrolyte changes. Similarly, the kind of initially adsorbed cation(s) does not affect, largely, the swelling pressure at higher bulk densities, which means that a large variety of bentonites are of potential use as highly compacted buffer materials.

The influence of the temperature on the swelling pressure has to be taken into consideration in the design of pressure-affected parts of deposition plants. For 70°C , a 30-50% reduction of the pressure at room temperature is expected.

The theoretical deduction of the swelling pressure was not successful mainly because a correct physical model has not yet been developed. For practical purposes empirical relationships should be sufficient. The average swelling pressure in MPa of bentonite saturated with "salt" water and "fresh" water, respectively, can be determined from:

$$p_s = e^{11.5(\rho-1.87)} \quad (\text{"salt"}) \quad (5)$$

with the boundary conditions $1.80 < \rho < 2.1 \text{ t/m}^3$
and

$$p_s = e^{11.5(\rho-1.81)} \quad (\text{"fresh" water}) \quad (6)$$

with the boundary conditions $1.75 < \rho < 2.1 \text{ t/m}^3$

These expressions are represented by the upper and lower boundaries of the hatched zone in Fig. 10. Most of the recorded swelling pressures fall within this zone. A temperature increase to 70°C reduces the pressure to approximately 50% of the value at 20°C .

Higher densities than 2.1 t/m^3 produce higher swelling pressures but they are likely to be reduced due to stress relaxation although this is probably a very slow process.

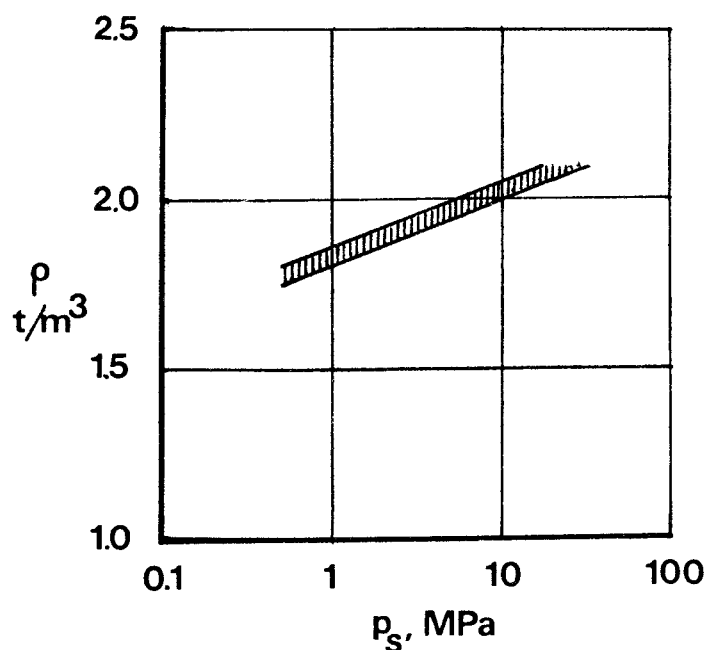


Fig. 10 Approximate relationship between swelling pressure and bulk density at complete saturation at 20°C .

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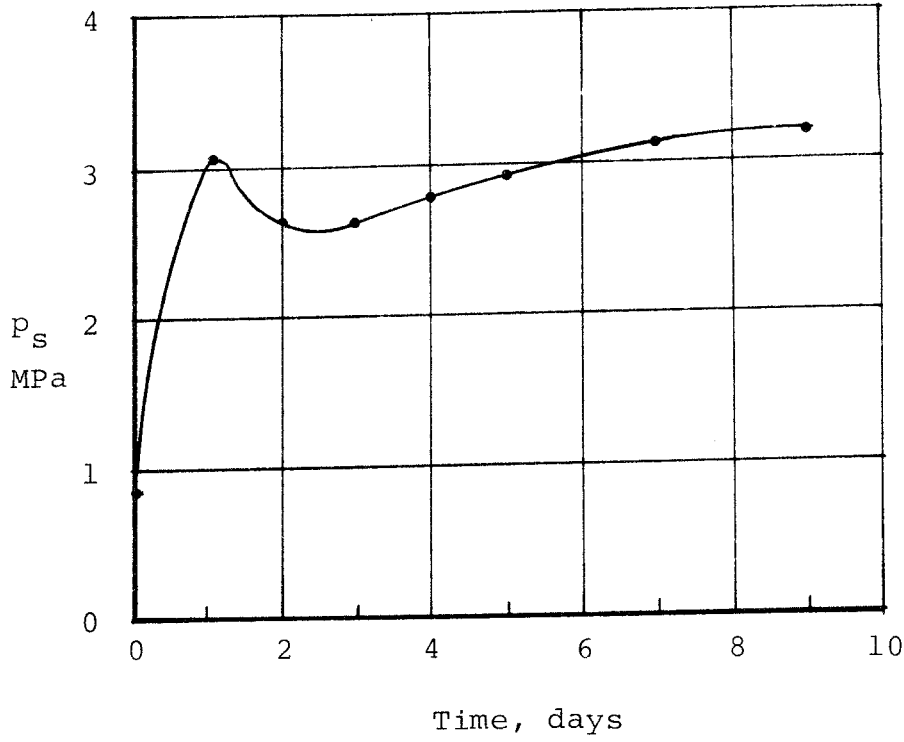
APPENDIX

Selection of records of swelling pressure versus time. The swelling pressure was measured with rather short intervals in these tests, while longer intervals were applied in the majority of the test series.

Legend:

ρ_m = Bulk density in water-saturated condition

height = Vertical extension of sample exposed to water at both ends



Test 44

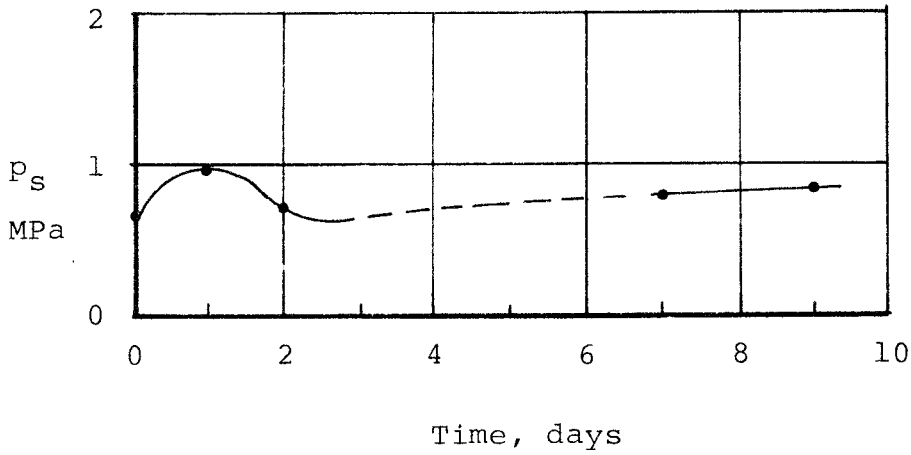
MX-80

$\rho_m = 1.98 \text{ t/m}^3$

NaCl-sol.

20°C

2.1 cm height



Test 45

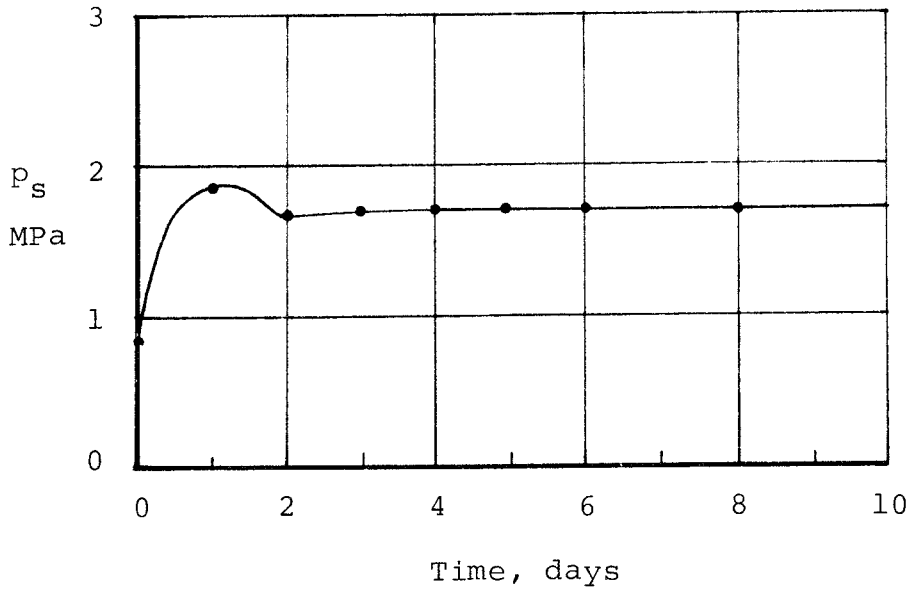
MX-80

$\rho_m = 1.84 \text{ t/m}^3$

NaCl-sol.

20°C

2.2 cm height



Test 46

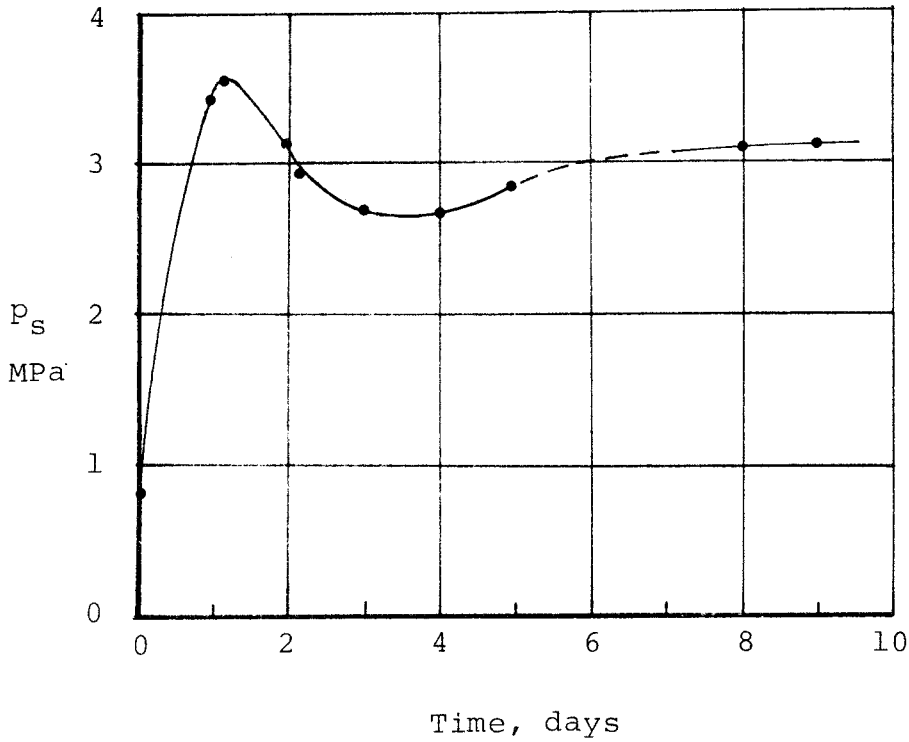
MX-80

$\rho_m = 2.01 \text{ t/m}^3$

NaCl-sol.

90°C

2.2 cm height



Test 51

MX-80

$\rho_m = 1.99 \text{ t/m}^3$

CaCl₂-sol.

20°C

2.2 cm height

Test 57

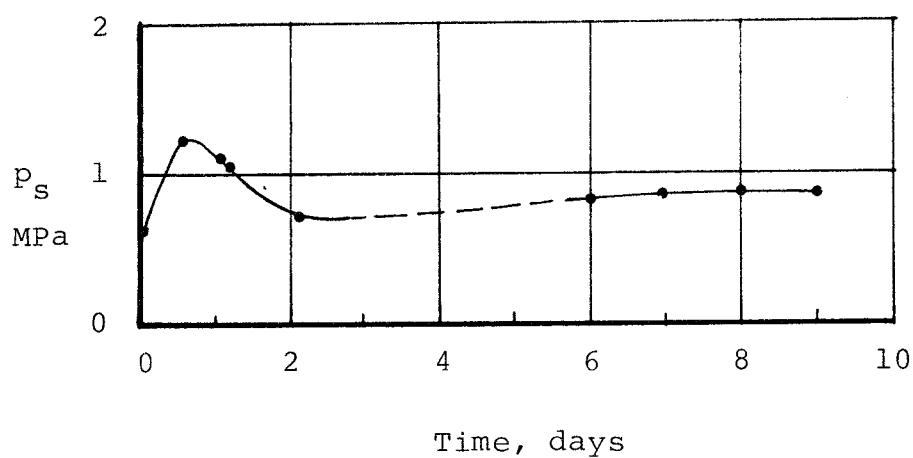
MX-80

$$\rho_m = 1.87 \text{ t/m}^3$$

CaCl₂-sol.

20°C

2.2 cm height

Test 63

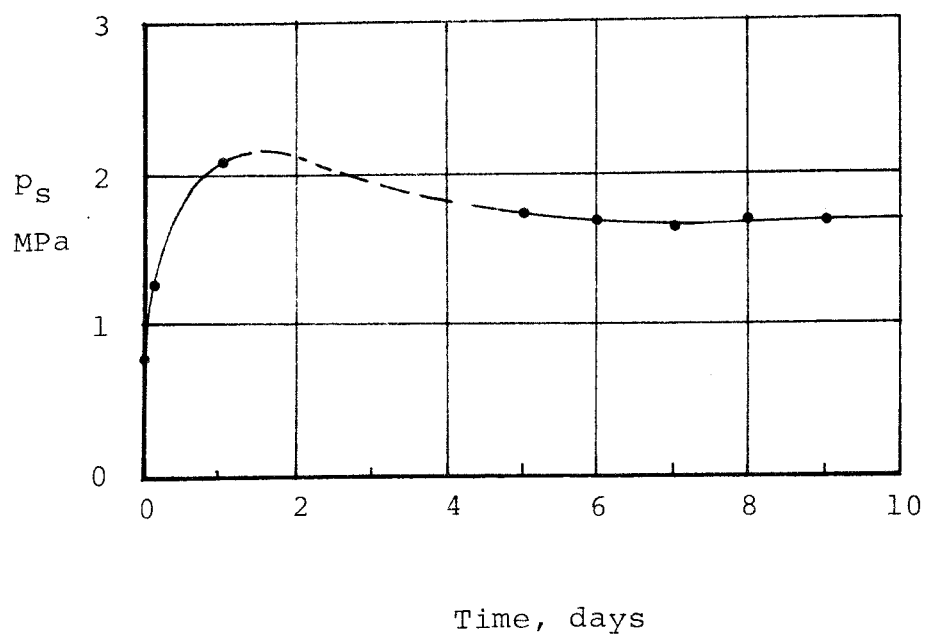
MX-80

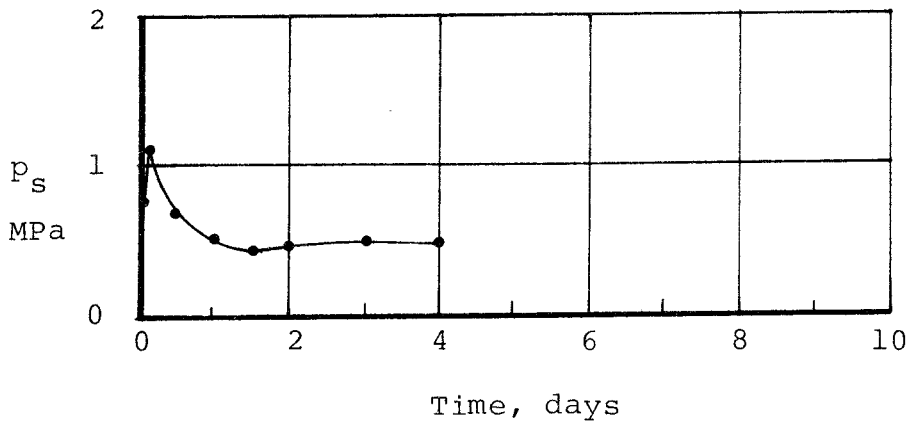
$$\rho_m = 1.99 \text{ t/m}^3$$

CaCl₂-sol.

90°C

2.2 cm height





Test 64

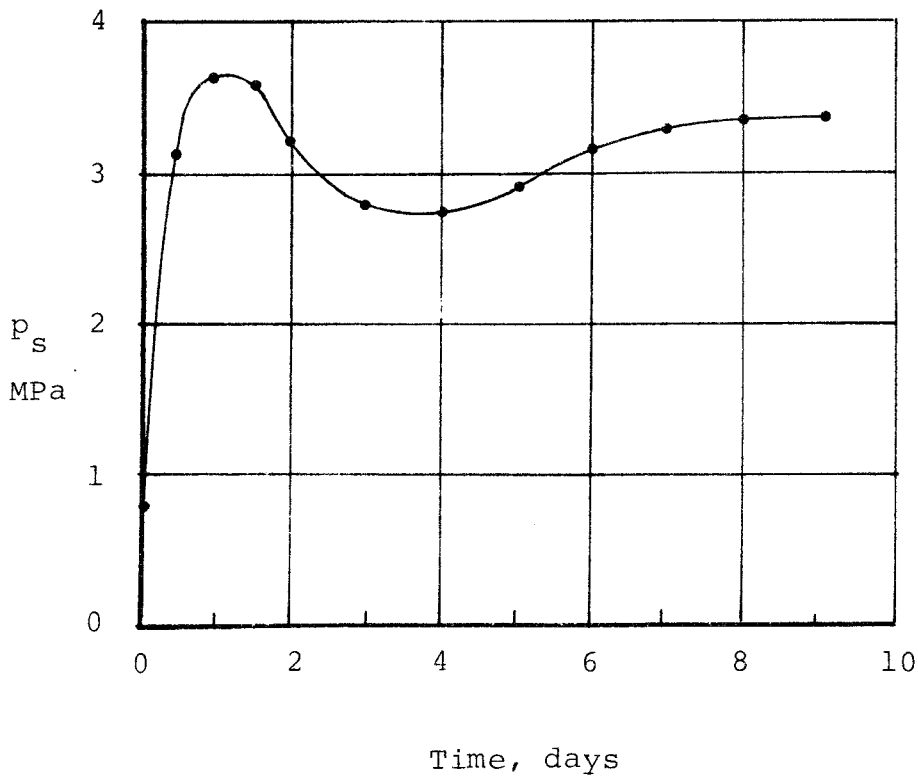
MX-80

$\rho_m = 1.96 \text{ t/m}^3$

CaCl₂

90°C

2.2 cm height



Test 70

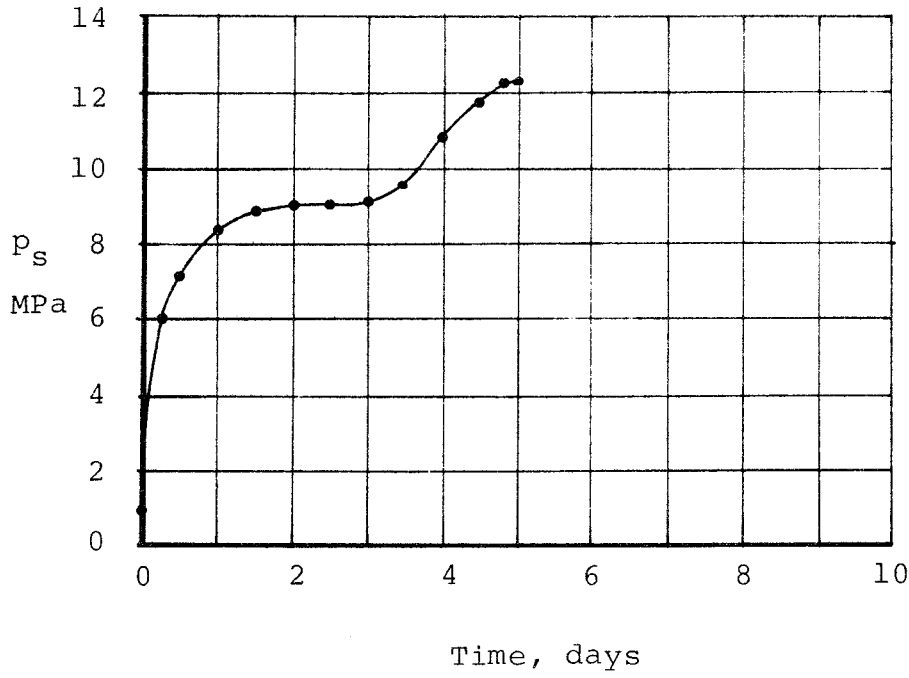
MX-80

$\rho_m = 2.00 \text{ t/m}^3$

CaCl₂

20°C

2.2 cm height



Test 73

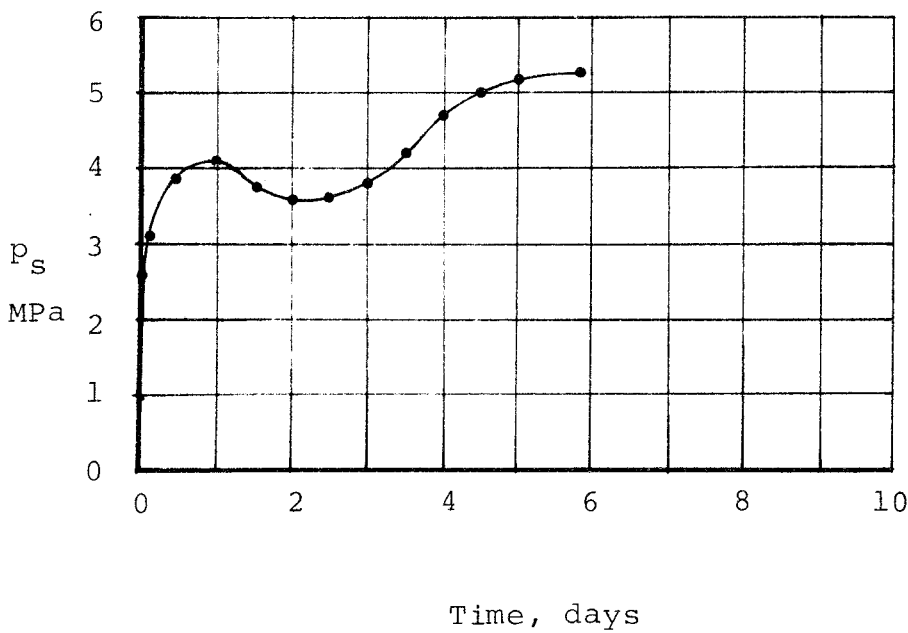
MX-80

$\rho_m = 2.10 \text{ t/m}^3$

Dist.water

20°C

2.1 cm height



Test 76

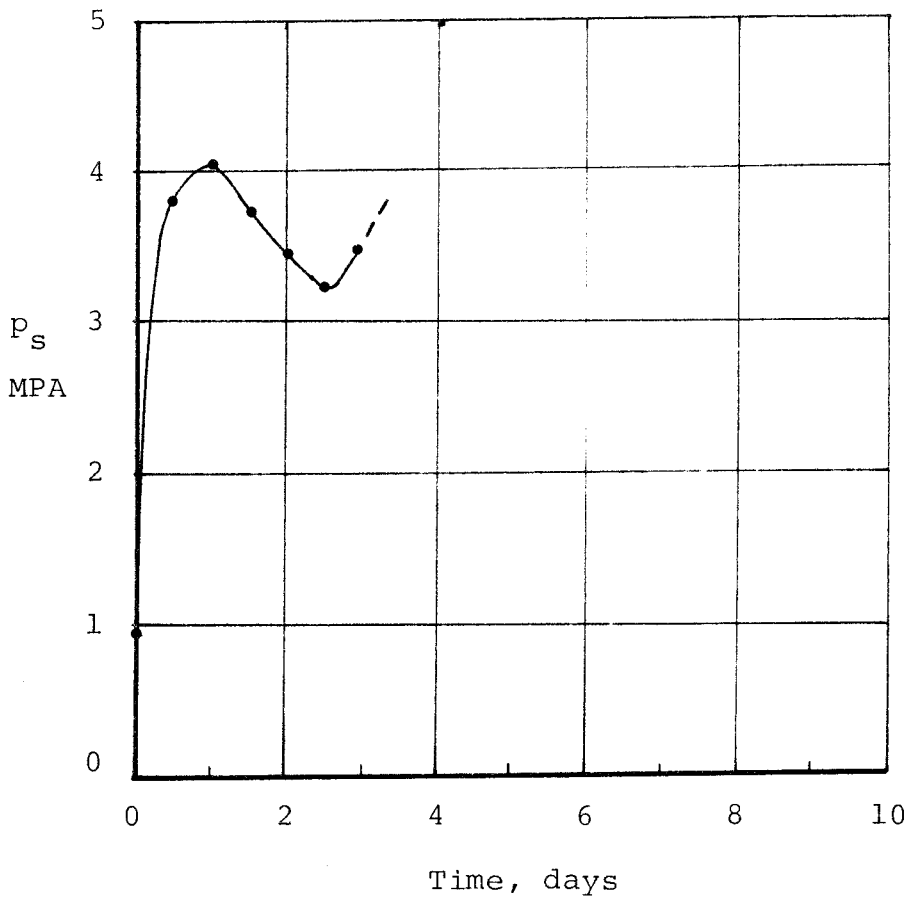
MX-80

$\rho_m = 2.00 \text{ t/m}^3$

Dist.water

20°C

2.0 cm height



Test 77

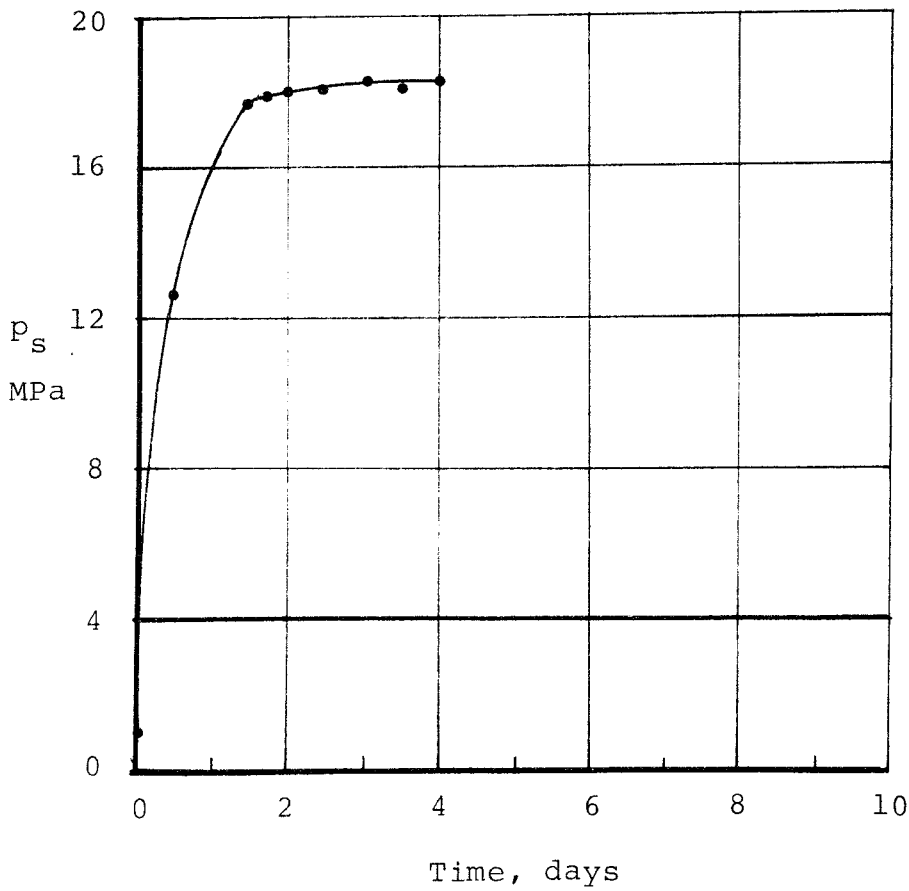
MX-80

$\rho_m = 2.01 \text{ t/m}^3$

Dist.water

20°C

2 cm height



Test 81

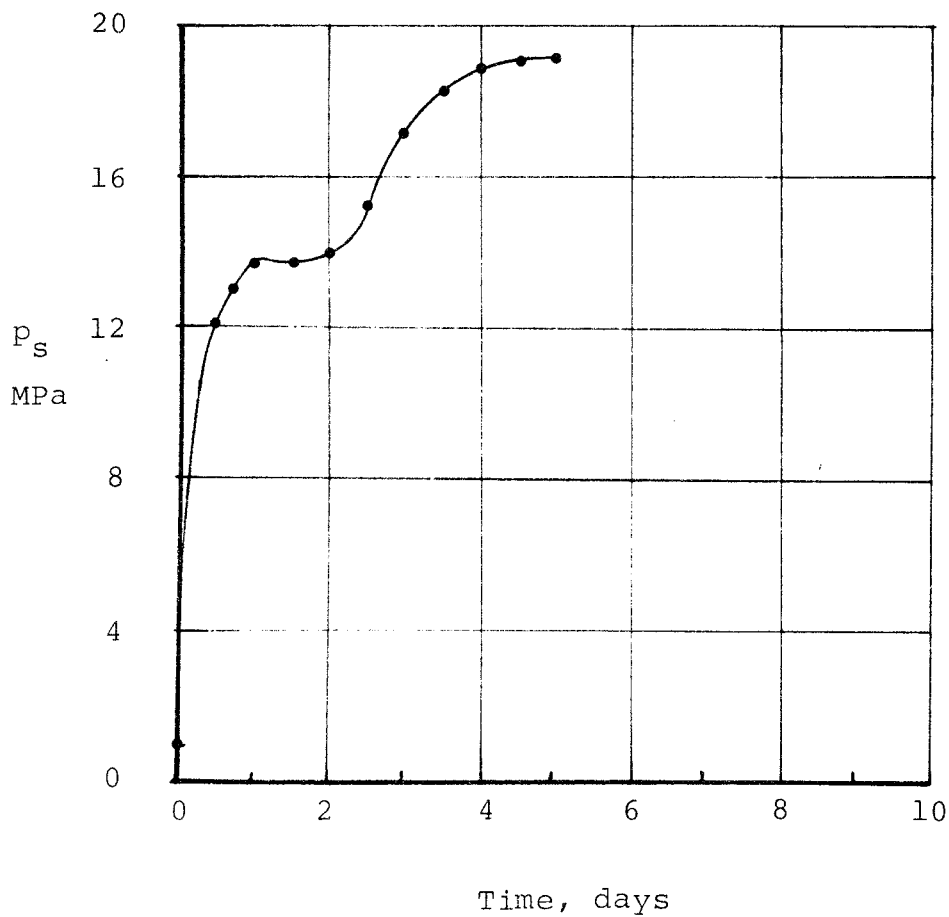
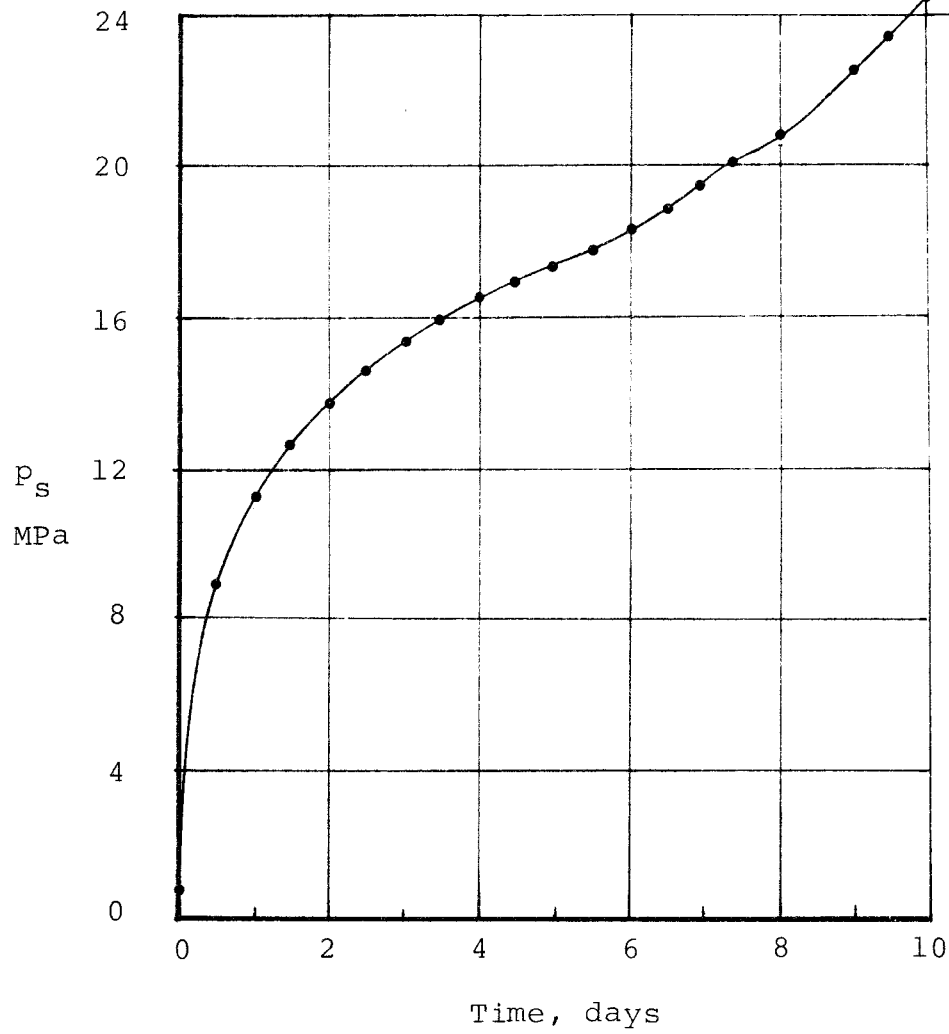
MX-80

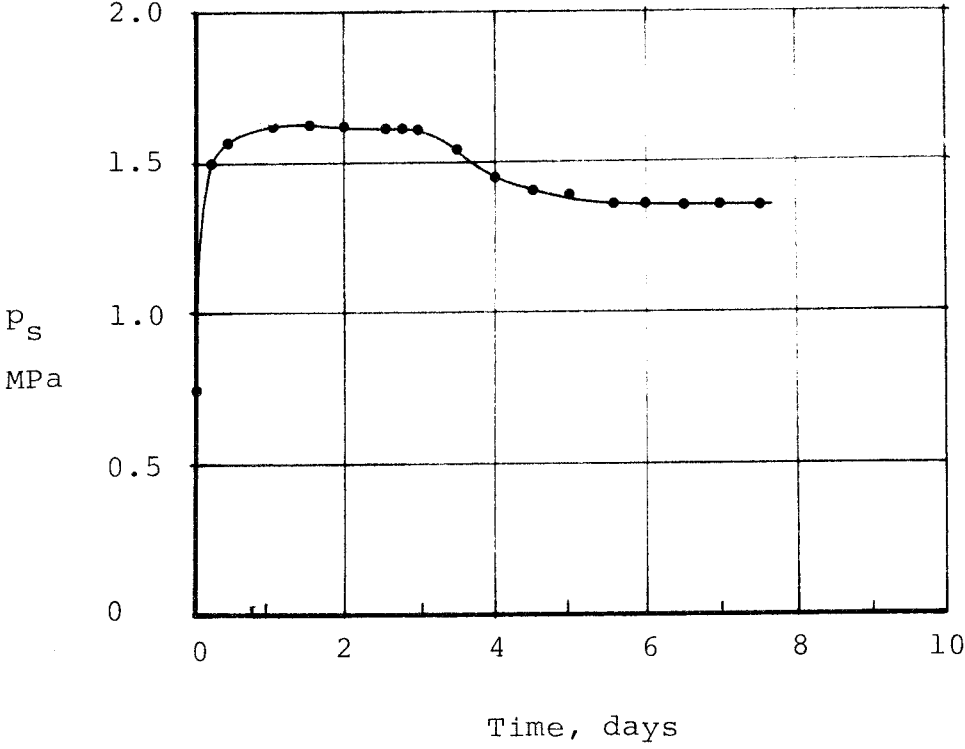
$\rho_m = 2.13 \text{ t/m}^3$

Dist.water

20°C

1 cm height





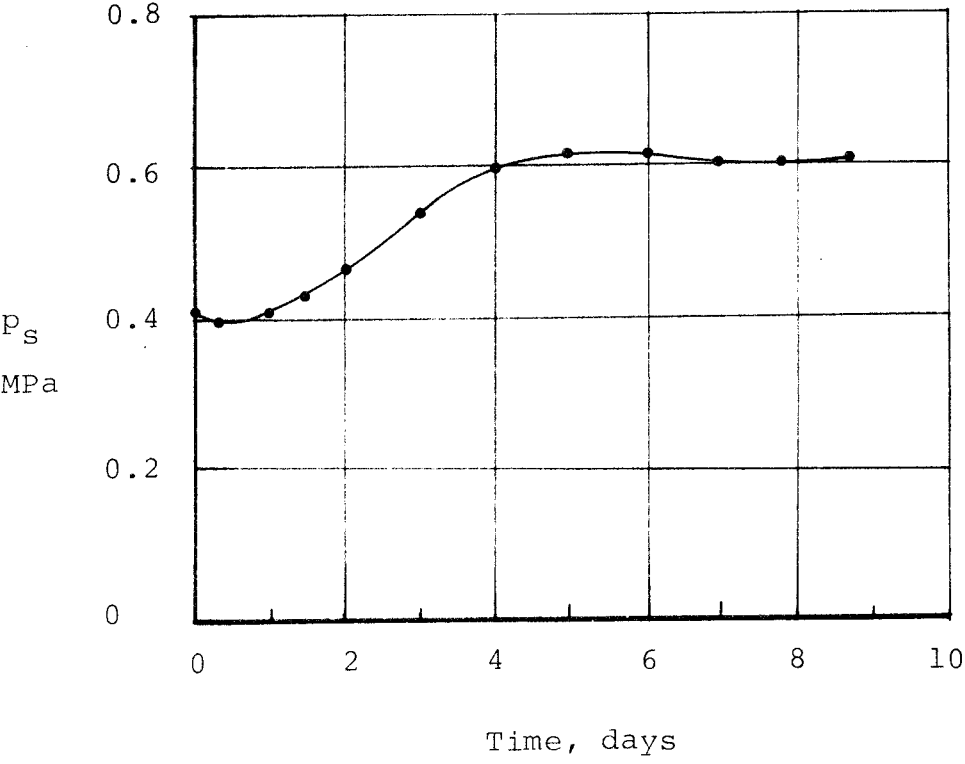
Test 87

MX-80

$\rho_m = 1.89 \text{ t/m}^3$

Dist.water

20°C



Test 91

MX-80

$\rho_m = 1.79 \text{ t/m}^3$

Dist.water

20°C

2.0 cm height

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