SKBF KBS TEKNISK RAPPORT
83-46

Use of clay as buffer in radioactive repositories

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Luleå May 25 1983

SVENSK KÄRNBRÄNSLEFÖRSÖRJNING AB / AVDELNING KBS

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USE OF CLAYS AS BUFFERS IN RADIOACTIVE REPOSITORIES

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DIV. OF SOIL MECHANICS
UNIVERSITY OF LULEÅ

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SUMMARY

The major physico/mechanical functional criteria which are specified for the three main applications of clay material in repositories, i.e. as canister "overpacks", as "plugs" in boreholes, shafts and tunnels, and as "backfills" in tunnels and shafts, are a low hydraulic conductivity and a swelling ability. The most suitable clay mineral is Na montmorillonite but illite may be accepted under certain circumstances although it does not swell readily. Illite-rich, natural clays are rare but a particular kind of late-glacial sedimentary clay is available in large quantities in Sweden.

Na montmorillonite is the dominant clay mineral in a few bentonites which are exploited commercially, but suitable sources can also be natural Ca bentonites which are converted to the Na state by ion exchange on an idustrial scale. Such bentonites are commercially available in bulk or bagged form from several plants in Europe and elsewhere. Refining of illite, on the other hand, requires the establishment of a Swedish plant for the production of sufficient quantities of powdered clay.

For use as canister "overpack", highly compacted bentonite is superior to illite and any reasonably montmorillonite-rich bentonite will do which is not too rich in sulphur. The organic content should be low and heat treatment may be required to bring this content down to an acceptable level. Heating to slightly more than 400 °C does not affect the physical properties of neither montmorillonite, nor illite to a significant extent. Bentonite is also very suitable for use as sealing plugs in the form of highly compacted blocks.

For use as backfill in tunnels and shafts, illitic clay is a candidate material which can be compacted on site to the rather high density that is required. Where a swelling capacity is needed, such as in the top part of tunnels, bentonite-based backfills are suitable and if Na saturated clay is used the bentonite fraction can be kept low. Thus, a 10 % content of Na bentonite by weight should generally be sufficient for a well compacted mixture with respect to the required hydraulic conductivity, while a 20-30 % content may be needed to arrive at a sufficient swelling power.

The choice of a suitable clay material requires that the substance be properly characterized and tested. It is concluded that rather rigorous analyses are necessary as concerns "overpacks", including mineralogical and granulometrical tests and the determination of the swelling characteristics as well as of certain chemical features. For backfills and for the current checking of all sorts of clay for use as buffer materials, the natural water content, the liquid limit and the swelling ability have to be determined, since they are the fingerprints of this type of soil.

The report ends with a tentative scheme for the possible use of various clay materials in repositories.

1. SCOPE OF STUDY

Much of the KBS-related clay buffer investigations refer to the use of MX-80, a commercial bentonite powder which stems from natural bentonite beds in Wyoming and South Dakota, USA. This material, which is rich in Na montmorillonite, is available in large quantities and is therefore of potential use in repositories. It is obvious, however, that other smectitic clays may be equally or sufficiently useful and considerably cheaper, the ultimate choice of clay material actually being a matter of optimum. Even quite different clay types, such as illitic sediments, may qualify and mixtures of smectite and ballast will certainly be useful for a number of applications.

This report presents a short review of the main properties of various clay and clay mixture candidates to serve as an aid in the search for suitable materials and to give an indication of the expected change in physical behavior in the event of mineral alteration, such as illitization of the smectite components of bentonite. More specifically, the following items will be dealt with here:

- Physico/mechanical criteria for use in repositories
- Physico/mechanical properties of certain candidate materials
- Characterization of clay materials
- Effect of addition of ballast (rock-forming minerals) on the properties of clays

2. PHYSICO-MECHANICAL CRITERIA

2.1 General

The choice of clay barriers for repositories, including those for disposal of low and medium level wastes, must be based on functional criteria. For the KBS concepts, such criteria were related to three main applications of clay technology: 1) near-field shielding of canisters with high level wastes, 2) backfilling of shafts and tunnels, as well as of the the space between rock and concrete silos with low and medium wastes, and 3) plugging of boreholes, shafts and tunnels. In the course of the work, other soil properties than purely physico/mechanical and chemical ones became important as well, and the present requirements therefore go beyond the initial specifications. Updated versions are given below for each application.

2.1.1 Near-field shielding of canisters in boreholes

The functional requirements of canister-embedding buffer materials, sometimes termed "overpack", are the following (cf. Fig.1):

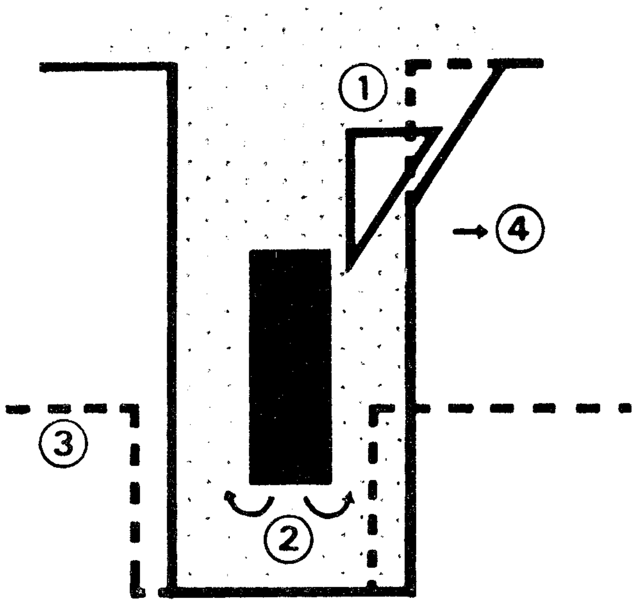
1a. Chemical stability to ensure sufficient function within one million years

1b. Sufficient strength to prevent rock fragments from falling, and to minimize canister settlement

1c. Sufficient ductility to dissipate stresses induced by slight rock displacements caused by tectonics etc

1d. Low hydraulic conductivity to minimize groundwater flow through the embedding substance

1e. Low diffusivity to retard ion migration from the rock as well as from the canisters



1,2,	Stiffness required
3	Softness required
4	Low flow and diffusion rates required

Fig.1. Schematic section through deposition hole with canister

1f. Sufficient heat conductivity to keep the temperature below any level that is critical to the embedding substance or the canisters

1g. Certain swelling potential to make the embedding substance fill up the space and establish a close contact with the rock and the canisters

1h. Easy access, from domestic sources if possible, and minimum industrial processing

1i. Suitable form for easy application without requiring highly specialized workers. This includes unexpected delay in ongoing deposition work and extreme climatic conditions due to ventilation breakdown, i.e. high temperature and humidity

1j. Minimum cost for preparation and application

In addition, it is considered to be valuable if the embedding substance has the ability of retaining radionuclides emerging from the canisters, through ion exchange mechanisms. Since this ability is not required until a late stage of canister corrosion has been reached, i.e. after several million years for the KBS 3 concept, the corrosion-retarding properties - such as a very low hydraulic conductivity - are ranked much higher.

2.1.2 Backfilling

The functional requirements of far-field buffer materials, i.e. backfills in tunnels and shafts (cf. Fig.2), are less rigorous than those of the canister-embedding substance. The major ones are:

2a. Chemical stability to ensure sufficient function within the operative lifetime of the repository: one million years for high level wastes, and a few thousand years for low and medium level wastes

2b. Sufficient strength and swelling potential of the backfill to prevent shallow rock from roofs and walls to separate from the rock mass, and to prevent settlement of the backfill under its own weight.

2c. Sufficiently low compressibility to yield small displacements at the interface between the backfill and "overpacks" which produce high swelling pressures

2d. Sufficient erosion resistance to prevent noticeable piping in the early stages of backfilling, i.e. when the hydraulic gradients are high

2e. Low hydraulic conductivity. For the backfill in tunnels with deposition holes it should not exceed that of the rock

2f. equal to 1h

2g. equal to 1i

2h. equal to 1j

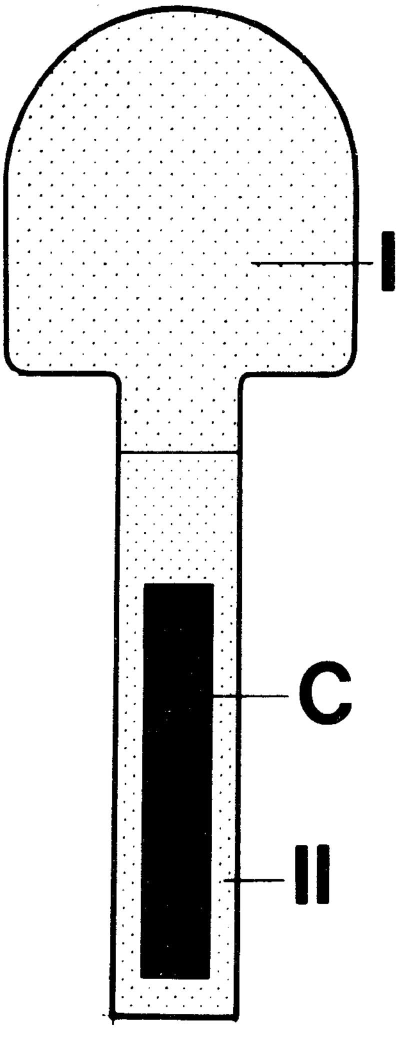


Fig.2. Schematic picture of tunnel with deposition hole. I is the backfill, II the overpack, and C the canister.

A major part of the backfill should be available at or close to mined repositories; preferably, crushed rock ballast from the excavation activities should be used, at least in the shafts and in tunnels with no deposition holes.

2.1.3 Borehole, shaft-, and tunnel plugging

Boreholes as well as tunnels and shafts may act as short circuits for groundwater driven by regional hydraulic gradients if they are not properly sealed. But even if the backfill is effective and has a lower hydraulic conductivity than the host rock, shafts and tunnels may still serve as water conduits because of the disturbed zone that is created in the rock mass adjacent to the penetration as a result of excavation (1). Boreholes, tunnels and shafts are also expected to traverse high conductivity rock zones, which may produce large changes in the hydraulic regime of the rock and bring water with less desirable chemical composition into the tunnels with deposition holes.

Effective plugging is required to seal off the penetrations locally, which may involve the cutting of slots (Fig.3), or over their entire length (2). Functional requirements also have to be specified, the major ones being the same as those of the tunnels and shafts, with the exception that the technique for application may require rather sophisticated but still very practical methods.

2.1.4 Conclusions

If the various criteria are put together we see that the most crucial physico/mechanical properties are the stress/strain/time behavior, the swelling potential, the hydraulic conductivity, and the thermal conductivity. It was concluded early in the Swedish search for suitable buffer substances, that clays or clay-based materials would fulfil the requirements. Smectites were preferred from the very start of the investigation, but the limited domestic resources has called for a closer examination of other clay types as well. Also, chemical alteration of smectite may yield other products in the repository environment and their behavior must of course be known. The final choice may also well involve mixing of several components or the use of natural soils of low purity, and it is therefore required that the properties of certain individual, relevant clay minerals are examined to begin with.

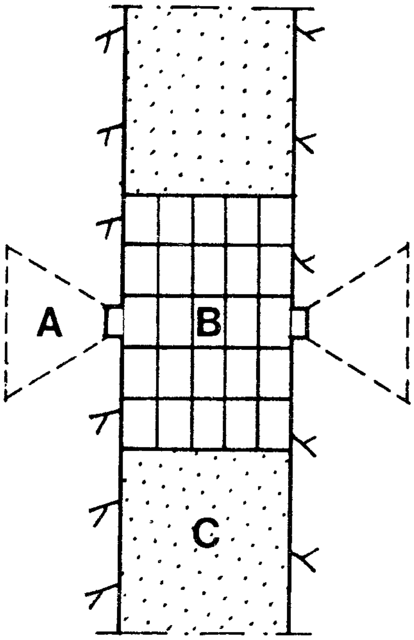


Fig.3. Shaft or tunnel plugging. A) Grouting, B) Effective, expansive sealing in the form of highly compacted smectite-rich clay, C) Backfill (3).

3. PHYSICO/MECHANICAL PROPERTIES OF CLAY CANDIDATES

3.1 Clay minerals

The main clay minerals are phyllosilicates forming crystals smaller than about 2-5 micrometers, with a structure based on composite layers of tetrahedrally and octahedrally coordinated cations. We will distinguish here between three main groups:

- Kandites (main minerals: kaolinite and halloysite)
- Illites (main minerals: illite/hydrous mica and glauconite)
- Smectites (Main minerals: montmorillonite, nontronite and beidellite)

They have a very wide occurence which makes them qualify as candidate minerals. Their chemistry, crystallographic characteristics etc. are described in a vast amount of literature to which the reader is referred. A useful summary has been given by CHAPMAN (3), whose characterization is applied here.

3.1.1 Kandites

The clay minerals belonging to this group have a very low layer charge and therefore a low cation exchange capacity. The crystal sheets are non-symmetric from a structural point of view, and adhere strongly to each other so that a kaolinite particle, for instance, usually consists of hundreds of sheets forming a stack, which is almost silt-sized. This means that the specific surface area is small and that these clay minerals adsorb very little water, which in turn yields a low plasticity and a low liquid limit, i.e. of the order of 20 - 40 %. If water is added to a water saturated sample in excess of the liquid limit, the clay turns semi-liquid like a silt. It does not show any swelling properties.

With the exception of halloysite, which is a rather rare mineral, kandites hold only small amounts of strongly adsorbed water and their microstructural arrangement yields large continuous passages for water, which is manifested by a much higher hydraulic conductivity than of clays dominated by the other clay minerals at one and the same bulk density.

Even in a very slightly compacted state, corresponding to a bulk density of 1.6 - 1.8 t/m^3 kandite clays have a high bearing capacity and a very low compressibility. The creep in shear is less obvious than that of illite- or smectite-rich clays.

Summing up, it can be stated that the mediocral properties of kandites as buffer materials are not balanced by the few valuable ones. Kandite clay should therefore not be considered as a candidate material. It should be mentioned that the (compact) theoretical density of kaolinite is 2.609 t/m^3 while measured values vary from 2.60 to 2.68 t/m^3, with 2.63 t/m^3 frequently quoted in mineralogical textbooks. Formation of kandites is favored by acid conditions.

3.1.2 Illites

Basically, illites are closely related to the micas and merge into that group by cation substitution. Their structure (Fig.4) is regarded as a condensed version of smectite through the establishment of a firm sheet-to-sheet coupling that is caused by potassium ions in interlayer positions. The crystal structure of illite is usually not very well defined; large variations in composition occur, such as interstratification of discrete lamellae of other silicates, and there are only a few known localities with larger quantities of reasonably uniform illite.

Illites are characterized by a cation exchange capacity of about 30 - 40 mE/100 g dry clay. Its ability to hydrate is intermediate to that of kandites and smectites and this is manifested by a liquid limit of 70 -90 % of finegrained, organic-free specimens.

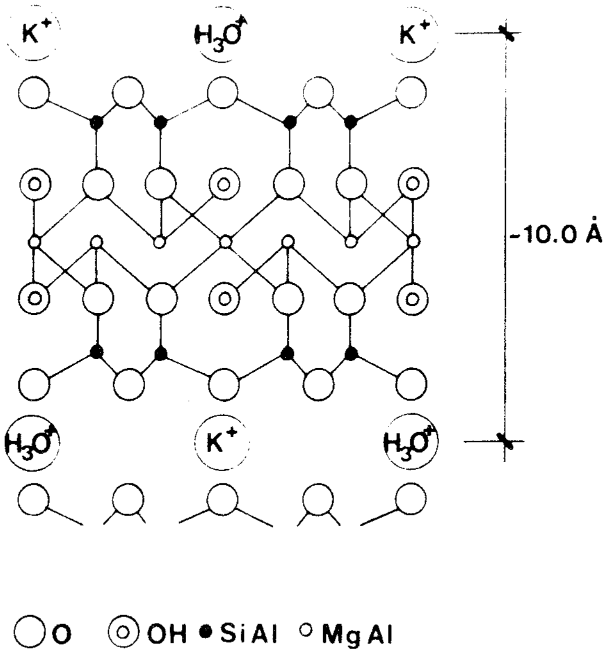


Fig.4. Schematic illite crystal structure.

The physical properties of illitic clays are largely determined by their characteristically high content of clay crystallites of colloidal size. This feature, which is illustrated by the histograms in Fig.5 of a typical natural illitic clay, may yield specific surface areas well over 100 m^2/g dry clay (4). The large fraction of colloidal mineral matter creates a strongly aggregated microstructure through the mobility and flocculating power of its minute crystals when such clays are formed in nature (Fig.6). The tortuosity with respect to water permeation is therefore considerable and the hydraulic conductivity consequently low.

Another property derived from the vast number of colloidal particles is the obvious thixotropic strength regain of such clays after remoulding (5).

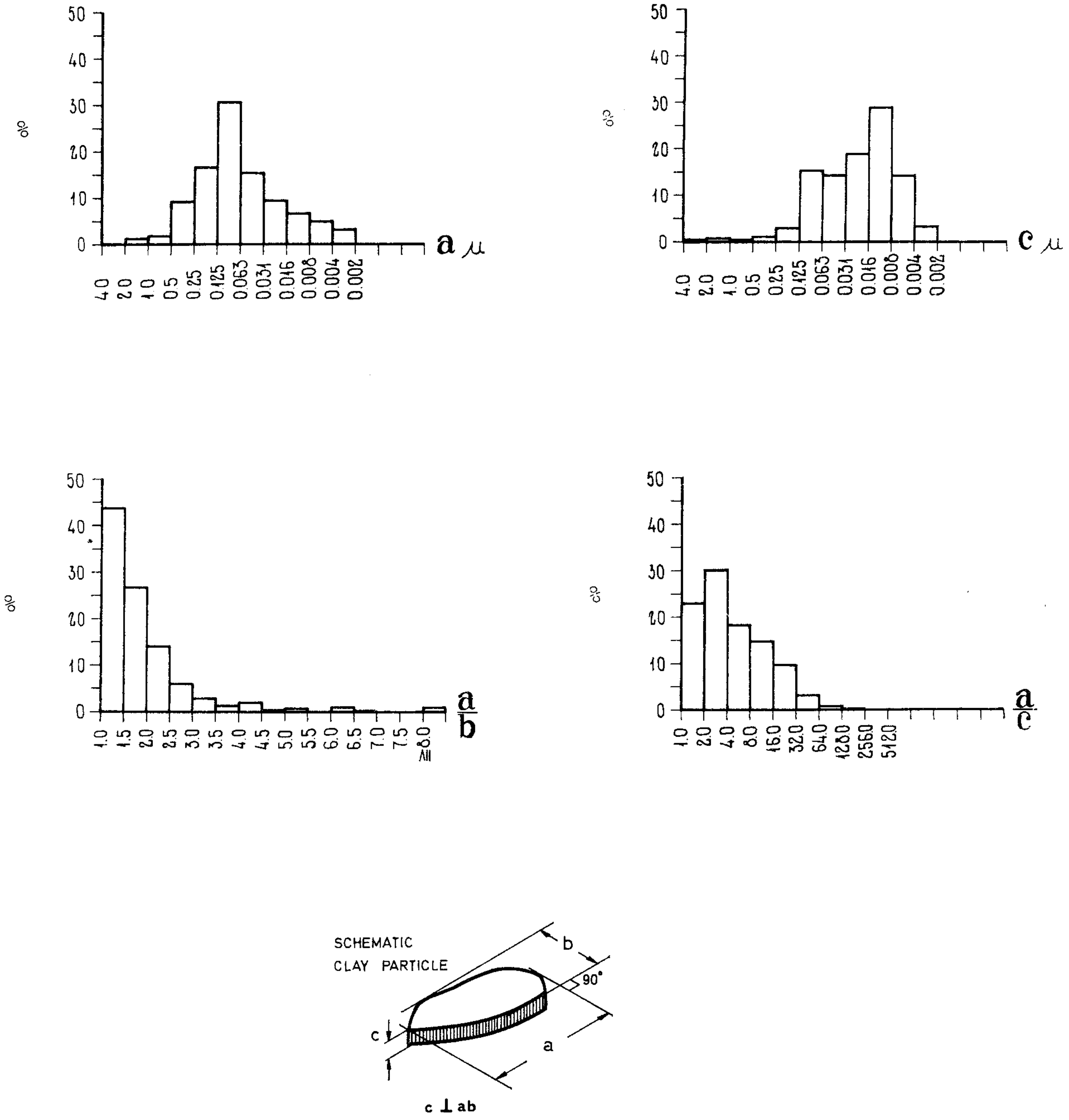


Fig.5. Clay particle size and shape in illitic clay.

The aggregated character of illitic clays yields a highly collapsible microstructure, which is manifested by the very obvious loss in shear strength by remolding, and by a high compressibility at low and moderate bulk densities. Turning then to microstructural details, the arrangement of interacting particles is found to be of particular significance with respect to the swelling potential. In the natural state of soft and moderately dense illitic clays, the majority of the interacting particles seem to be coupled edge-to-edge or edge-to-face. Shearing as well as compression yields comprehensive microstructural breakdown, by which domains, i.e. oriented, face-to-face grouped flaky particles are created (Fig.7). Here, equally charged, hydrated mineral surfaces are brought close to each other, their repulsion giving rise to expansion of the domains and to macroscopic swelling. If it is hindered, a swelling pressure will be exerted on the confinement. Experience tells that the swelling is very moderate compared to that of smectites, which is explained by a stronger hydration and different charge properties of the latter.

The (compact) density of illite with no adsorbed water has been found to range between 2.64 to 2.69 t/m. In room atmosphere with 50 % relative humidity, the hygroscopic water content is usually less than 5 %.

In the present context it should be mentioned that illite may be formed from smectites through charge change and potassium uptake. For this, heating to at least 100 C and subsequent percolation of potassium-holding water, or diffusion of potassium ions, seem to be required (6).

Summing up, we can conclude that the mechanical properties in terms of shear strength and stress/strain/time behavior may be suitable for certain practical applications, and that the hydraulic conductivity and swelling properties may also be acceptable. Illitic clay therefore deserves to be ranked as a candidate buffer material.

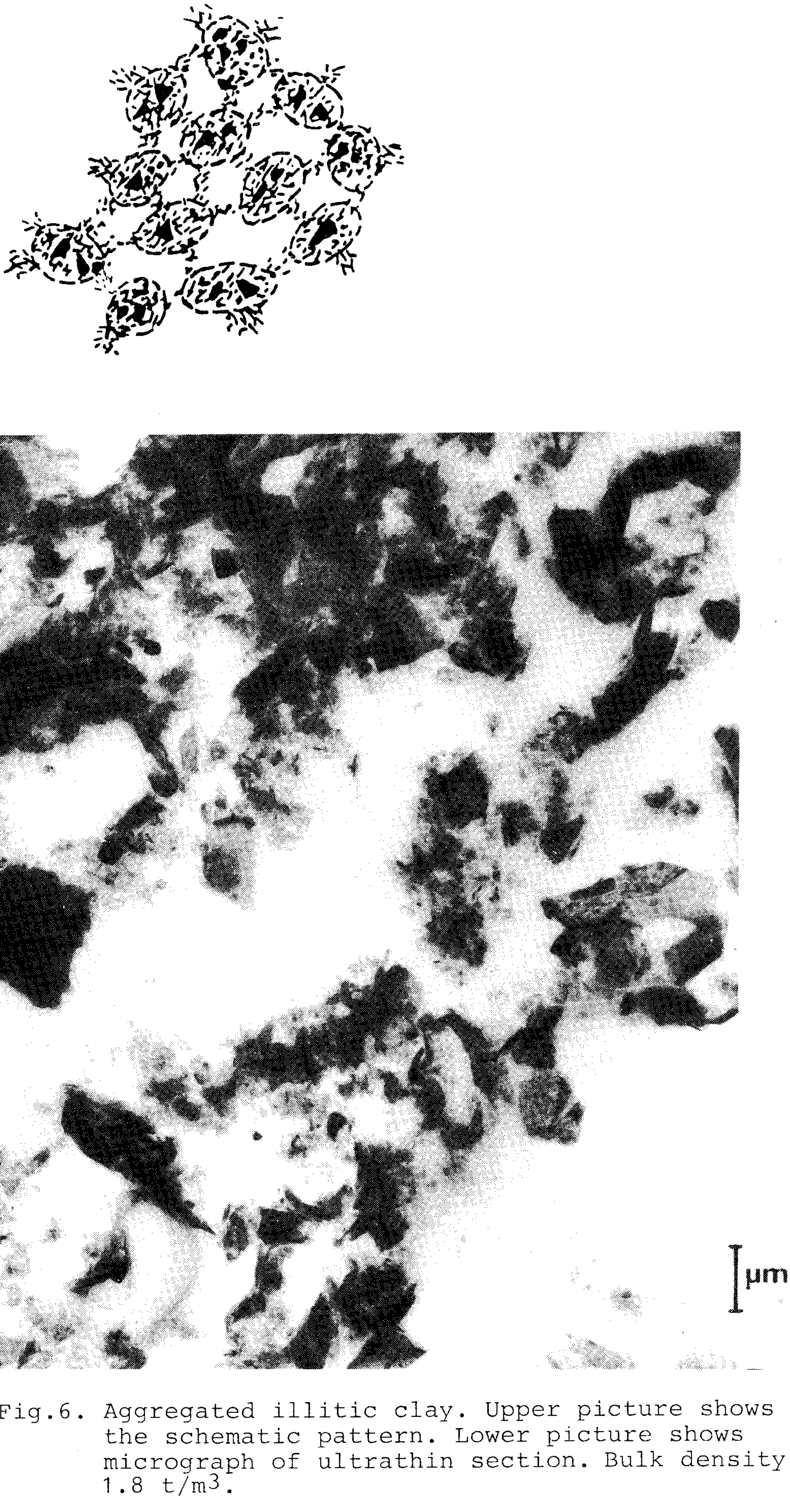


Fig.6. Aggregated illitic clay. Upper picture shows the schematic pattern. Lower picture shows micrograph of ultrathin section. Bulk density 1.8 t/m^3.

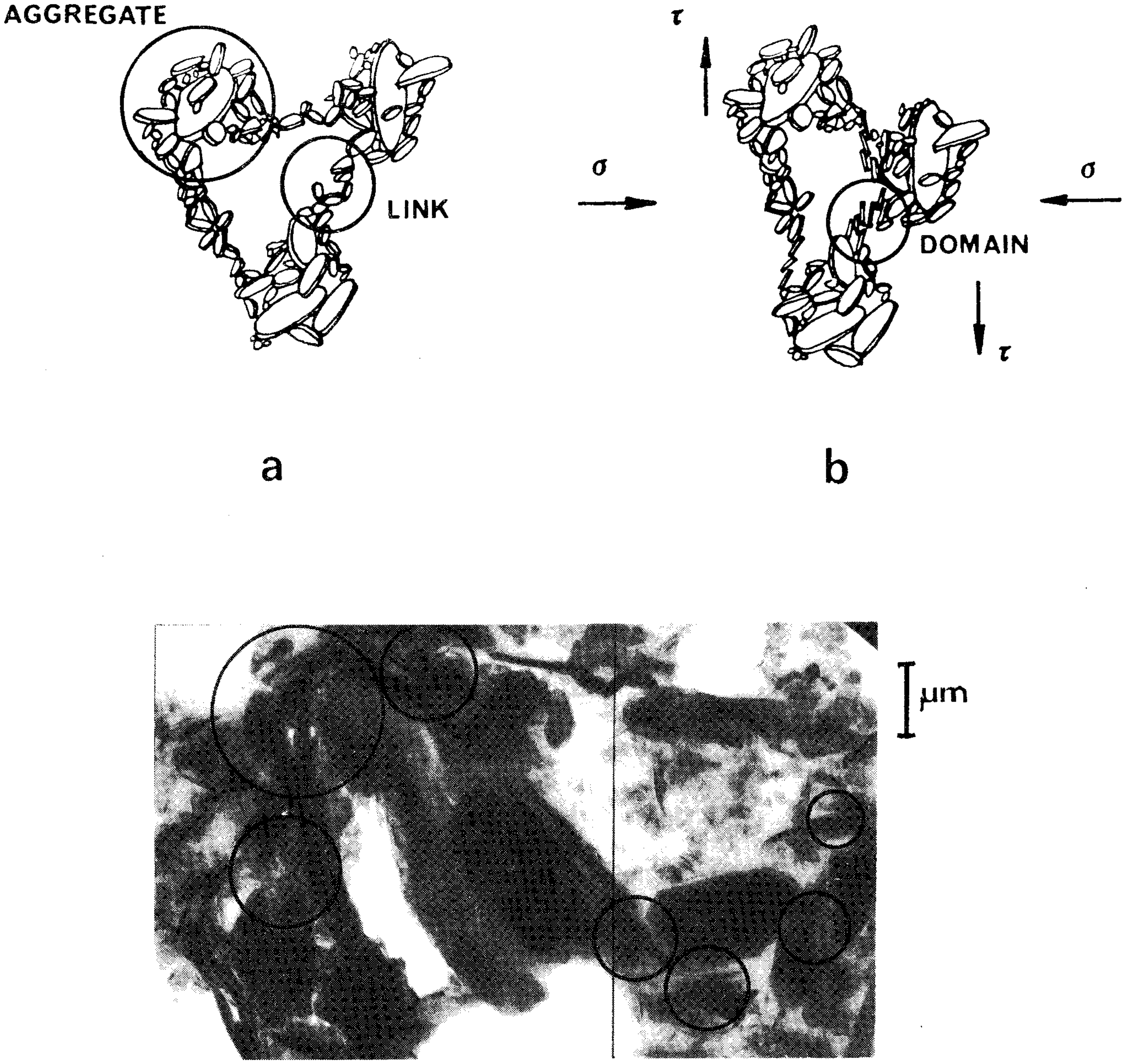


Fig.7. Structural breakdown through shear and/or compression.

Upper picture: a) Natural aggregated pattern
b) Links converted to domains by overstressing

Lower picture: Electron micrograph of ultrathin section of illitic clay. Domains (encircled) formed by compression of soft clay at 1.3 MPa.

3.1.3 Smectites

Smectites have an octahedral sheet coordinated with two tetrahedral sheets in which oxygens are shared. Cationic substitution occurs in octahedral as well as tetrahedral sheets, which yields different properties and forms the basis of classification: montmorillonite with Si in tetrahedral positions and Al and Mg in octahedral sites, beidellite with Si and Al in tetrahedral positions and Al in octahedral sites, and nontronite with Si and Al in tetrahedral positions and Fe in octahedral sites. In practice, most smectites have compositions that deviate from the idealized versions. Montmorillonitic minerals are the most common smectites, their crystal structure being illustrated in Fig.8.

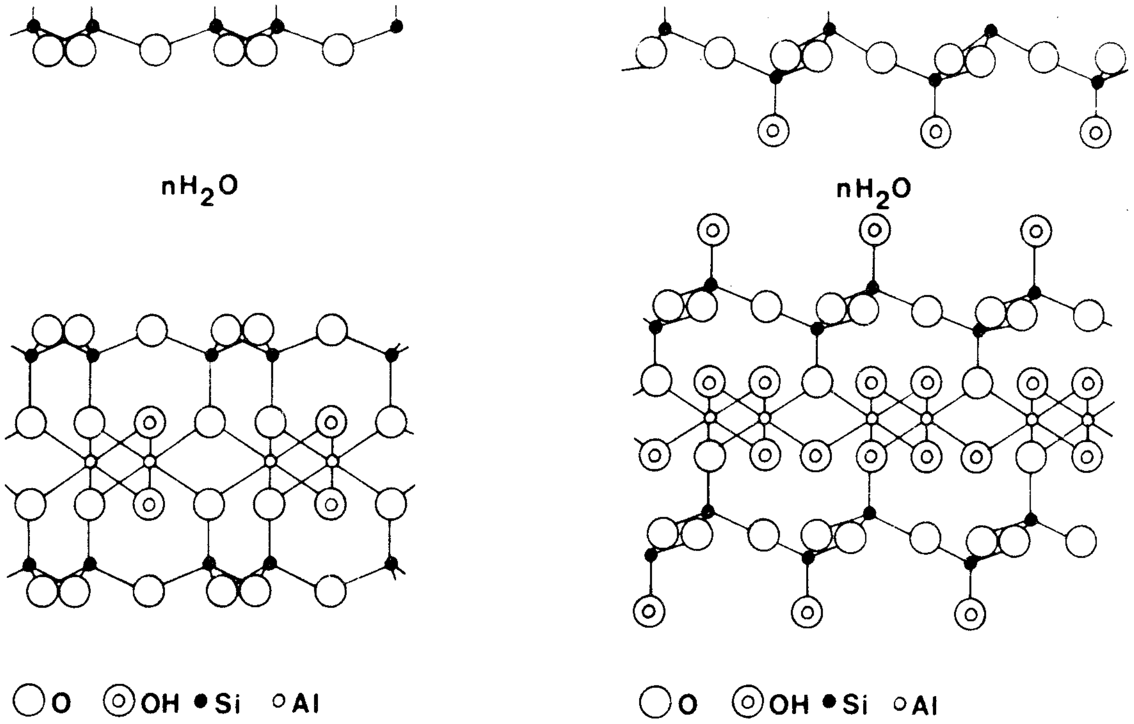


Fig.8. Possible montmorillonite structures. Left: Traditional version (HOFMANN, ENDELL & WILM). Right: Alternative version (EDELMAN & FAVEJEE). $n(H_2O)$ represents intercrystalline water.

Expansion and shrinkage of the smectite lattice are affected by the nature of adsorbed cations or organic molecules, and it is usually assumed that the amount of adsorbed water depends on the exchangeable species. NMR and dielectric measurements (7), as well as systematic determination of the swelling pressure of smectite-rich clays (8), indicate, however, that the adsorption of intercrystalline water leads to a structural organization of the molecules that does not primarily depend on the presence of exchangeable cations. Such ions only seem to give rise to well-defined structural modifications of the adsorbed water lattice.

A number of observations, such as the obvious pH-buffering ability of smectite, support the assumption that montmorillonite is best described by the EDELMAN/FAVEJEE model for temperatures below 100 °C. It deviates from the other, earlier model with respect to the coordination of tetrahedra, implying that a certain fraction of the tetrahedra of the silica sheets is converted.

Thermogravimetric measurements show that intercrystalline water is completely removed at about 100°C, eventually including some surface hydroxyls, while differential thermal analyses indicate additional lattice reorganization of surface hydroxyls extending into the temperature interval 150 - 200°C (7). Not until the temperature exceeds 300 - 400°C,partial dehydroxylation of the octahedral sheet takes place.

Water uptake by smectites is crucial to the understanding of their swelling characteristics. In air of low humidity there is only a monomolecular water layer adsorbed on the crystal basal planes, but in moist air and with free access to water, the number of layers is successively increased with a concomitant separation of the lattice lamellae. If sufficient space is provided, the water uptake proceeds until 4 or possibly 5 ordered water molecule layers are fully developed. The adsorbed cations do then determine to what extent further swelling can take place. Thus, with Na as adsorbed ion, additional water is adsorbed but it is less ordered and yields separation of the lamellae, which are rearranged to form edge-to-edge or edge-to-face flocs if the salinity is sufficient.

For montmorillonite with Ca as adsorbed cation, the expansion stops when the 4 to 5 water layers have been established and the clay appears in the form of fairly stable stacks ("domains") of face-to-face oriented lamellae. This amount of adsorbed interlayer water corresponds to a bulk water content of about 50 - 70 %, and below this interval the strong mineral/water interaction sets up "suction" gradients that leads to a uniform interlayer distance and therefore to a very homogeneous microstructure, especially for Na montmorillonite. This is illustrated by Fig.9, which shows the characteristic pattern of densely grouped, interwoven laminae of montmorillonite-rich clay.

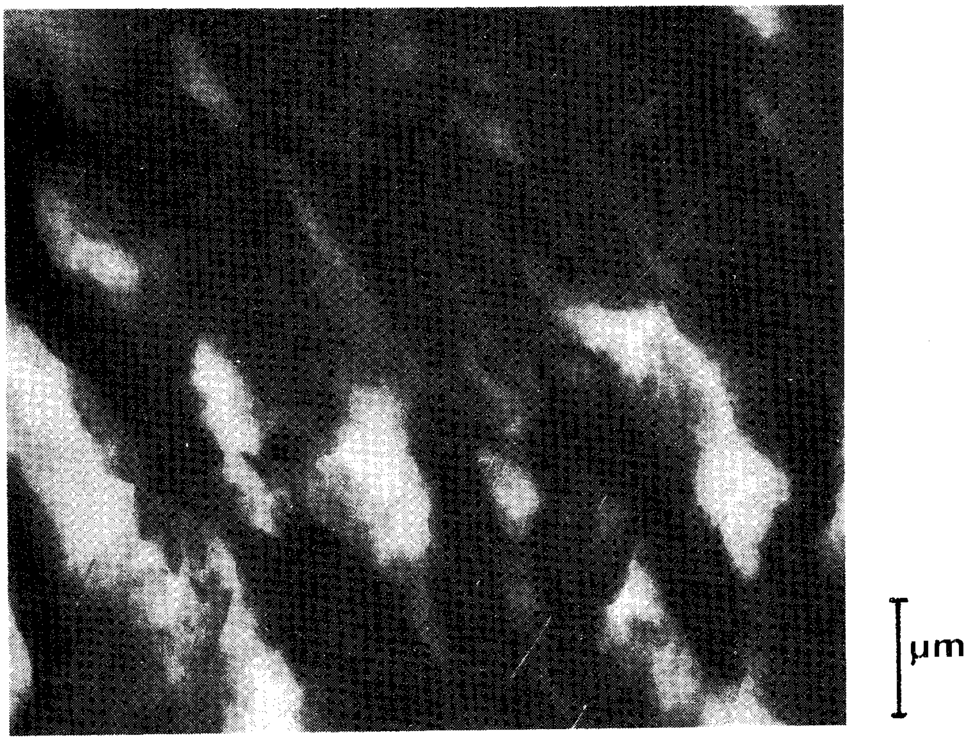


Fig.9. Electron micrograph of ultrathin section through a montmorillonitic, dense clay of Triassic age.

The extremely narrow passages for water flow and the structured nature of much of the adsorbed interlayer water, cause a very low hydraulic conductivity at high bulk densities. The large exposed mineral surface area at the dispersion of Na montmorillonite that results from swelling beyond the 50 - 70 % water content, yields a low conductivity also of diluted gels, which is of great practical significance in the present context. Completely dispersed smectite clays have a specific surface area of 600 - 800 m^2/g dry clay.

If a smectite mineral mass is contacted with water but kept confined so that expansion is prevented or limited, it will exert a swelling pressure which may be very high at high bulk densities, but which is noticeable also at very low densities. At higher densities than about 1.9 t/m^3 the pore water composition is hardly a determinant of its magnitude, which suggests that the "disjoining" pressure exerted by the interlayer water is primarily due to its structured nature, i.e. to the tendency of surface-near water to "crystallize". This would imply that very dense Na and Ca montmorillonites yield approximately the same swelling pressure, which also turns out to be the case.

The cation exchange capacity, which is partly caused by replacement of the protons of protruding hydroxyls in the EDELMAN/FAVEJEE model, and entirely caused by replacement of interlayer cations in the HOFMANN/ENDELL/WILM version, amounts to about 80 - 130 mE/100 g dry clay. Lattice vacations and substitutions are assumed to produce the net negative charge that is balanced by adsorbed cations in the latter model.

The (compact) density of smectites with no adsorbed water has been found to range from about 2.2 to more than 2.7 t/m^3. The experimental difficulties in the determination of the density are considerable and it is obvious from the discussion of the structural models that the definition of crystal lattice density is not quite clear in the case of montmorillonite. Thus, after drying to 105°C some surface hydroxyls, which should actually be considered as structural members of the EDELMAN/FAVEJEE model, have gone and the remainder therefore has a higher density than at 80 - 90°C when practically no water is adsorbed but the OH:s are back in position. From a practical point of view, the difference between montmorillonite, for instance, and illite can be made more clear by comparing their densities in humid air. Thus, at 50 % humidity, the density of montmorillonite is of the order of 2.2 to 2.3 t/m^3, while it is about 2.5 t/m^3 of illite (9). This means that conversion from montmorillonite to illite involves contraction of the lattice and liberation of water.

It should be added that recent studies (6) indicate that prolonged heating to more than 100°C of water saturated montmorillonite seems to be critical to its crystal lattice stability. Thus, the previously mentioned charge change probably has the form of silica leaving the tetrahedrons to be replaced by aluminum. These silicons, possibly in hydrated forms, may precipitate at the domain edges, thereby "spotwelding" their individual lamellae together. By this, spontaneous swelling may be prevented (10).

The usefulness of smectites as barrier materials is obvious from what has been referred to in this text and in numerous reports and textbooks, especially considering the major consequences of their rather extreme hydrophilic nature: the very low hydraulic conductivity and the ability of self-sealing through internal redistribution of water and solid matter. However, smectites do also present problems which result from their affinity to water and which are manifested by a dramatic loss in strength and a substantially increased compressibility if they are allowed to swell in an uncontrolled fashion. Another difficulty may be the very high swelling pressure that will be exerted on supporting constructions, such as confining rock, if the density of the clay is too high.

3.2 Clays of potential use

3.2.1 General

The two candidate clay mineral groups which passed our examination do not occur in a completely pure form in nature; they are mixed with other minerals and contaminated with organics. Yet, there are large deposits which have fairly high contents of the respective mineral and a few of them will be referred to in this report, partly because they are reasonably well known, partly because they represent possible exploitable resources for Swedish purposes.

Basic physico/mechanical data are given for each of the materials and the chapter ends with a list of possible applications.

3.2.2 Illitic clay

Economical and practical aspects point to the need of using natural sedimentary clays of Quaternary age, since they are available in huge quantities in this country. Unfortunately, there is a spectrum not only of clay minerals but also of rock-forming minerals in these clays and it is hardly possible to find clay strata which hold more than 50 % illite. Some of the other mineral constituents have properties similar to those of illite, i.e. chlorite and degraded mica, so that clay-rich materials actually offer a mineral substance which behaves as illite. This requires a clay percentage of 60 - 70, which suggests that the most convenient strata are the Ancylus and Litorina sediments, which form the transient between older, varved glacial deposits and younger, usually organic-rich post-glacial sediments. The late-glacial clays are of wide occurrence in the present coastal areas, large river valleys and lake districts in glaciated regions like Sweden, but they usually do not form thick beds - the average being 0.5 - 2 m. Normally, they are located less than 5 m from the present ground surface.

The late-glacial clays, to which we will confine ourselves in this chapter, have a color ranging from grey to bluish, and they can be identified by the richness in clay-sized particles, and by the correspondingly high liquid limit, while the organic content is low to moderate. Fig.10 serves to illustrate their stratigraphical position, while Table 1 summarizes some major data from two classical geotechnical test sites, which both have about 2 m thick clay-rich horizons of interest. The clay fraction of the presently discussed fine-grained clays consists of illite, quartz, feldspars, calcite, kaolinite, chlorite, vermiculite, and mixed-layer minerals (11). On average, illite forms 30 to 50 % of the total mineral mass.

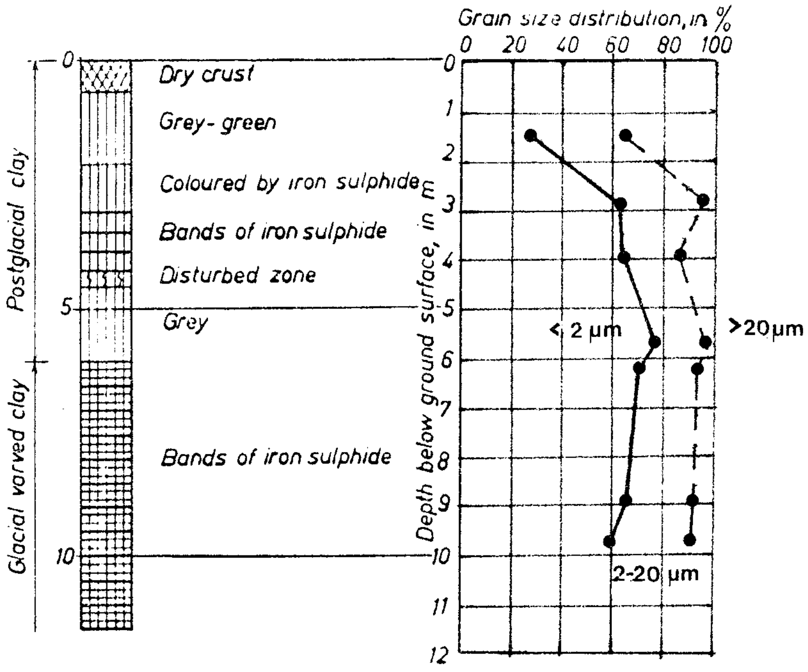


Fig.10. Stratigraphical profile at Skå-Edeby near Stockholm (after Hansbo). Here, the Ancylus/Litorina stratum is located at 5-6 m depth.

The considerable structural variations and the very wet condition of the clay make it impossible to produce highly compacted blocks, or to apply and compact it to obtain reasonable homogeneity and sufficient density. For this purpose the excavated material must

Table 1. Geotechnical soil data for characteristic Swedish clay profiles.

Site	Depth	Bulk density	Shear strength	$w^{1)}$	W_L	Clay fraction ($\leq$ 2 µm)	Ignition loss %	Description
	m	t/m^3	kPa	%	%	%		
Skå-Edeby (fresh-,or brackish-water clays)	2.0	1.43	15	106	120	60	7.5	Green-grey muddy clay
	5.0	1.49	9	105	98	77	4.6	Grey clay
	7.0	1.59	11	73	58	52	5.5	Brown-grey varved clay
	8.0	1.61	10	70	56	63	5.6	Brown-grey varved clay
	9.0	1.62	12	67	55	56	5.1	Brown-grey varved clay
	10.0	1.61	11	65	52	54	5.1	Brown-grey varved clay
Lilla Edet (marine clays)	3.0	1.48	16	97	68	65	3.5	Grey clay
	3.0	1.48	12	97	73	70	3.4	Grey clay
	6.0	1.50	16	91	58	74	3.0	Grey clay
	19.0	1.64	24	58	54	55	4.0	Dark-grey clay

1) Natural water content

be dried and ground - possibly also washed to remove coarser grains - before it can be applied for field compaction or for the production of compacted blocks in the laboratory.

The usefulness of ordinary clay of the presently discussed type was checked through a pilot study which involved laboratory investigations of samples taken arbitrarily from Ancylus/Litorina strata at Ekerö near Stockholm. The clay content was found to range between 55 and 70 %, while the liquid limit averaged 55 %. Keeping in mind the criteria for the choice of clay barriers, the following properties should be determined and evaluated: 1) the behavior in the preparation phase, 2) the stress/strain relationships, 3) the sensitivity to erosion, 4) the hydraulic conductivity, 5) the diffusivity, 6) the thermal properties, 7) the swelling behavior, and 8) the chemical constitution including the mineral composition and the organic content. The pilot study did not comprise all these subjects, but experience offers some insight in the missing ones, and all of them will be commented on.

Preparation

The preparation of the natural, wet clay which had a water content of 59 - 65 % and the granulometric characteristics shown in Fig.11, was made so as to simulate possible full scale processing. For this purpose, lumps of clay were frozen at -5°C for three weeks and were then thawed and dried at 150°C for one day. The dry lumps, which had a water content of 2-3 %, were slightly ground to yield silt-sized aggregates, which were compacted in the swelling oedometer to various densities (8). Water saturation was achieved by contacting one end of the 2 cm thick samples with the KBS synthetic "Allard" water (12).

It should be mentioned here that compaction of this clay powder under a pressure of 50 MPa gave physically stable and very strong cylindrical blocks. Their density was of the order of 2 t/m^3.

The impression is that clay powder of this sort, i.e. not very finely ground, is suitable for layer-wise application and field compaction and that a dry density of 1.5 - 1.6 t/m^3 can be achieved although this may require some slight addition of water (cf. Fig. 12). This would yield a bulk density of 1.9 to 2.0 t/m^3 after water saturation.

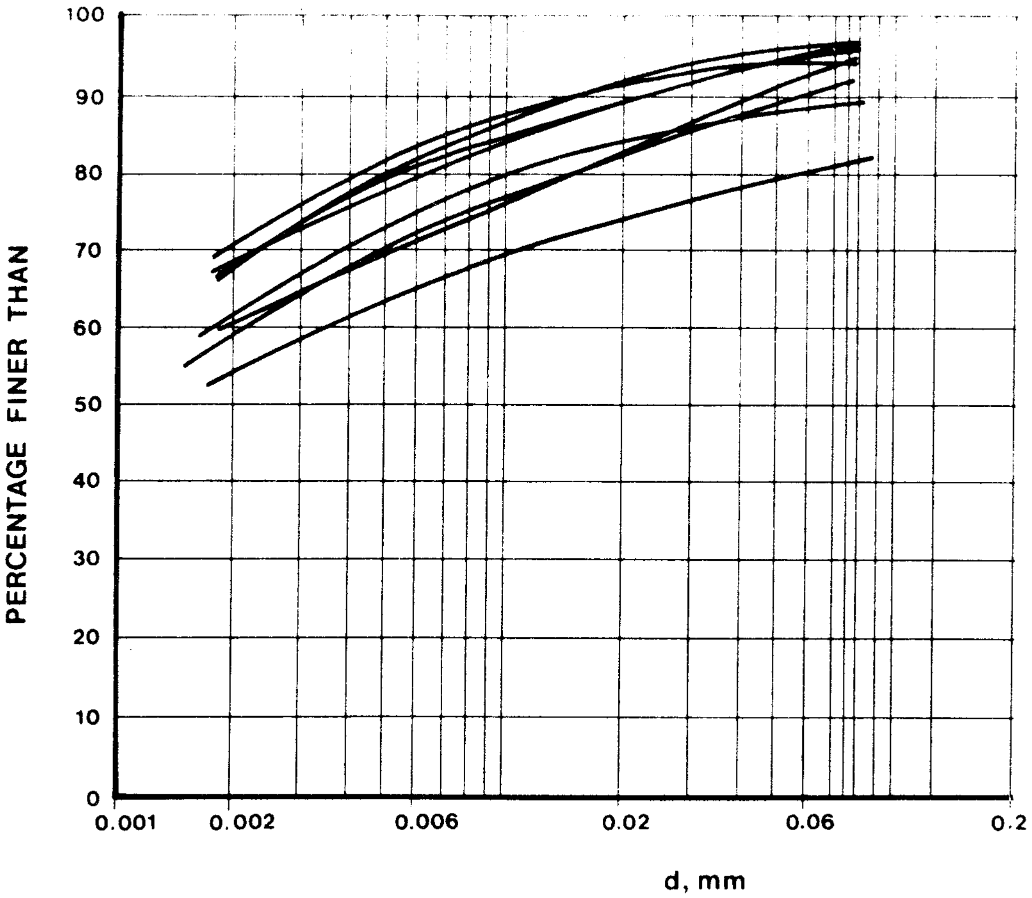


Fig.11. Grain size distributions for 8 Ekerö samples.

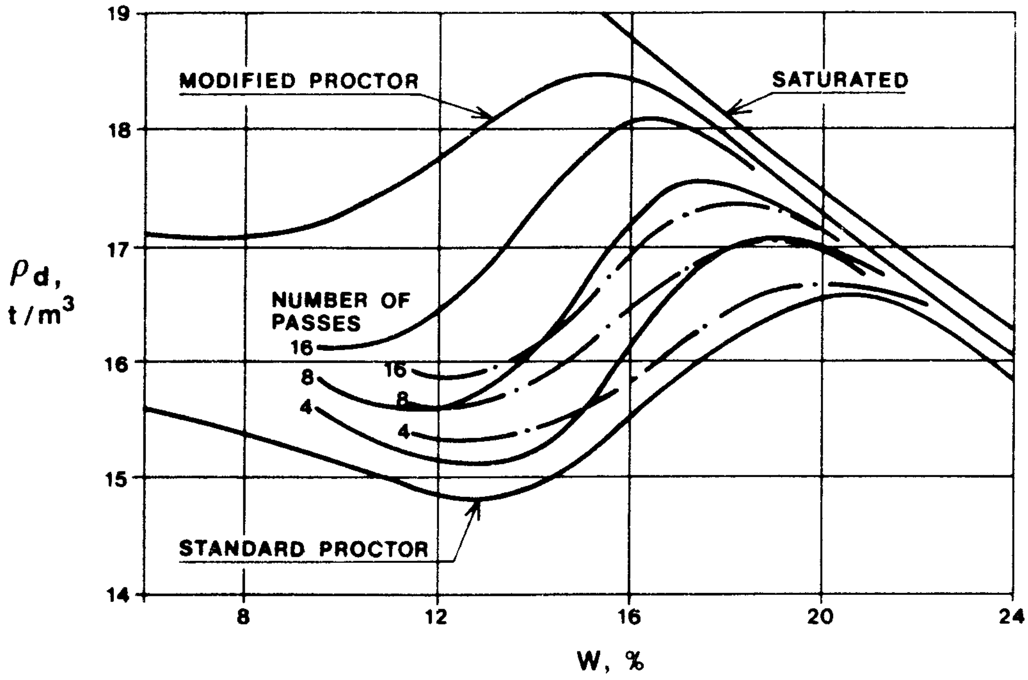


Fig.12. Field compaction curves of clay (13). The curves do not refer to the Ekerö material.

——— 10 ton vibratory sheepsfoot roller
—·— 35 ton pneumatic-tired roller

Stress/strain relationships

One postulate to make illitic clay qualify as near-field shielding of canisters in deposition holes, as well as of sealing plugs in boreholes, is to reduce its hydraulic conductivity and diffusivity as much as possible, which calls for a very high bulk density. It can be achieved by compaction of air-dry clay powder under very high pressures to form blocks. We will see later that for smectite, the density must not be too high in order to avoid unacceptable swelling pressures; for illite this will not be a problem.

A bulk density of 2.2 t/m^3 after water saturation in situ would be a practical maximum and this yields strength and deformation properties that are expected to be similar to those of moraine clay. The required softness calls for a slightly lower bulk density, the optimum probably being in the range of 1,95 - 2.05 t/m^3, or possibly somewhat less than that. This is concluded from creep and strength tests of smectite-rich clays as well as from theoretical considerations concerning the physical nature of their interparticle bonds (14).

Experience from various tests of smectites, which are characterized by lower deformation moduli and more expressed creep behavior than illitic clays, suggest that even if the bulk density is lowered to 1.9 t/m^3, the bearing capacity will be quite sufficient to carry heavy canisters of the KBS type with practically negligible settlement.

For use as layer-wise applied and compacted backfill, maximum bulk density that can be obtained through field compaction is required, and it is assumed here that it will be possible to reach at least 1.9 t/m^3 after subsequent water saturation if effective padfoot rollers are used for the compaction.

Sensitivity to erosion

Clays are generally considered to be more resistant to erosion than silt and fine-grained sand but this is not generally true. The whole matter of soil erosion is actually being reconsidered in connection with the development of improved soil dam design principles, and much improved laboratory test procedures and classification schemes have been developed in recent years. Thus, the use of the pinhole test and other means of determining, in a quantitative manner, the erodibility of clays have become widely used (cf. 15). The particular sensitivity of expansive clays is verified through such tests, the ranking list being based on the so-called SAR and ESP numbers, which refer to the physico/chemical properties of the clay and to the groundwater composition. Illites turn out to be much less sensitive to erosion than smectite.

In the standard pinhole test, which involves flow of water through a prebored fine hole that crosses the soil sample and analysis of the percolate with respect to its content of eroded matter, the gradient is slightly higher than unity, while it will be very much lower in a sealed repository. In the course of the application of the backfill and until the final sealing has taken place, the gradients will be higher but the inflow so small and slow that erosion will be negligible. This is also the case of highly compacted clay in deposition holes and boreholes, which will be exposed to slow groundwater flow through joints traversing the holes. Here, illitic clay may appear to be more easily "slaked" than smectites since the cohesive interparticle forces are higher in the latter clays. On the other hand, smectite expands and migrates far into the rock if the joints are wide.

Hydraulic conductivity

Fine-grained, natural illitic clay is known to have a low hydraulic conductivity aven at low bulk densities and deviations from Darcy's law are frequently reported.

The Ekerö clay was tested by injecting "Allard" water into saturated samples in the swelling pressure oedometer through its lower filter (Fig.13). The pressure was kept constant at a about 50 kPa, which gave a gradient of approximately 300. The flow was recorded at the inflow as well as at the outflow by determining the rate of displacement of a water meniscus in calibrated capillaries. Each test was run for about 1 week, reasonably stationary flow being recorded already after a few hours. The evaluated hydraulic conductivity is plotted in Fig.14.

The conductivity of the Ekerö clay that corresponded to the bulk density 2.0 t/m^3, was found to be an order of magnitude higher than that of an equally dense very illite-rich clay sample from France, which was put to the author's disposal by Dr Max Müller-Vonmoos, ETH, Zürich. This material, which was prepared and tested in the oedometer like the Ekerö samples, has a clay content of 85 %, a specific surface area of 114 m^2/g dry clay, a liquid limit of 70 %, and a cation exchange capacity of 28 mE/100 g dry clay. It is assumed to be a montmorillonite clay that was transformed to illite by potassium uptake and fixation. The lower conductivity of the French illite than of the Ekerö clay can be explained by its higher tortuosity, which results from the higher content of small particles and by the large specific surface area that is also associated with the fineness.

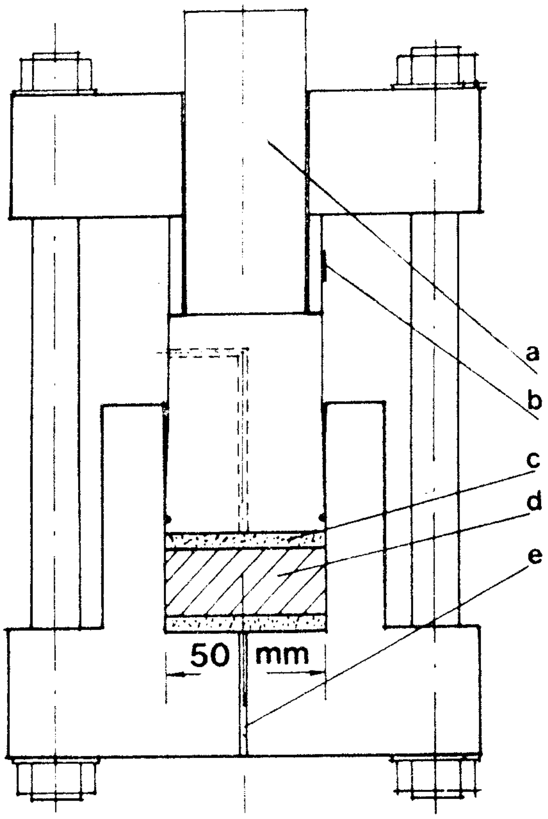


Fig.13. The LuH swelling pressure oedometer.

a) Free piston for loading the sample; when the ring (b) is stress-free the applied load balances the swelling pressure.

c) Filter

d) Sample

e) Lower water inlet

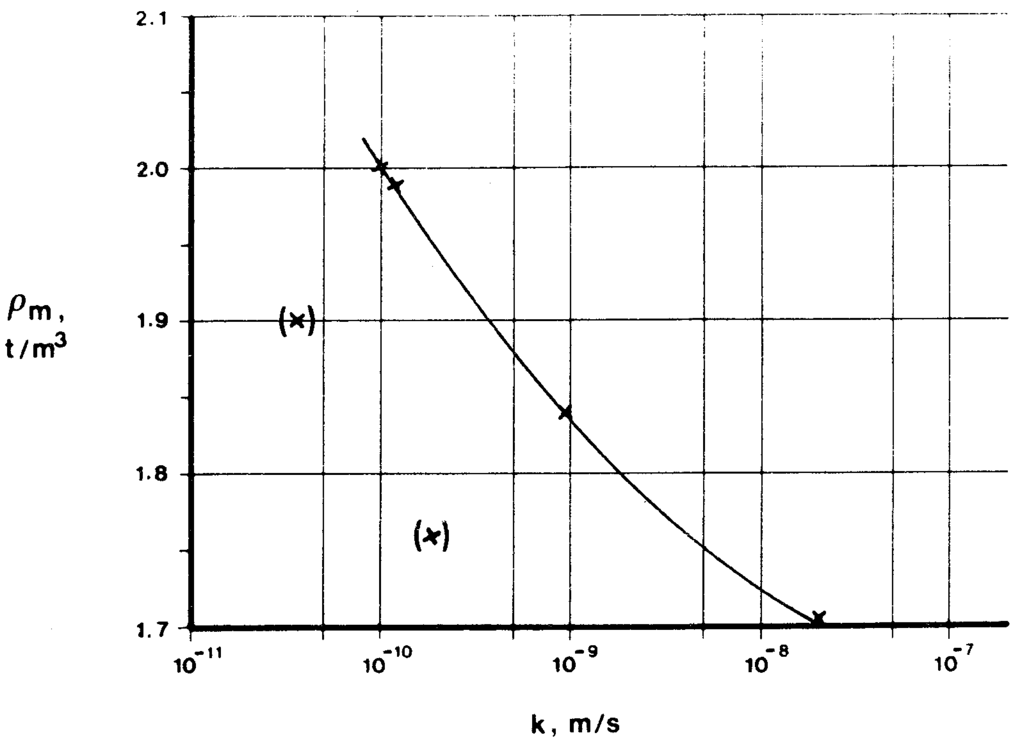


Fig.14. Hydraulic conductivity (k) of the Ekerö clay versus bulk density (ρ_m).

Diffusivity

The diffusion coefficient for individual ion migration is not the same since the size, degree of hydration, and charge are determinants of the diffusion rates. Furthermore, cations, and to a smaller extent also anions, interact with the crystallites through adsorption, for which a correction term K must be introduced in order to evaluate a true diffusion coefficient from experiments.

No diffusion tests have been run of the Ekerö samples, but reasonably representative values of this kind of clay can be derived from various field and laboratory tests of similar clays. Thus, for Ca $6 \cdot 10^{-12}$ m^2/s seems to be a proper diffusion coefficient, while $6 \cdot 10^{-10}$ m^2/s has been evaluated for K in Norwegian field tests (16). For Cl, the latter diffusion coefficient actually fits concentration profiles that have been recorded in marine illitic clay slopes percolated by fresh water for some 11000 years. All these data refer to soft clays.

Thermal properties

The heat conductivity of illitic clays is fairly well known from various investigations, particularly in Norway and Canada, and comprehensive data are available in literature (17). We will confine ourselves here to show its dependence on the water content of water saturated clay (Fig.15). The strong influence of quartz is illustrated by the boundaries q= 0 for no quartz contained in the clay, and q= 30 for 30 % quartz particles expressed in terms of weight percent of the total mineral content.

The heat capacitivity of clays is largely determined by the water content. For very dense, water saturated illitic clay with a water content of 20 % this thermal parameter can be taken as $2.9 \cdot 10^6$ $J/m^3,K$, while a water content of 100 % would yield about $3.7 \cdot 10^6$ $J/m^3,K$.

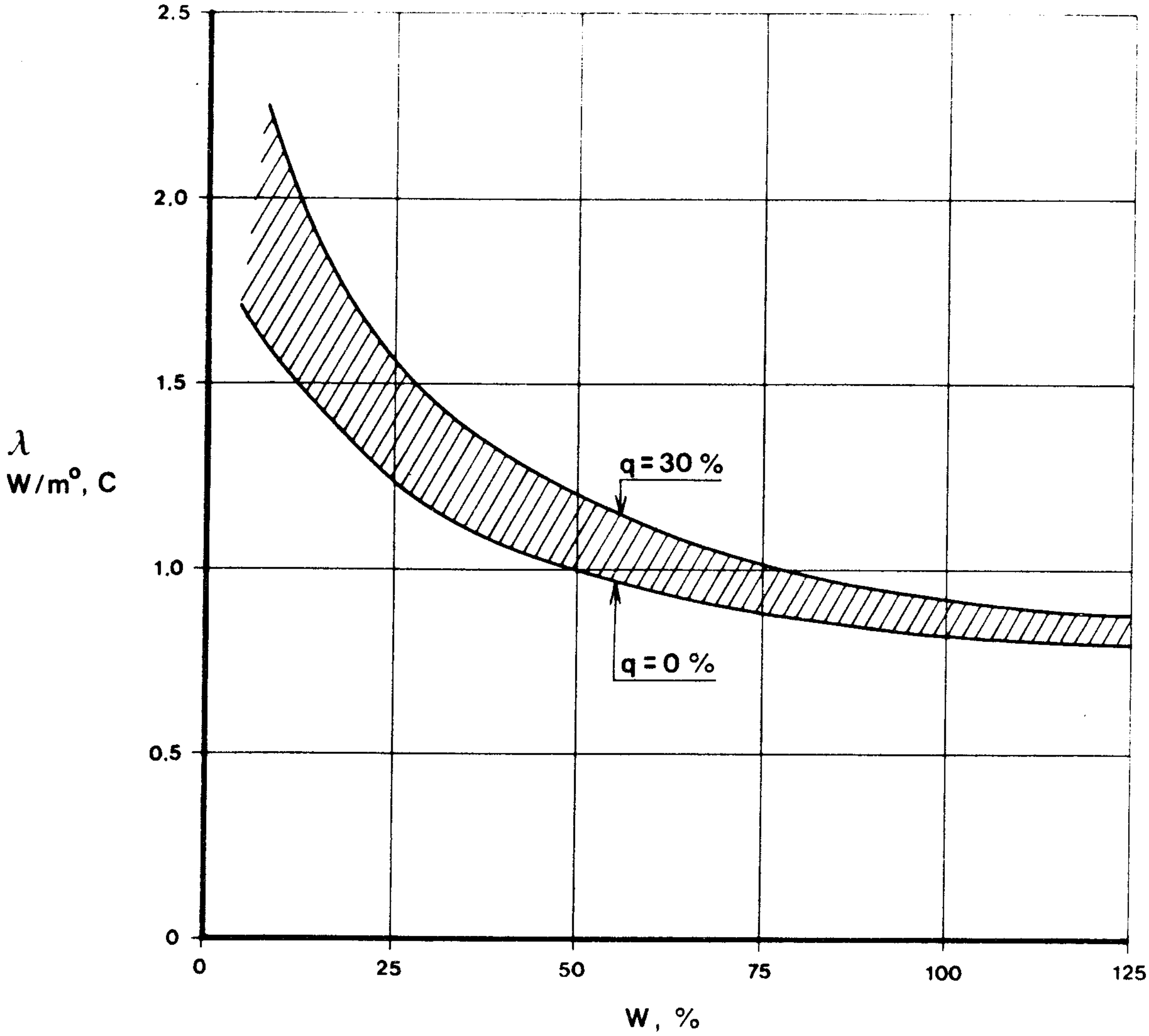


Fig.15. Heat conductivity of water saturated illitic clay. q represents the content of quartz particles in weight percent.
(Sven Knutsson, Div.of Soil Mech., Univ. of Luleå).

Swelling behavior

Illitic clay shows only slight expansion when it is free to swell from a moderately dense state, and this is partly due to elastic rebound and partly to rehydration of the crystallites.

Ekerö clay samples with 15 mm thickness and 50 mm diameter were wetted in oedometers on which the load was successively adjusted so as to maintain a constant volume during the water uptake of the air-dry clay powder. The recorded rate of wetting is illustrated by tests of samples with 50 mm thickness and 50 mm diameter (Fig.16). We see that saturation was almost completed in less than one day, which is considerably faster than the corresponding process in dense smectite. This points to somewhat different physical mechanisms, the wetting of illite probably being more related to capillary effects.

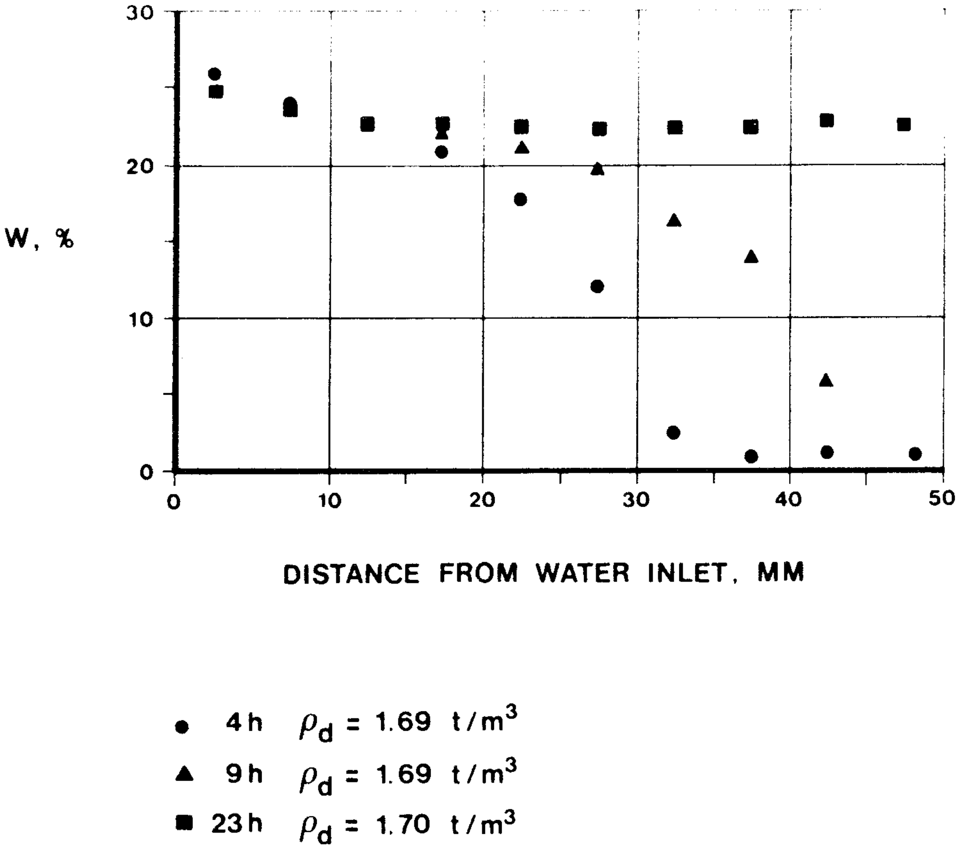


Fig.16. Distribution of water in terms of water content at different times after the onset of water uptake.

The swelling pressures recorded about one week after the test start are plotted in Fig.17 versus bulk density.

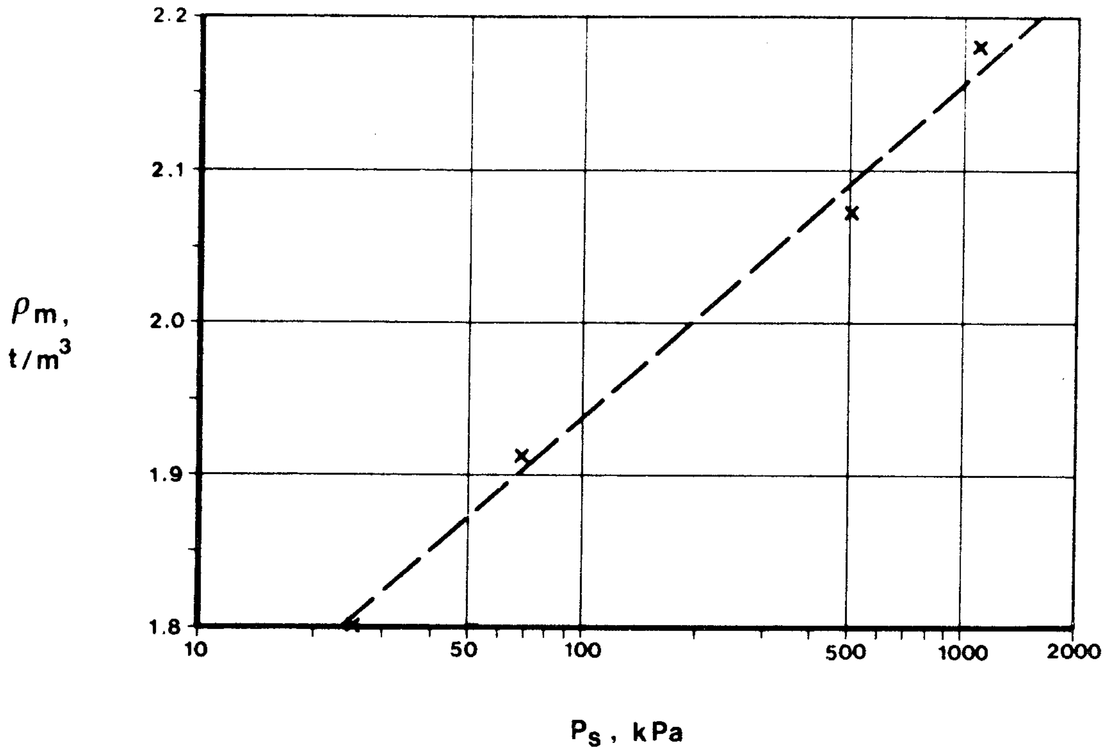


Fig.17. The swelling pressure of Ekerö clay as a function of its bulk density.

The French illite referred to earlier in the text was tested also with respect to the swelling pressure, and it turned out to be high: 0.7 MPa at 1.85 t/m^3, 5.5 MPa at 1.92 t/m^3, and 19 MPa at 2.08 t/m^3. Probably, the high values are largely due to stored elastic strain built in at the compression of the clay powder. Partly, however, hydration through direct water molecule coupling to the crystal lattice also took place and this may have contributed fairly much considering the large specific surface area of this clay. Accordingly, the swelling ability on stress release was very obvious and much stronger than that of ordinary Swedish illitic clays. Thus, a water saturated sample of the French illite took up additional water and swelled spontaneously by unloading from a bulk density of 2.07 t/m^3 to 1.90 t/m^3.

Chemical composition

The mineral composition has a bearing on the chemical stability of the clay, while its pore water chemistry, sulphur, and organic contents affect the corrosion rate of the copper canisters.

The mineral composition, which is actually similar to that of smectite-poor weathered granitic rock, is illustrated by the X-ray diffraction diagram in Fig.18. It is representative of many fine-grained late-glacial illitic clays of the presently discussed type. Illite, quartz and feldspars are recognized as main minerals, accessory minerals being carbonate minerals (mainly calcite), kaolinite, chlorite and some heavy minerals. Usually, the cation exchange capacity ranges from 30 to 40 mE/100 g dry clay.

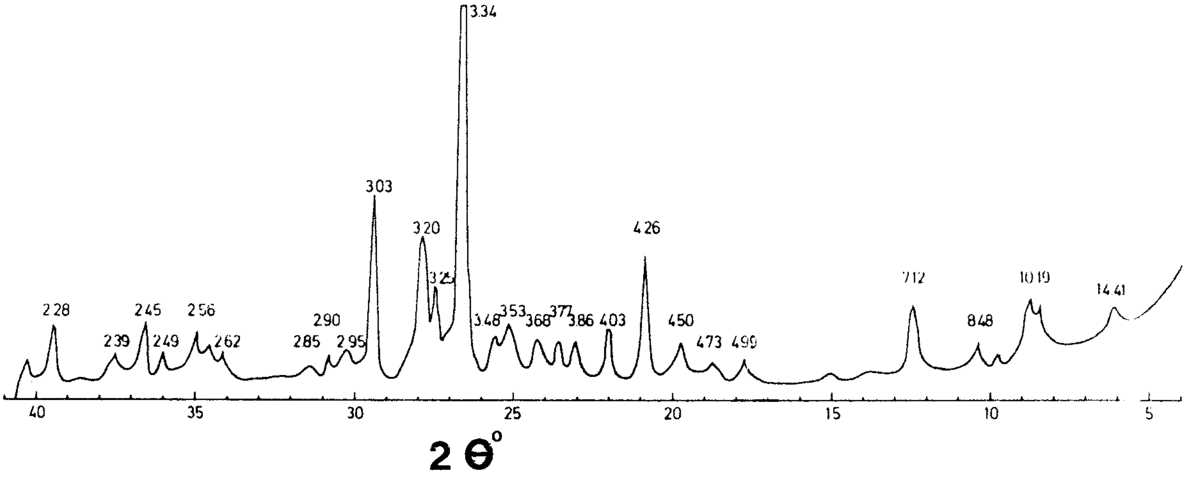


Fig.18. Representative X-ray diffractogram of late-glacial clay.

Sulphides, which are usually present in small amounts, result from a reduction process for which a number of ingredients are required, such as sources of iron and sulphate, a suitable redox potential, sulphate-reducing bacteria, and organic material. The resulting iron sulphide ranges from amorphous to well crystallized mineral matter, such as pyrite. A large fraction of the ignition loss on heating to 600 - 800°C that is not due to hydroxyl escape, may actually be caused by amorphous iron sulphides. This means that many of the late-glacial clays have sulphide contents in excess of 1000 ppm.

The organic content is usually relatively high also in "organic-poor" clays, and values lower than 5000 - 10000 ppm are rarely found. It originates from microorganisms which added various products to the soil from the autolysis of their body substance. Due to the lack of suitable identification techniques these substances, as well as not yet fully decomposed matter, commonly masquerade as humus, which therefore appears to range from fairly low molecular weight compounds to high molecular ones. The most important representatives are humic acids and fulvic acids. They contain several functional groups that interact with clay crystal lattices, which suggests that organic molecules are partly sorbed on clay minerals and not free to migrate, unless drag forces from percolating water liberate them (18). They are probably relatively well protected from microorganisms in pores smaller than 1 micrometer.

3.2.3 Smectitic clays

We will confine ourselves here to discuss bentonite, which is the geological term for smectite-rich clay formed by devitrification of the natural glass component of volcanish ash, deposited in the sea or estuaries in prehistoric time. As a consequence of the numerous volcanic activities in Paleozoic and later time, bentonite is a very common constituent of thick sediment sequences all over the world. The chemical composition is a function of the nature of the parent magma and of the water in which the deposition and transformation took place. The smectite content and homogeneity of the sediments are determined by these factors but also by contaminations in the water, such as river-transported minerals that were brought to the sedimentation area by currents.

In most bentonite beds, the clay-sized particles constitute 50 - 90 % of the total mineral mass, and smectite usually forms 50 - 90 % of the clay fraction. Very high clay concentrations (85 - 90 %) are offered by Wyoming and South Dakota (Mowry) bentonites, and these beds usually have a smectite content of 70 - 80 % of the clay fraction. They are, thus, extraordinarily rich in smectite.

Thin bentonite seams occur frequently in many sediments but they are often too thin and too sparse to be exploited. Several decimeter and even a few meter thick beds are fairly frequent in the Mediterranean area and in continental Europe, North Africa, India and North America. The dominant cation in most bentonites is Ca/Mg, and only a few formations like the Wyoming deposits hold mainly Na in their natural state.

The very wide use of bentonite in various industries, such as foundry, deep drilling for oil exploitation, and various chemical plants, has led to extensive mining in Europe and north America. Since the Na form offers the best physical properties for many applications, effective methods have been developed for cationic exchange from the Ca state on an industrial scale in many countries.

Industrial use of bentonite requires that it is conditioned to form dry powder and such clay is therefore commercially available in bulk for shipping, or bagged for delivery in smaller quantities. Protection from rain and snow is a necessary prerequisite in the use of bentonite.

This chapter is mainly concerned with the same, practically important properties that were specified for illitic clay. Various bentonites that are of potential use in Swedish repositories will be referred to in the text.

Preparation

Industrial processing of mined bentonites which require ion exchange to be converted into the Na state is schematically illustrated in Fig.19.

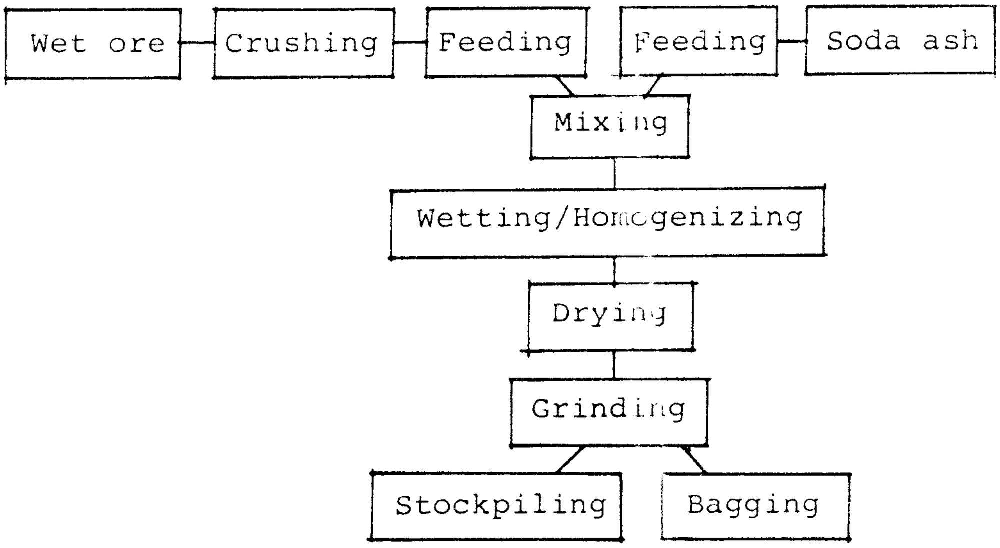


Fig.19. General flow pattern for soda activation of Ca bentonite.

The amount of soda added to the clay is usually 2 - 5 weight percent of dried clay. This treatment yields a slight amount of calcium and calcium/magnesium carbonates in the clay by which the original content of such compounds may be at least doubled. Grinding and drying brings the bentonite into the desired powder form, the water content of the material being a function of the relative humidity of the air. Na bentonite powder usually has a water content that ranges between 7 and 14 %. The liquid limit varies between 70 and 100 % for Ca bentonites and between 200 and 450 % for Na bentonites that are commercially available. This consistency limit has a wide use in soil classification and is a practical and valuable parameter for characterization of bentonites.

It should be added that industrial processing of bentonite may also involve extrusion and pelletization, which yields coarse aggregates of uniform size. This form may be advantageous for the backfilling of narrow slots etc. in which traditional compaction operations cannot take place.

Stress/strain relationships

A high density of water saturated bentonite yields a very low hydraulic conductivity and is therefore required for near-field shielding of canisters and for sealing plugs in boreholes, shafts and tunnels. It can be achieved by compaction of air-dry clay powder under very high pressures to form blocks as shown in several experimental investigations in the course of the KBS project. However, high bulk densities also cause high swelling pressures and since a practical maximum of the latter is about 10 MPa for the KBS concepts, the bulk density of water saturated bentonite should not exceed 2.0 - 2.1 t/m^3 provided that the bentonite is rich in Na smectite. Densities of this order of magnitude give the matured bentonite mechanical properties that are similar to those of stiff moraine clay, which means that its bearing capacity is much more than sufficient for carrying heavy KBS canisters and to prevent them from settling more than a fraction of one centimeter in one million years (14). This is concluded from settlement predictions that are based on creep theory and experimental identification of relevant creep parameters. Preliminary investigations indicate that the undrained shear strength of water saturated Wyoming Na bentonite (MX-80) with a bulk density of 2.0 t/m^3 is in the interval of 0.4 to 4 MPa depending on the rate of strain, while its modulus of elasticity seems to range between 100 and 1000 MPa. These parameters are of profound importance for a proper evaluation of the canister stress state in the event of rock shear of the kind shown in Fig.1, and so is the stress relaxation behavior which is presently being investigated.

Current analyses indicate that a slightly reduced bulk density, i.e. in the interval 1.9 - 2.0 t/m^3, may represent optimum conditions with respect to possibly induced canister stresses and to the bearing capacity of the clay.

For certain purposes the rheological properties of bentonite can be expressed as a viscosity, the order of magnitude of which is illustrated in Table 2.

Table 2. Approximate viscosity of Na bentonite (MX-80)

Density t/m	Viscosity MPas	Source
1.9	$10^6 - 10^8$	Creep tests
1.5	$10 - 10^4$	Literature data
1.2	$2 \cdot 10^{-2}$	Viscometer tests
1.1	10^{-4}	" "

The much more modest requirements of backfills with respect to the hydraulic conductivity than of "overpacks", means that bentonite can be used as a sealing component of mixtures with coarser soil.

Experience indicates that the required property of the backfill to exert a certain, slight swelling pressure on the confining rock is fulfilled even at a content of Na bentonite of only 20 %, provided that the bulk density in the water saturated state is at least 1.8 t/m^3. Such soft mixtures can be accepted for a small fraction of backfilled tunnel sections, such as the uppermost parts where shotcreting has to be applied. The major part of such sections should be backfilled with denser and much less compressible material in order to minimize the displacement of the interface between near-field, expansive "overpacks" and the overlying backfill. This is easily achieved by field compaction of silt/sand/gravel- type ballast with about 10 % Na bentonite mixed with it. The bulk density of such a mixture will be at least 2.1 t/m^3 after water saturation and its stress/strain properties similar to those of very stiff moraine clay.

It is estimated that different commercial bentonites affect the stress/strain/time properties mainly through the varying smectite content, and that this influence is minor for highly compacted bentonite. For mixed backfills larger differences are expected. The matter is looked into in the subsequent text.

Sensitivity to erosion

Smectites are the most easily eroded clays, except for the case of extremely slow water flow through diluted clay gels. The exception is because the stability of smectite gels of low concentration is better than that of illitic gels if the salinity is not too low. For high flow rates smectites show rapid "swelling/slaking" except when the clay is calcium saturated. The choice of Ca bentonite may thus be advantageous in composing backfills. However, as shown previously, the capacity of the water flow from rock joints is expected to be very moderate in the type of rock that is being considered for repositories, so that erosion will actually not be a problem even when Na bentonite is used as a backfill component. This is confirmed by the current "Buffer Mass Test" (BMT) of the current Stripa Project (19).

Hydraulic conductivity

The very low hydraulic conductivity of bentonite has been verified through various laboratory investigations in Sweden and elsewhere. Thus, for the very low hydraulic gradients that will prevail some time after the final sealing of a repository and for which Darcy's law is not valid, the earlier derived general relationship between the hydraulic conductivity and the bulk density of Na smectite is still applicable (Fig.20).

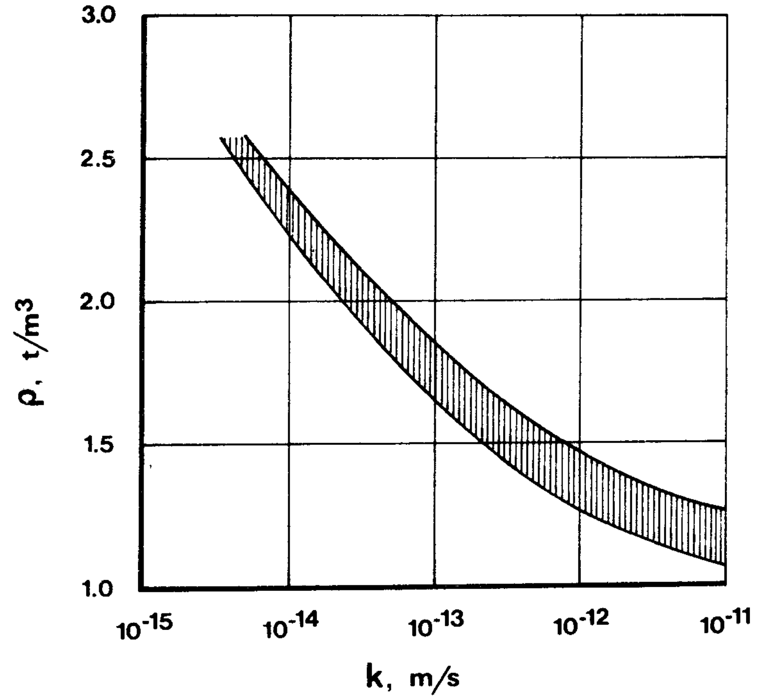


Fig.20. Hydraulic conductivity versus bulk density for Na smectite.

More explicitely, the hydraulic conductivity of MX-80 is given in Table 3. The tests have been run at different temperatures by using the swelling pressure oedometer (20).

Table 3. Average hydraulic conductivity k in m/s versus bulk density of MX-80 ("Allard" water)

Bulk density t/m^3	k, m/s Room temp.& low gradient	70 C,& very high grad.
2.1	$1.5\ 10^{-14}$	$1.5\ 10^{-13}$
2.0	$2.0\ 10^{-14}$	$2.0\ 10^{-13}$
1.9	$3.0\ 10^{-14}$	$5.0\ 10^{-13}$
1.8	$5.0\ 10^{-14}$	$8.0\ 10^{-13}$
1.7	$8.0\ 10^{-14}$	10^{-12}

An equally comprehensive study of commercial Ca bentonite (Erbslöh, West Germany) gave a hydraulic conductivity that was approximately 10 times as high as that of MX-80.

A matter of great practical importance is the influence on the hydraulic conductivity of the choice of commercial bentonite for sealing purposes. From a scientific point of view this would require a comprehensive determination of the various soil data of potential importance and this has not been worked out yet. A first approach can be made by considering only the importance of the liquid limit, since it is an integrated measure of the specific surface area and the surface activity, and thus of the content of clay minerals. Fig.21, which is a compilation of such data, clearly shows the profound importance of the bulk density but also the usefulness of the liquid limit as a rather sensitive measure of the physico/chemical properties that govern the hydraulic conductivity. We can conclude from this diagram that ordinary commercial bentonites with a liquid limit of more than 200 % will serve well as "overpack" with respect to the required hydraulic conductivity. Consequently, they should also be useful as sealing components in mixed backfills.

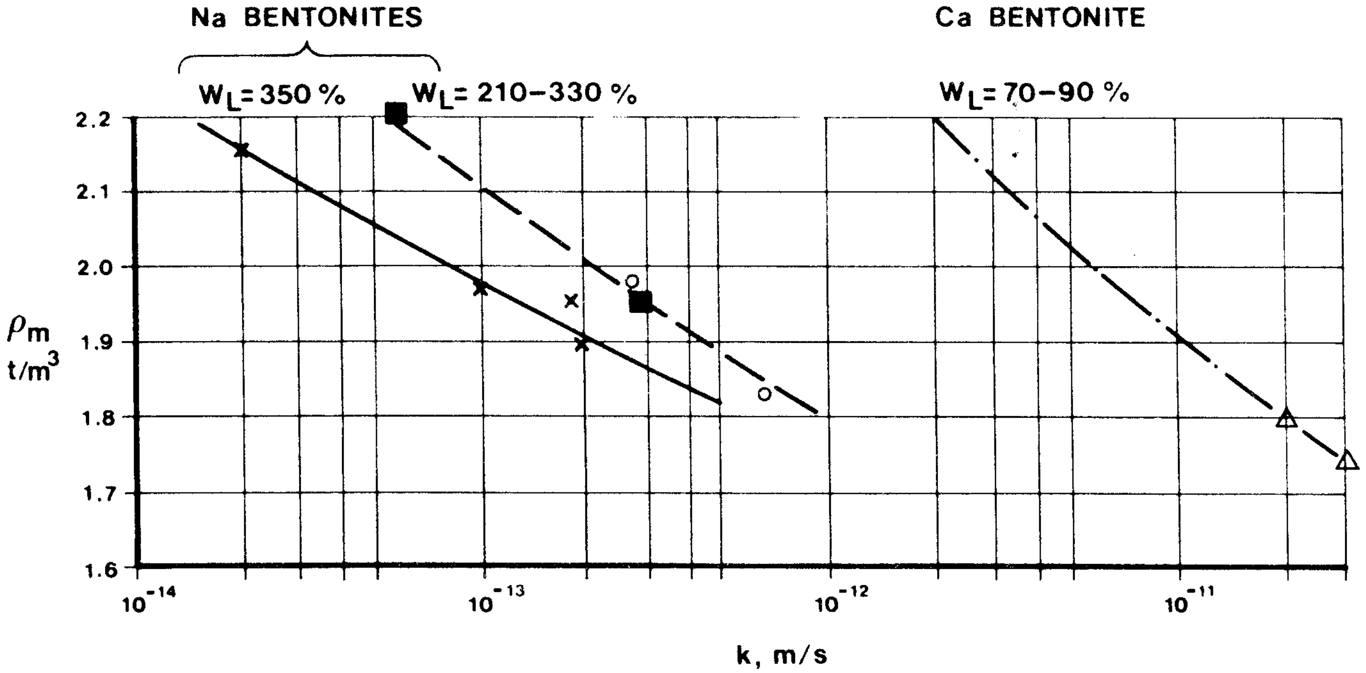


Fig.21. Bulk density versus hydraulic conductivity for commercial bentonites with different characteristic liquid limits.

Diffusivity

Ion diffusion through bentonites has been investigated and reported in a number of papers (cf.21). Various experimental set-ups have been tried to avoid artefacts. Thus, tests have been run by use of the swelling pressure oedometer with the saturated clay sample in contact with aqueous solutions of Cs, Sr, I and Cl, and the diffusivities evaluated from the tracer concentration/distance profiles in sectioned samples 10 days after the onset of diffusion. Other series have been run with pairs of saturated samples, one of them being tracer-doped and put in contact with the virgin sample at the test start.

Assuming the K_d coefficient to be a constant, the application of diffusion/sorption equations have yielded the "true" diffusion constants of Na and Ca bentonites with a bulk density of 2.1 t/m^3 that are given in Table 4.

Table 4. Average diffusion coefficients of Na bentonite (MX-80) and Ca bentonite (Erbslöh).

Ion	Diffusion coefficient, m^2/s
Sr^{2+}	$2.3 \cdot 10^{-11} - 4.8 \cdot 10^{-11}$
Cs^{+}	$3.4 \cdot 10^{-12} - 7.5 \cdot 10^{-12}$
I^{-}	$10^{-12} - 4.0 \cdot 10^{-12}$
Cl^{-}	$6.0 \cdot 10^{-12}$ (MX-80 only)

It has been concluded that the mechanisms for migration is different for cations and anions. Thus, cations migrate not only through the tortuous system of voids, which is the only passage for anions, but also through interlayer space and - depending on the size of the ion - through the crystal lattices. Also, it seems as if the ions move through the clay fully or at least partly stripped of their hydration shells, which is in agreement with the concept of largely immobilized interlayer water in very dense smectites.

A further conclusion from these tests was that Na and Ca bentonite with equal, high bulk densities offer approximately the same diffusion resistance to migrating ions, which indicates that surface phenomena as well as tortuosity are determinants of the migration processes. Reduction of the bulk density to about 1.6 t/m^3 and large differences in time for the samples to mature after the water saturation phase did not have any substantial effect on the diffusion rates.

Thermal properties

The thermal properties of bentonite do not deviate substantially from those of illitic clay. A number of experimental determinations as well as back-calculation using recorded BMT data have yielded the results specified in Table 5.

Table 5. Heat conductivity and capacitivity versus bulk density and water content of unsaturated and saturated MX-80, av. temp. 20°C.

Bulk density t/m^3	Water content %	Heat conduct. W/m,K	Heat capacitiv. MJ/m^3,K	Remark
2.2	11	1.01	2.50	50 % satur.
2.0-2.1	20-25	1.50	2.90-2.95	100 % satur.
2.1	14	1.15	2.56	
2.1	5	0.96	2.02	
2.0	5	0.83	1.92	
1.2	10	0.33	1.33	

The very moderate difference between these data and the ones given earlier for illite suggests that other bentonites will not behave very differently.

Swelling behavior

The swelling behavior of bentonite is probably the property of this clay type that has been most thoroughly investigated. Thus, much of the early work that concerns the swelling pressure has been repeated and extended to cover also lower bulk densities. Other bentonites than MX-80 and Erbslöh and also other smectitic clays have been tested. However, the conclusions are still the same, namely:

1. The pore water chemistry does hardly affect the swelling pressure as long as the bulk density is higher than about 1.95 t/m^3

2. At lower densities, the effect of salt water on the swelling pressure is very obvious: with oceanic salinity of the pore water, the swelling pressure will be 2-5 times lower than that produced by fresh water

3. Ca bentonite tends to give the same swelling pressure as Na bentonite. Actually, Ca bentonite tends to give a slightly higher pressure at high densities

Additional observations are that soda activated bentonites (Greekish, Bulgarian, and Sardinian) fit nicely with the MX-80 although their liquid limits are only 60 - 90 % of that of MX-80 (cf. Fig.22). This suggests that the liquid limit is not a determinant of the swelling pressure, which is actually clearly demonstrated by the Ca bentonites. One explanation of this rather odd behavior of such bentonites may be that a major contribution to their swelling pressure is given by elastic strain built in by the compression of the clay powder in the oedometer. This would also imply that the continuity of stiff structural units (domains) is more effective in Ca than in Na clays at one and the same void ratio, which would explain that also slight hydration generates high pressures.

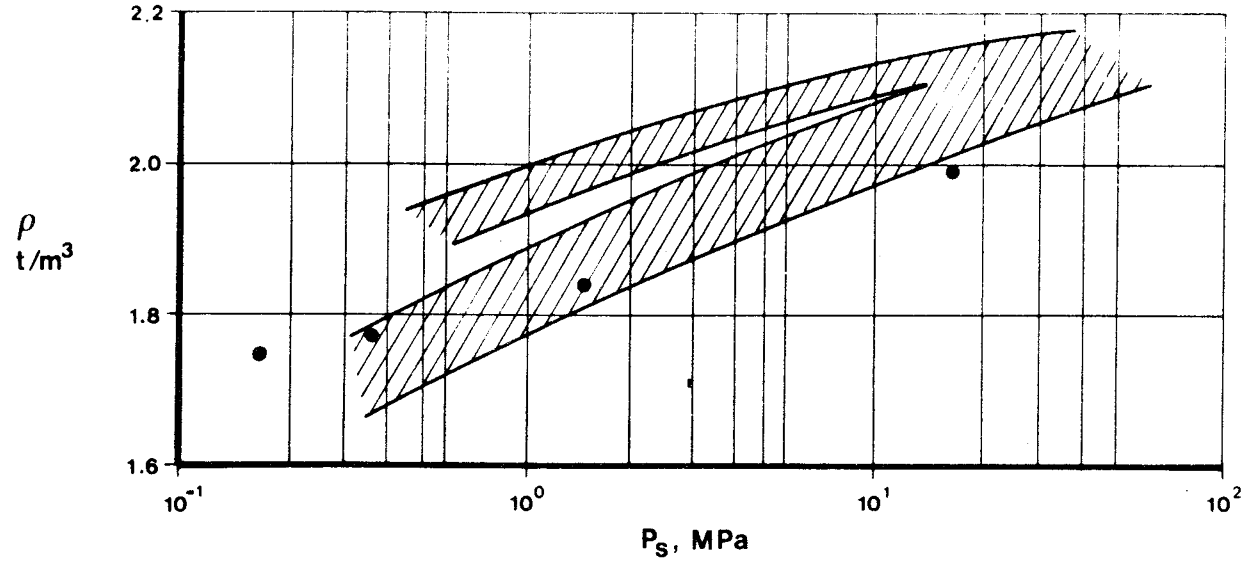


Fig.22. Swelling pressure p_s versus bulk density of water saturated bentonite. The bands represent MX-80; the upper branch corresponding to ocean salinity, the lower to fresh water ("Allard" and distilled). The black dots represent commercial bentonites of various brands.

Although the various bentonites did not show a clear difference in swelling pressure at higher bulk densities as we can see from the diagram in Fig.22, the scattering is very obvious at low densities (Fig.23). This has an impact on the swelling ability of mixed backfills since the clay component forms such a small fraction of the total mineral mass that the bulk behavior becomes very sensitive to variations in swelling power of the clay. The choice of a suitable bentonite for backfilling purposes should therefore always be based on experimental determination of the swelling properties of candidate bentonites.

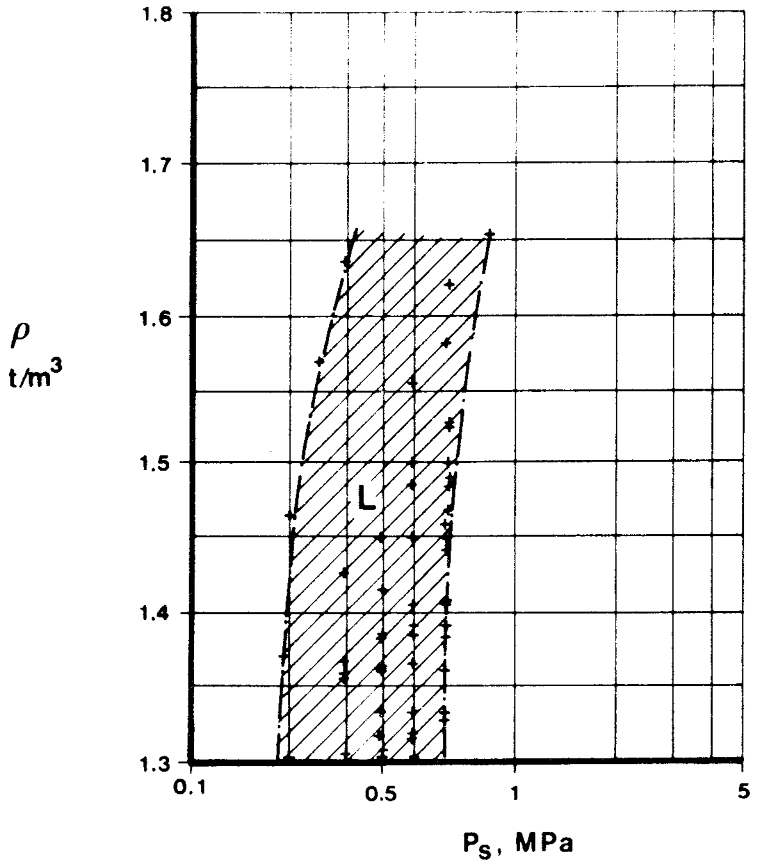


Fig.23. Variation in swelling pressure of natural Na smectites. Compilation of data published by Philip Low (22).

Some of the recent work at the Div. of Soil Mechanics, University of Luleå, has involved reconsideration of earlier and evaluation of recent data, which has yielded the updated, explicit correlation between swelling pressure and bulk density of MX-80 given in Table 6. The influence of temperature has been indicated as well.

Table 6. Swelling pressure p in MPa versus bulk density of MX-80 at different temperatures

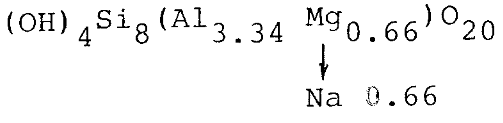
Bulk density t/m^3	p_s, MPa		
	20°C	90°C	70°C (estim)
2.15	45	35	40
2.10	30	17	20
2.05	15	8	10
2.00	7	4	5
1.95	4.5	2.5	3

Chemical composition

As for the illite case, the mineral composition and the organic content are of primary importance.

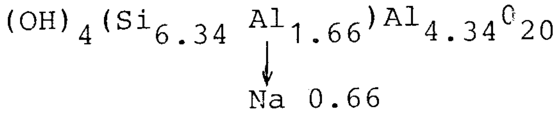
A key question, which has to do with the chemical stability of bentonites in repository environment, is whether the smectite component is montmorillonitic or beidellitic, since the latter has the charge properties that make it vulnerable to illite conversion. The identification has to be based on chemical analyses, the characteristic compositions* being:

Montmorillonite



$(OH)_4Si_8(Al_{3.34}\,\underset{\substack{\downarrow\\ Na\ 0.66}}{Mg_{0.66}})O_{20}$

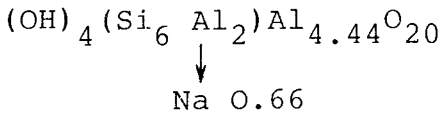
Beidellite



$(OH)_4(Si_{6.34}\,\underset{\substack{\downarrow\\ Na\ 0.66}}{Al_{1.66}})Al_{4.34}O_{20}$

or

Beidellite



$(OH)_4(Si_6\,\underset{\substack{\downarrow\\ Na\ 0.66}}{Al_2})Al_{4.44}O_{20}$

---

* After Ralph E. Grim

In the above formula, arrows are placed below the groups with charge deficiency, thus requiring the addition of cations external to the crystal lattice to balance the structure. Sodium has been picked here as balancing adsorbed cation.

The potential risk of beidellite being transformed to illite, which takes place if potassium is available, suggests that only montmorillonitic bentonites should be considered. A further restriction is that K-bearing minerals, such as K-feldspar and biotite, should be sparse in candidate bentonites at least if the temperature is expected to approach or exceed 100°C. Since these minerals typically represent silt- and sand-sized particles, they are automatically "sieved" out in the most fine-grained bentonites.

The MX-80 material is a typical exponent of bentonites that are very suitable for use as buffer material in repositories. The clay fraction holds montmorillonite with mostly sodium in exchange positions and only traces of quartz, kaolinite and zeolites. The silt and sand fractions, which are very small, hold quartz, feldspars, micas, pyrite, iron oxide and hydroxide, calcite, apatite, glassy components and organics. Various investigations (cf. 23) have shown that bentonites with very similar compositions, i.e."Wyoming-type montmorillonites", are available in Japan, Argentina, and elsewhere in the US.

Large European bentonite deposits, which are exploited today and which can be regarded as potential resources, show some deviations from the Wyoming beds as is illustrated by Table 7. As long as the clay content is high and their smectite content is also high, such deviations are not important with the exception of the content of sulphides and sulphates. If this content is expressed in terms of total sulphur, it turns out to range between 0.01 % and about 0.3 % for most bentonites. Bentonites with very low S contents are to be preferred as "overpacks", while this content is not critical for backfill components or sealing plugs.

Table 7. Chemical constitution of commercial bentonites

Compound %	MX-80	Bavarian	Greekish	Bulgarian	Cyprian	Sardinian
SiO_2	63.0	58.3	69.0	59.5	57.9	70.0
TiO_2	0.1	0.4	0.3	0.9	-	-
Al_2O_3	16.1	20.1	14.1	15.2	14.6	15.9
Fe_2O_3	3.0	5.4	2.4	5.1	5.6	2.0
MgO	1.6	4.2	2.2	3.1	1.6	2.1
CaO	1.1	1.9	2.0	5.3	1.1	1.7
NaO_2	2.2	0.1	2.9	1.7	1.0	1.1
K_2O	0.5	2.0	0.5	0.8	1.2	0.3

The organic material in bentonites originates from three sources: present vegetation on site, carbon-type contamination from heating during the processing, and original compounds derived from microorganisms etc. that were entrapped when the sediment was deposited. The two first-mentioned, which may well form the major part in to-day's production of several plants, can be avoided by proper handling. The inhereted organics, which seem to be kerogen-type end products of the decomposition outlined earlier in the text (cf. 24), are practically absent in certain bentonites while they amount to several thousand ppm in others.

Since the organic content should be kept low, i.e. below 200 ppm for the KBS concepts, only a limited number of commercial bentonites qualify. It has been demonstrated, however, that the organic content can be reduced to the required upper limit by heating to about 400°C, and that this treatment does not affect the swelling characteristics of montmorillonite (25). Thus, heating to 425°C of air-dry montmorillonite for 15 hours did not cause permanent breakdown of the crystal lattices, merely a slight weakening of the reflections. Such weakening became obvious when the temperature was raised to 600°C for 6 hours, however.

It is anticipated that much of the organic matter in bentonites is strongly held at smectite crystal surfaces through hydrophilic functional groups, and that it cannot be displaced by water flow under low and moderate hydraulic gradients. The even smaller and less continuous void passages in smectite clays than in ilite means that it is virtually impossible for most bacteria to find their way through highly compacted bentonite and to get access to the well confined and widely spaced organic complexes that might serve as nutrients (25).

4. CHARACTERIZATION OF CLAY MATERIALS

4.1 General

The main criteria for the selection of suitable clay materials for repositories imply that the chemical constitution be identified with respect to the art and content of certain specified minerals and organic constituents, and that the swelling and sealing ability are checked. Only smectitic clays are considered here.

4.2 Chemical composition

The checklist with respect to chemistry is:

a. General mineralogy

X-ray diffraction analysis is required to demonstrate the presence of smectite minerals and to get a general picture of the mineral composition with particular respect to calcite, K feldspar, and biotite.

b. Type of smectite

For use as "overpack", chemical analysis is required to verify that montmorillonite is the main smectite mineral.

c. Organic material

For use as "overpack", the organic content has to be determined.
The rather low content of organic matter in the bentonites of potential use point to the need of chemical analyses, since the standard procedure in soil mechanics of determining the ignition loss will be far from accurate. Wet combustion techniques for determination of the total carbon content are recommended.

4.3 Physical properties

A general characterization is required with respect to the granulometrical composition, since it yields clear information of the content of clay-sized particles and thereby, indirectly, of the amounts of smectite and rock-forming minerals. Subsequently, special tests are applied to characterize the clay with reference to its swelling and sealing properties. A suitable checklist would be:

a. Clay content

The clay is dispersed in distilled water and effectively stirred without adding dispersing agents and without applying ultrasonic treatment. By following this recommendation, possible cementation can be identified.

The clay content is suitably determined by applying the Andreasen pipette method although the hydrometer method can usually be accepted as well.

b. Swelling ability

For use as "overpack" or as plug material in boreholes, shafts, and tunnels, the swelling pressure should be determined by using an adequate swelling pressure oedometer, preferably the LuH type (Fig.13). In selecting a suitable clay for backfilling purposes, where the swelling pressure will be in the low range, an oedometer with a continuously adjusted counter-pressure is preferable.

c. Sealing ability

Permeability tests are very tedious and require an experienced laboratory staff and special equipment. It is therefore recommended that an indirect measure of the hydraulic conductivity is used, and for this purpose the liquid limit can be used. Since the Casagrande method and the Swedish cone penetration test yield substantially different values, it is recommended that one of them is selected for standard testing. It should be the cone method since it is much simpler and much more rapid and sufficiently accurate.

It is important to notice that certain clays that have been investigated in the current study, have yielded rather low liquid limits although their X-ray fingerprints definitely show that they are smectite-rich and also show correspondingly low hydraulic conductivities. This discrepancy is caused by cementation which is strong enough to withstand major breakdown at the liquid limit testing, but which allows a sufficient amount of smectite flakes to be separated to give nice X-ray peak displacement at ethylene glycol treatment, and to give a low hydraulic conductivity. In consequence with this the swelling pressure turned out to be low. Such rebelling clays need to be revealed and this requires that the swelling pressure be determined. An alternative, very simple check of the swelling ability can be used in the current quality control as shown in the subsequent text.

4.4 Current quality checking

Once the clay material has been selected, regular checking is required and usually there is very little time for running the tests. This rules out detailed mineral analyses, oedometer tests etc. and leaves only a few simple investigations which have to be conducted in large numbers in a few days only. They should comprise determination of the natural water content, which is required to select proper compaction pressures in the production of highly compacted blocks, and to add a correct amount of water in preparing backfills; the determination of the liquid limit; and the Enslin swelling test.

The need of making rapid and yet sufficiently accurate checking of the swelling ability, i.e. of the hydration capacity, which in turn is measure of the surface activity and therefore of the smectite content, was realized early by manufacturers of bentonite products. Various techniques, such as determination of the adsorption power of certain organic dye substances, have been in use for a long time in the laboratories of the processing plants. The most suitable method seems to be the Enslin test, which is presently being tried at the Swedish State Power Board. It deserves a short presentation.

4.4.1 The Enslin test

The original apparatus described by ENSLIN (26) in 1933 consisted of two funnels connected by a U-shaped tube, while the present device has the shape shown in Fig.24. To make the test, a weighed amount of air-dry clay is placed on the filter plate which is in contact with distilled water that fills the system and terminates in the horizontal capillary.

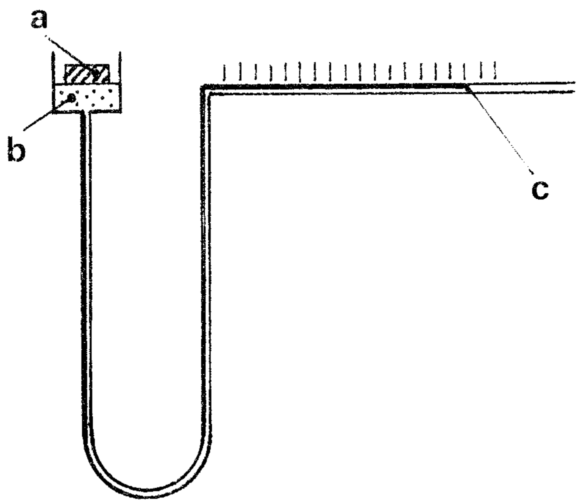


Fig.24. Schematic picture of the Enslin device.
a) Sample, b) Porous filter, c) Water meniscus

A suitable amount of dry clay is 0.5 to a few grams and the time for suction is usually 1000 minutes (27), although a somewhat longer time may be required for the identification of possible significant differences in the smectite content in a large series of samples.

The result is plotted as the water uptake, directly read from the scale of the capillary tube, versus time, as illustrated by Fig.25.

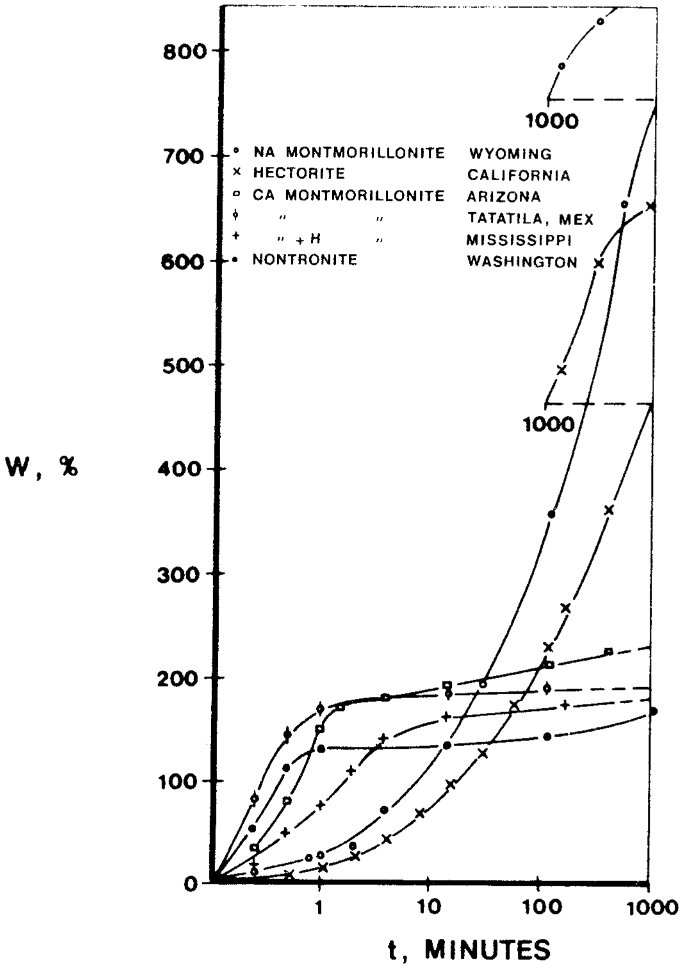


Fig.25. Water sorption curves for smectitic clays (27).

5. EFFECT OF THE ADDITION OF BALLAST

5.1 General

No clay mineral occurs in a perfectly pure form in nature. Thus, contamination by other clay mineral representatives as well as rock-forming minerals has always taken place, either in the course of the deposition of sedimentary materials or as a result of transient weathering processes of originally heterogeneous soil or rock. This means that we always deal with mineral mixtures when clays are considered, and that synthetic mixtures such as backfills involves a further decrease in clay mineral concentration. When this concentration drops below a few tens of percent of the total mineral mass, the governing role of the clay substance with respect to the physico/mechanical properties in bulk, is successively lost. The matter is of great practical and economical importance and is almost entirely dealt with empirically in practice. Some examples will be presented in this chapter, which also gives a short theoretical introduction.

5.2 Theoretical aspects

We will disregard here, to start with, from the practical difficulties of obtaining ideal distributions of the various constituents of soil mixtures. Thus, it will be assumed that voids between larger contacting grains can be occupied by smaller grains, without facing the fact that mixing involves some clay-smearing of the surfaces of the larger grains.

The goal of arriving at a very low hydraulic conductivity can be reached by filling the voids between larger grains with a smectite gel of very low density. Actually, if such a gel completely filled up the space it could have a bulk density of only about 1.3 t/m^3 to give the mass a hydraulic conductivity of less than 10^{-10} m/s, which is an order of magnitude lower than what is specified for backfills according to the KBS 3 concept. In fact, at the low gradients that are considered in sealed repositories, an effective sealing can be offered by even softer gels. A swelling ability on the other hand, requires that the swelling pressure exceeds the own weight of the backfill, and this usually means that the density of the clay gel needs to be higher.

The rather extreme sealing effect of the relatively expensive smectite clays, calls for the selection of a grain size distribution of the coarser components so as to yield very small voids in which clay aggregates fit. A dense clay skin covering those parts of larger grains that are not occupied in forming intergranular contacts would also be required in order to minimize the hydraulic conductivity.

Starting from a theoretical approach we know that spherical grains can be arranged in various regular patterns with different porosities (Fig.26). Compaction means that pressure and shear produce displacements within the particle assembly, and the more or less spherical grains are brought closer, approaching a minimum porosity of 25 - 30 % and to less than 25 % if a certain fraction of the grains are slightly oblate.

The diameter of spheres that can be inscribed in the remaining pores is theoretically 0.154 d where d is the diameter of the spheres forming the primary grain system if it is in the tightest state of packing. Assuming these spheres to be represented by very well rounded 2 mm diameter sand grains with a high degree of sphericity, a considerable part of the pore volume would thus be occupied by introducing silt grains with an average diameter of 0.3 mm. The net porosity is reduced to less than 20 % by this operation, the ultimately remaining pores having dimensions in which 0.05 mm diameter spheres fit.

The theoretical concepts cannot be applied too literally because we do not deal with ideal spheres and packing cannot be achieved in a wholly systematic manner. It is concluded, however, that if the mixture is composed of a primary ballast material of sand grains with diameters ranging between 0.1 and 2 mm, with some additional fines added, the major part of the remaining voids can be filled with a very moderate amount of bentonite powder. Per m^3 of water saturated, compacted mixture this should represent about 0.2 m^3 of clay with a bulk density of 1.3 t/m^3, which thus represents about 0.26 t clay gel per m^3 of the mixture. This corresponds to a few weight percent of air-dry bentonite in preparing the mixture. In practice, such low clay concentrations have been applied frequently with great success. For repository purposes it is required to use a slightly higher percentage, however, partly because of the difficulty in preparing large amounts of homogeneous mixtures with very low clay contents, partly because a slight excess of clay yields particularly effective sealing since it tends to fill all the irregular, narrow voids which extend from larger pores (Fig.27). This is especially important when moderately expanding Ca smectite is used.

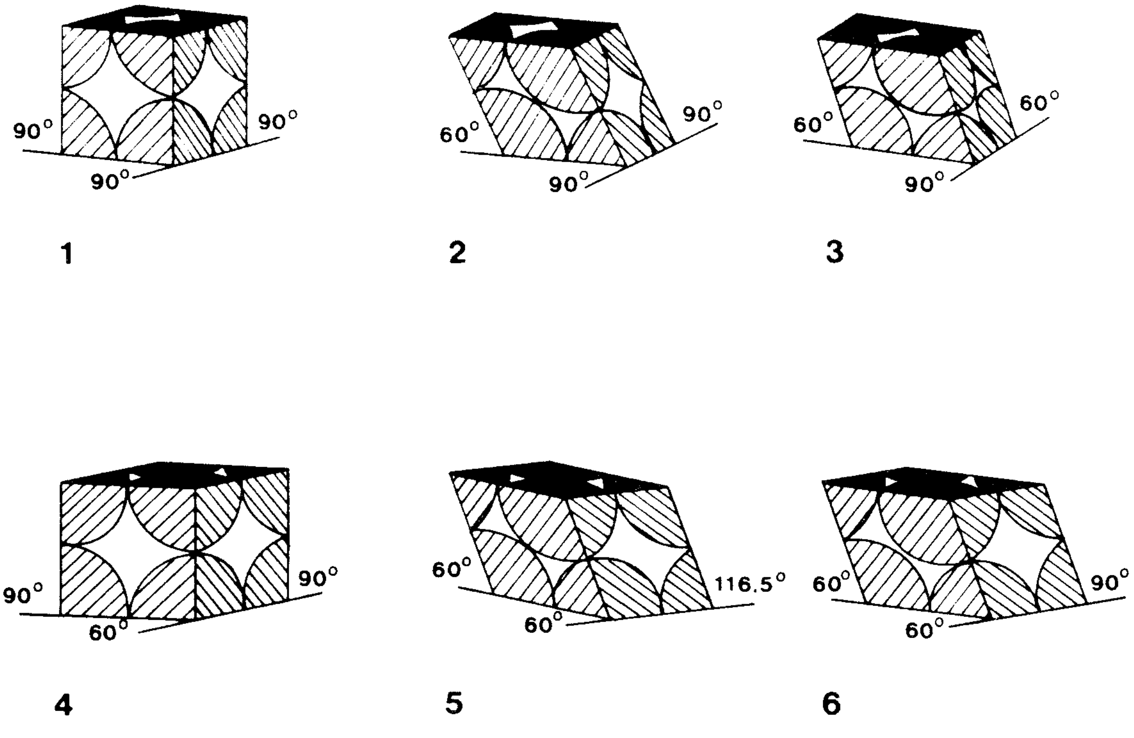


Fig.26. Schematic coordination modes of equally sized spheres (After Pettijohn). Case 1 is most open, cubical packing with n=0.48, while case 6 is the closest, rhombohedral packing with n=0.26.

n is the porosity expressed as the ratio of pore volume and total volume.

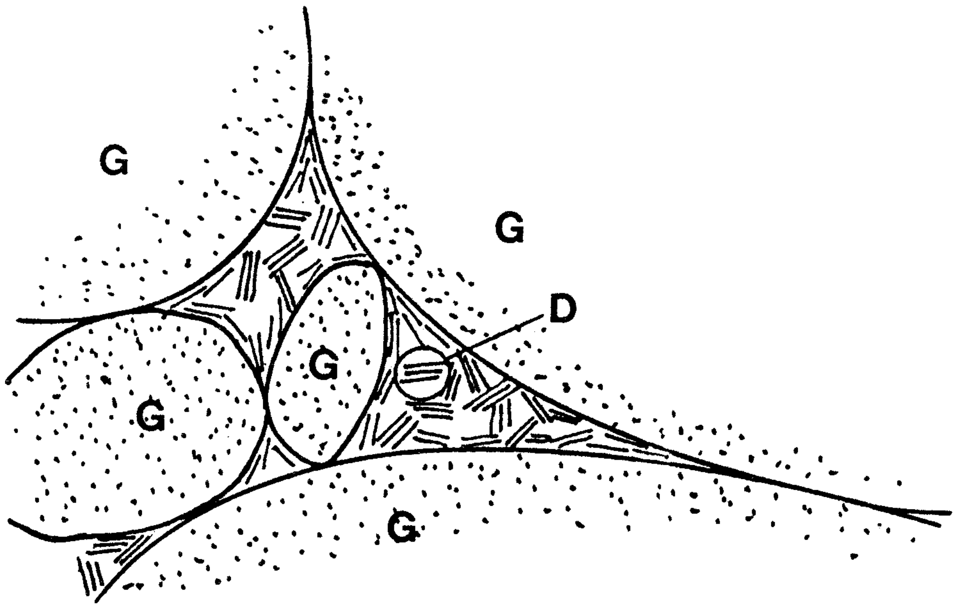


Fig.27. Microstructural features of clay/silt/sand mixtures. G= silt and sand grains, D= clay aggregates.

This amount of additional clay suggests that the clay content should be 5 - 10 weight percent of the total mineral mass to yield a bulk hydraulic conductivity of less than 10^{-10} m/s.

Actually, even slight swelling of a densely compacted backfill increases the pore volume dramatically, by which the density of the clay gel is reduced. Thus, a swelling of 10 % of the bulk mass involves 50 % volume increase of the space that hosts the clay gel in our example, whereby its swelling pressure is correspondingly reduced.

5.3 Practical experience

5.3.1 Consistency

Water sorption Enslin testing of mixtures of Na montmorillonite and well rounded, fine Ottawa sand suggests that when the montmorillonite content exceeds about 30 weight percent of the total mass, it governs the behavior of the mixture (Fig.28).

Determination of the plastic and liquid limits (Atterberg consistency limits) give a slightly different pattern. Thus, these quantities seem to be, approximately at least, in proportion to the clay content (Table 8).

Table 8. Atterberg consistency limits of quartz sand/montmorillonite mixtures (28)

Mixture		Liquid limit %	Plastic limit %
33 % Ca montmorillonite	(by weight)	53	18
50 % Ca montmorillonite	"	75	22
100 % Ca montmorillonite	"	140	50
33 % Na montmorillonite	"	122	19
50 % Na montmorillonite	"	214	24
100 % Na montmorillonite	"	475	47

The latter investigation illustrates the bulk properties of mixtures that were thoroughly remolded in a wet condition. This procedure yields a much more homogeneous distribution of the clay component than mixing of dry, aggregated clay with sand, and the influence of the clay on the bulk properties was therefore particularly obvious.

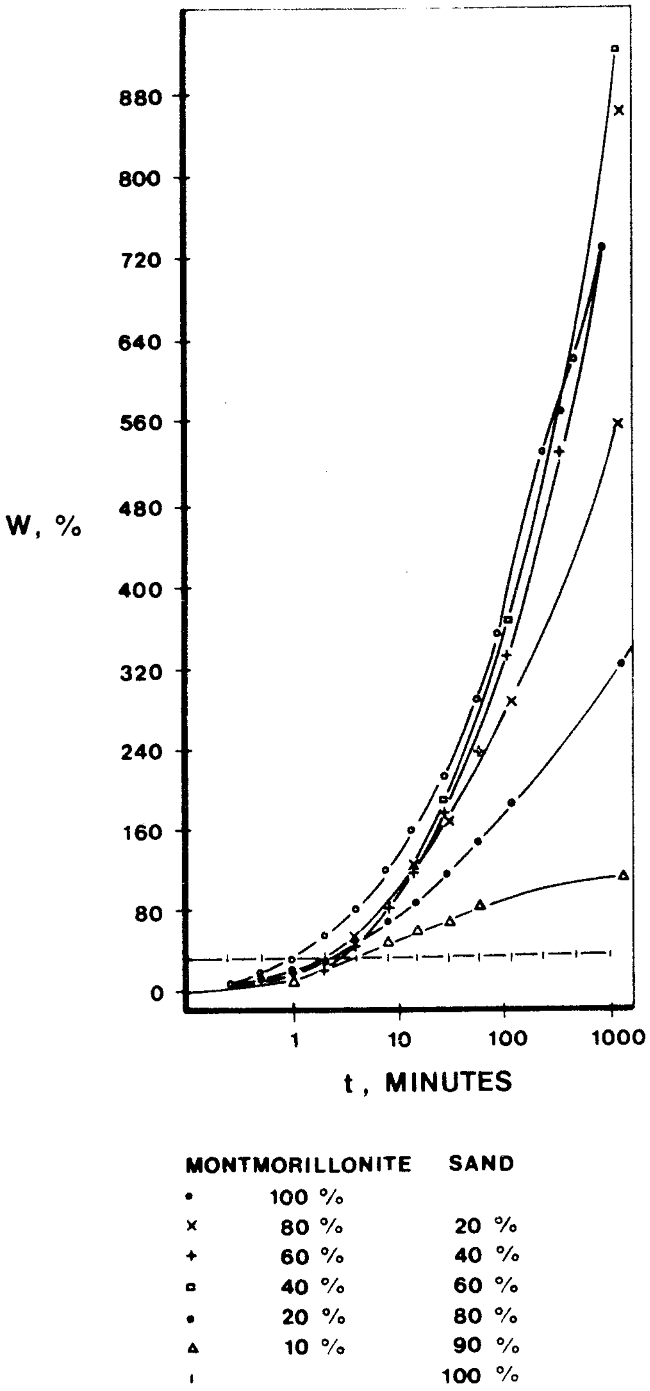


Fig.28. Water sorption characteristics of Na montmorillonite/sand mixtures (27).

5.3.2 Hydraulic conductivity

A large number of percolation tests on a laboratory scale have been conducted using MX-80 and silty sand. They show that the hydraulic conductivity is very much dependent on the bulk density, clay percentage, and hydraulic gradient (29, 30). Table 9 shows that the conductivity is surprisingly high, which illustrates that theoretically derived relationships of the kind we discussed in the preceding text, may not be very relevant. It should be noticed , however, that all the tests were run at high hydraulic gradients with some associated piping, and that the conductivity is probably an order of magnitude higher than we can expect at the very low gradients that will prevail in sealed repositories.

Table 9. Evaluated hydraulic conductivity from laboratory percolation tests with MX-80/ballast materials

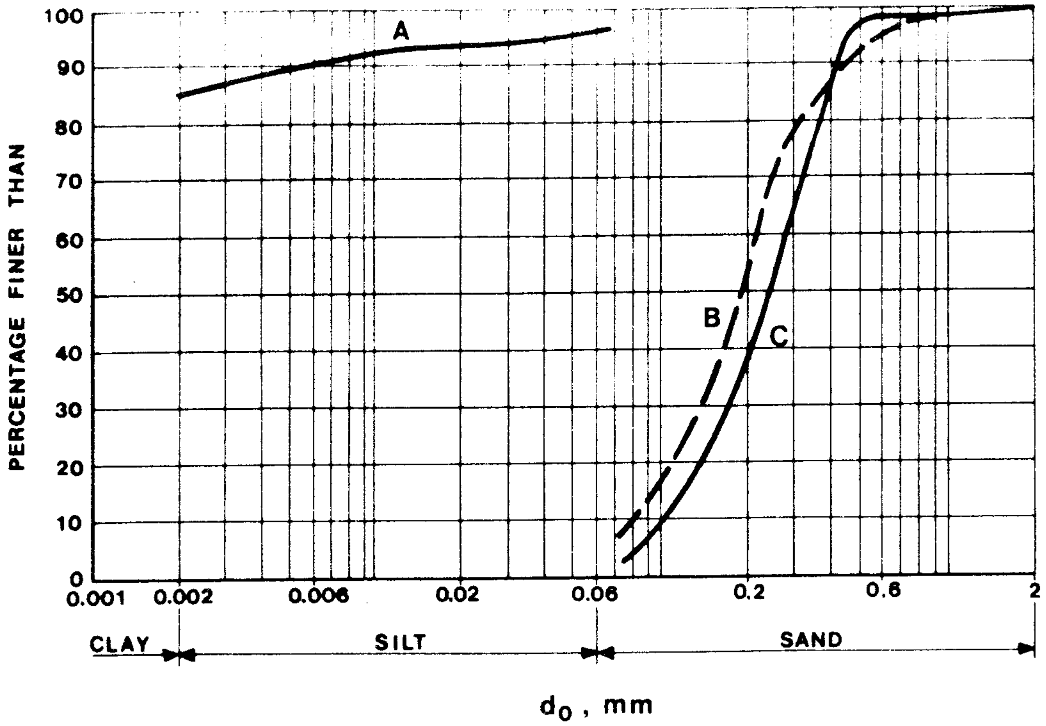
Clay percentage	Bulk density t/m^3	Hydraulic gradient	Hydraulic conductivity	Ballast
4	2.1	50	$3\cdot10^{-6}$	10% < 0.06 mm
4	2.1	25	$5\cdot10^{-7}$	35% < 0.2 "
8	2.1	50	$4\cdot10^{-8}$	80% < 1.0 "
8	2.1	25	$1.5\cdot10^{-8}$	100% < 6.0 "
10	2.1	100	10^{-9}	
16-18	2.15	100	10^{-9}	10% < 0.02 mm
16-18	2.15	25	$5\cdot10^{-10}$	40% < 0.2 "
20	2.0	100	$1.5\cdot10^{-10}$	90% < 1.0 "
20	2.1	100	10^{-10}	100% < 2.0 "

5.3.3 Swelling pressure

The swelling pressure of mixtures of MX-80 bentonite and silt/sand ballast has been determined by using swelling pressure oedometers with pressure transducers instead of the stress-release principle. Fig.29 shows the grain characteristics of the bentonite powder and of the granular composition of the ballast that was mixed with air-dry bentonite and compacted to various densities. The granulometrics of 10 % and 20 % bentonite mixtures are shown in Fig.30.

Fig.31 gives the general relationship between the swelling pressure and the bulk density of the "Allard" water saturated mixtures with the clay percentages 10, 20, 30, and 100. Probable curves for intermediate clay contents, shown as broken lines, are based on simple interpolation.

We see that for bulk densities of about 2.1 t/m^3, which are easily obtained in practice, the 10 % bentonite content yields a very low swelling pressure, i.e. less than 0.2 Mpa, while the 30 % bentonite content can be varied between slightly more than 0.1 and 0.7 MPa by varying the bulk density from about 2 to 2.15 t/m^3, or between 0.3 and 0.9 MPa by varying the bulk density between about 2.05 and 2.20 t/m^3. This points to the need of applying and compacting such relatively smectite-rich backfills in a controlled fashion so that large differential swelling pressures are avoided and that supporting constructions do not become overstressed. The insignificant swelling pressure of the 10 % bentonite mixture confirms the approximate validity of the physical model, which implies that 5 to 10 % air-dry bentonite would be required to fill the finest pores with a clay gel that has a bulk density of about 1.3 t/m^3. It is concluded from the earlier statements, that a Na bentonite gel of this density is expected to produce a swelling pressure of 0.1 to 0.4 MPa.



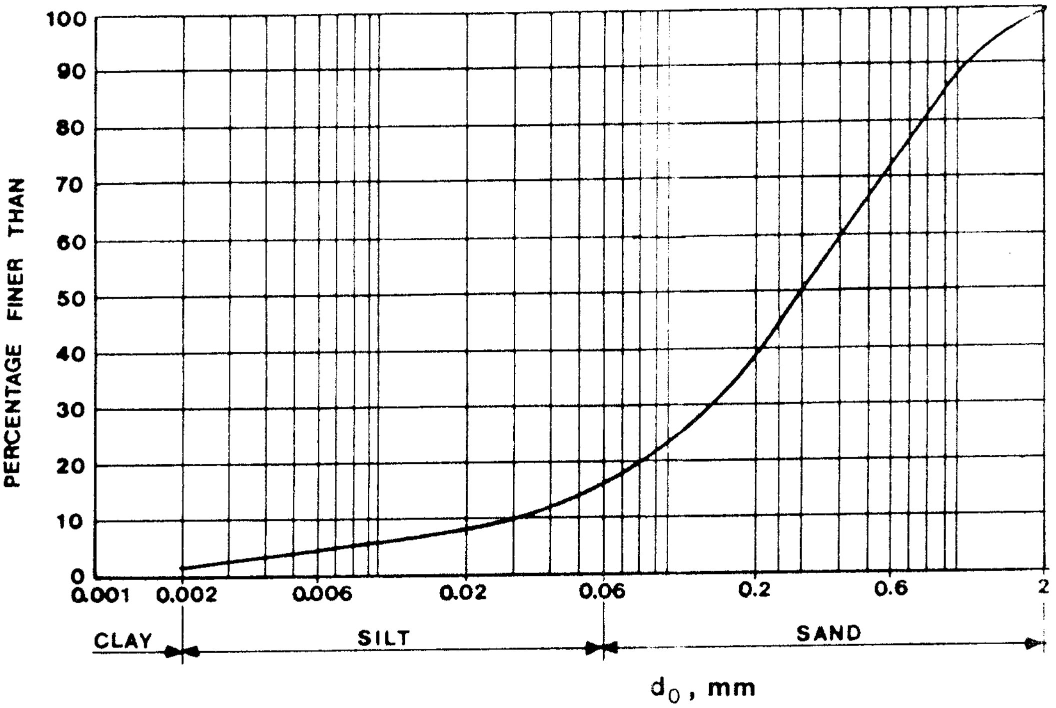
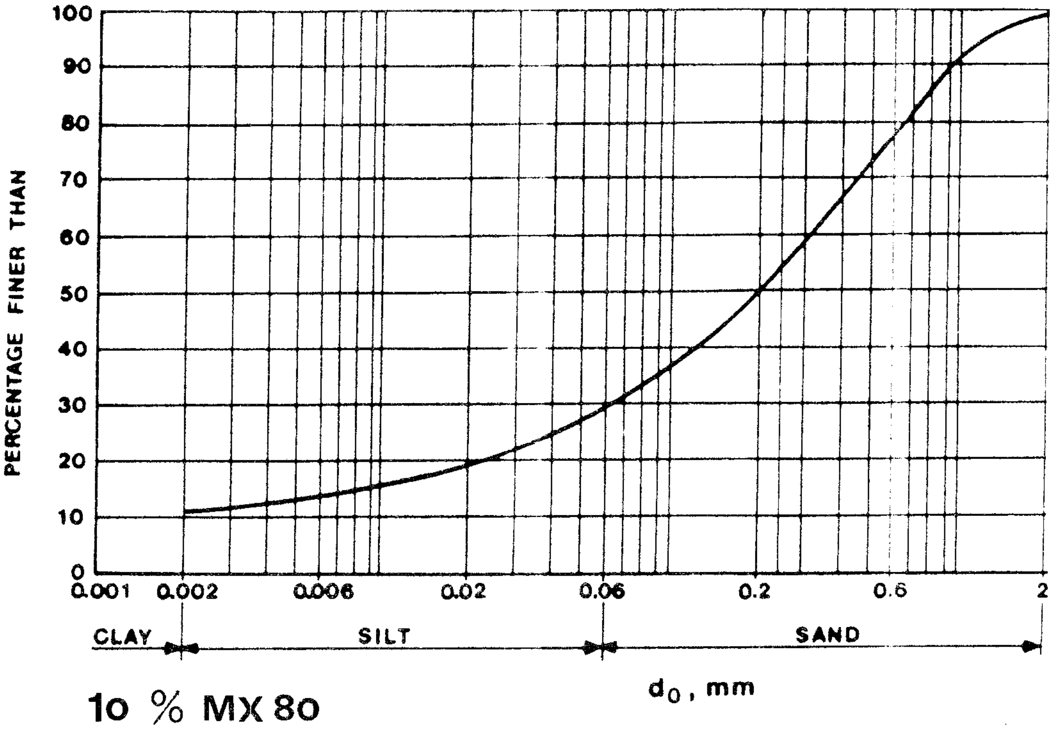


Fig.29. Upper diagram: Grain size characteristics of MX-80. A) sedimentation analyses after dispersion. B) and C) powder being sieved in air-dry condition.

Lower diagram: Grain size distribution of the ballast material.



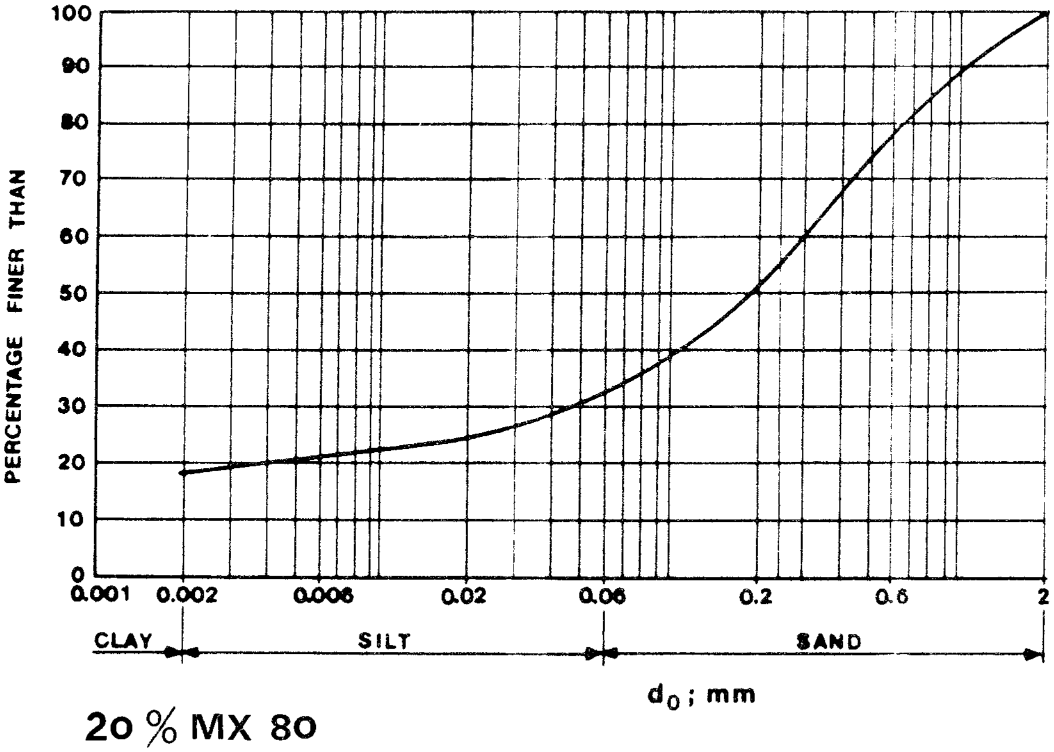


Fig.30. Grain characteristics of MX-80/ballast mixtures.

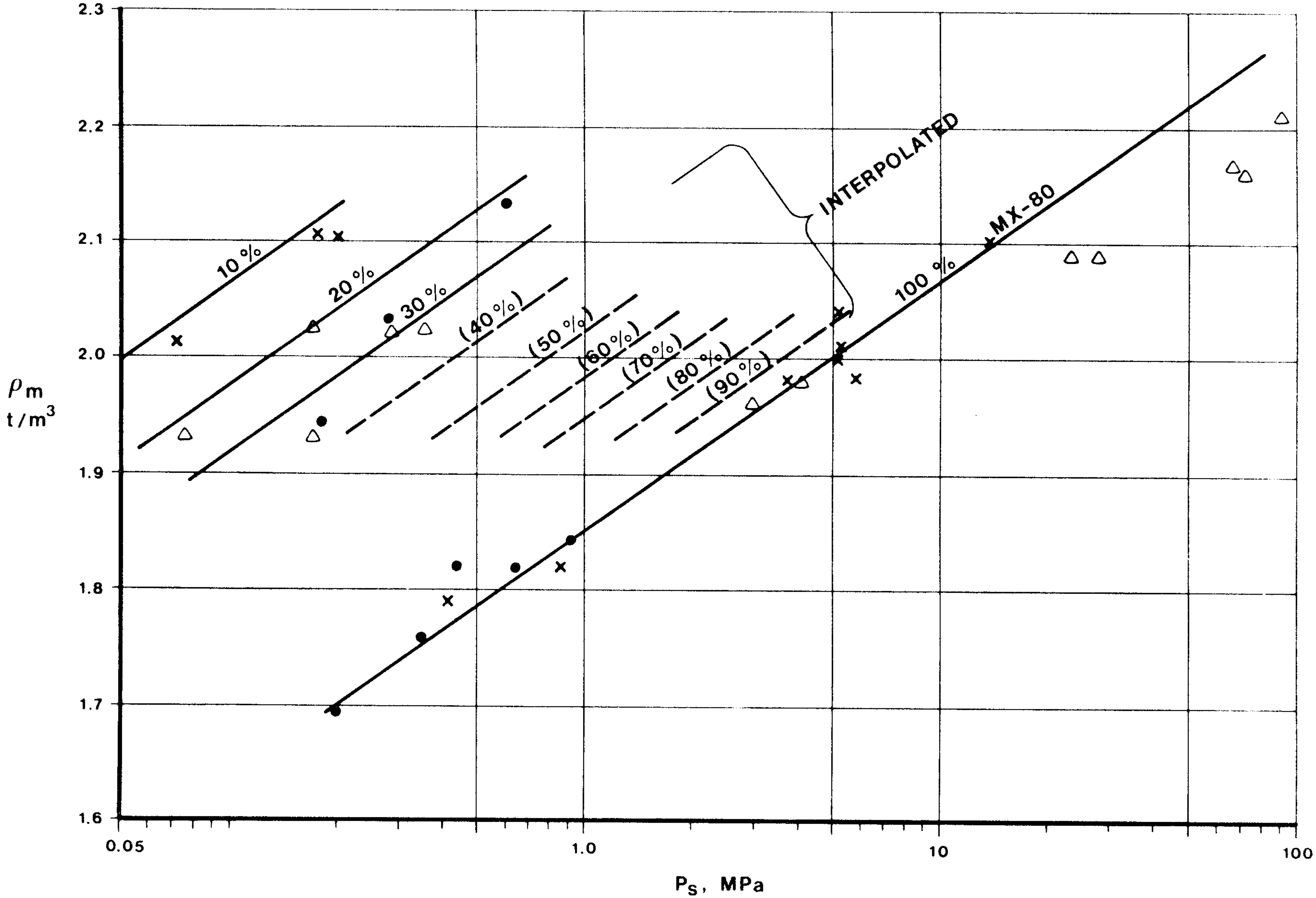


Fig.31. Swelling pressure versus bulk density for MX-80/ballast mixtures. After Lennart Börgesson.

5.3.4 Compaction properties

Experience from extensive earth dam construction activities in Sweden and elsewhere shows that homogeneous mixing of clay-based soil materials can be achieved also in large-scale operations, provided that the clay concentration is not too low. The Buffer Mass Test in Stripa offers an example of how effective such mixing can be when concrete mixers are used and how the mixed mass can be applied and compacted (19,20).

Fig.32 shows laboratory compaction curves for the Stripa BMT tunnel backfills (cf. Figs.29 and 30) with 10, 20, and 30 % bentonite mixtures. It is obvious that compaction on the dry side of optimum is hardly sensitive at all with respect to the water content when the bentonite percentage ranges from 10 to 20, while it is clearly affected also by small moisture changes when the bentonite forms 30 % of the total mass.

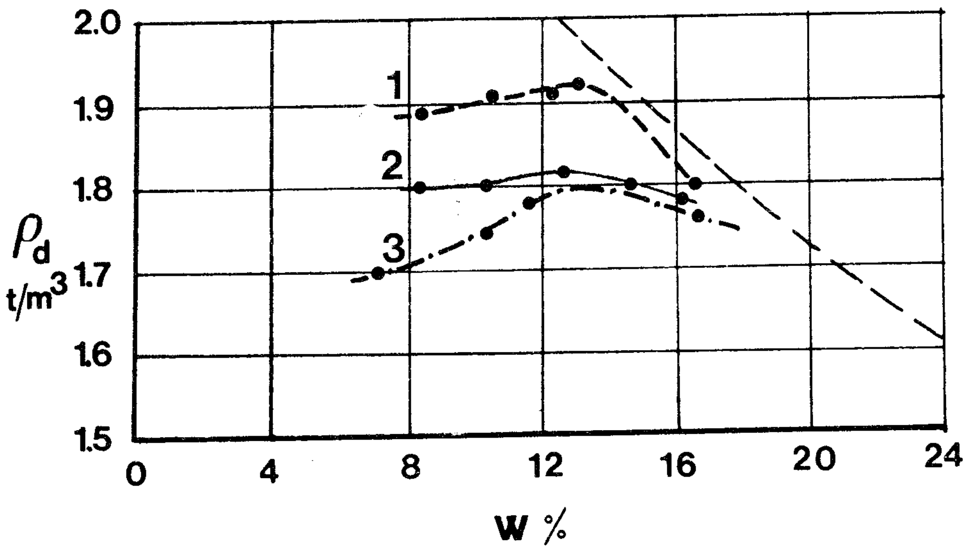


Fig.32. Compaction curves for the Stripa BMT backfills (Modified Proctor technique). 1) 10 % MX-80, 2) 20 % MX-80, 3) 30 % MX-80.

The expected bulk density of the in-situ compacted backfill with 10 % bentonite content was 2.15 - 2.20 t/m^3 after water saturation, while the average actual density will be slightly higher than that, as indicated by sampling on site. The fill was applied in 0.15 - 0.30 m layers and compacted by 10 - 15 runs with a 400 kg plate vibrator.

The backfill with 20 % bentonite content was applied by use of shotcreting technique. Its bulk density was found to be considerably lower than that of the 10 % mixture; actually, it will only be of the order of 1.8 - 1.9 t/m^3 after water saturation.

5.3.5 Compressibility

The compressibility of backfills is of practical importance where they are contacted by "overpacks" or plugs of strongly expanding clay. The traditional parameters m and β used in soil mechanics may well be used to describe the compressibility and to form the basis of the calculation of compression or displacement. Although it is anticipated that the stress exponent β does not deviate too much from unity and that the compression modulus m is in the interval of 100 - 2000 for low and moderate bentonite contents of heavily compacted backfills, there is still not sufficient experience to allow for the application of these values. A general idea of the compression characteristics of 10 % bentonite mixtures are offered by Fig.33, which relates to the Stripa BMT backfill.

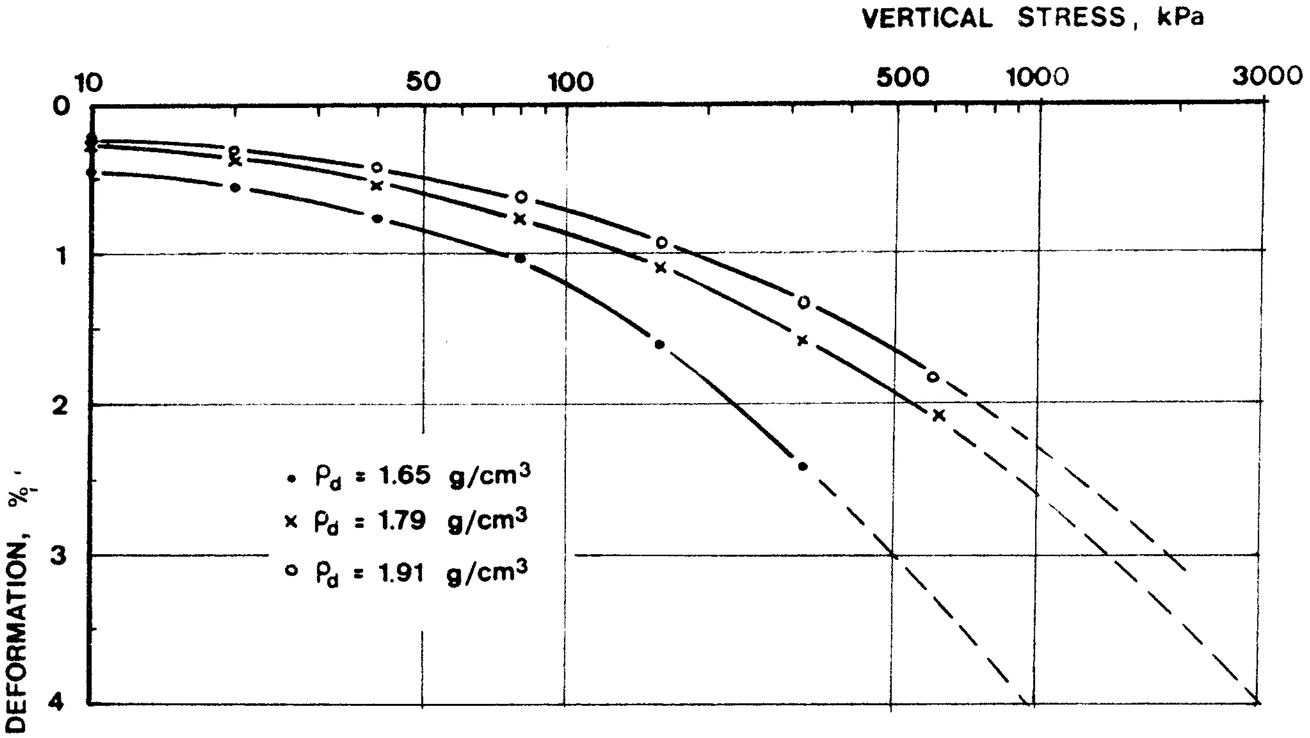


Fig.33. Compression characteristics of the 10 % bentonite Stripa BMT mixture at different dry densities.

6. CONCLUSIONS AND COMMENTS

The criteria specified for "overpacks" and expansive plugs unanimously point to the use of smectite-rich clay. Na montmorillonite has many advantages, particularly with respect to the hydraulic conductivity and the swelling and self-healing ability, and it is therefore suggested as the No.1 candidate. This mineral is the dominant constituent of the KBS reference clay material, the MX-80 bentonite, which has excellent properties with the possible exception of the contents of sulphur and organic matter. There are a few other bentonites which may serve equally well, the major difference being that the MX-80 material is derived from natural Na bentonite, while the competitors are Ca saturated and need to be transformed to the desired Na state by soda treatment. The carbonates resulting from this treatment form such a small part of the solid mass that they are expected to have a negligible influence on the physico/mechanical properties of the activated clay. Sufficiently effective removal of organics presents no serious problem.

Another difference is the slightly lower contents of clay-sized particles and montmorillonite in the latter bentonites, which are commercially available in Europe. This is manifested by the their lower liquid limit and the slightly higher hydraulic conductivity. The difference is negligible from a practical point of view, however, since the hydraulic conductivity will still be less than about $5 \cdot 10^{-13}$ m/s for very high gradients and bulk densities exceeding 1.9 t/m. The KBS 3 concept refers to the figure 10^{-13} m/s, which is the anticipated value also for the activated bentonites at the very low hydraulic gradients that prevail in sealed repositories.

When it comes to backfilling, the major object of which is to replace excavated rock by a medium with the same or with a lower conductivity, the requirements are much more moderate. Thus, natural illitic, late-glacial clays; dried, ground, and applied with subsequent compaction, are sufficiently impervious to be accepted for many backfilling purposes, although Na bentonite-based mixtures are superior. Since illitic clay powder requires effective compaction, it cannot be used to fill the top part of tunnels and drifts, for which shotcreting needs to be used. The shotcreted backfill must also have a swelling ability in order to establish and maintain a firm contact with the confining rock and this calls for the use expanding clays. Ballast mixed with 20 to 30 % Na bentonite is suitable for this purpose. Non-activated Ca bentonite has a moderate swelling capacity and would therefore not meet the requirements equally well, unless the backfill can be very densely compacted.

A tentative scheme for the possible use of various clay materials is given in Table 10.

Table 10. Tentative use of clays in repositories of the KBS 3 type. Ranking scale 1-3, 1 being excellent, 3 being possibly acceptable.

Clay	Clay % of mixture	Treatment	Application	Ranking
Illitic, late-glacial	100	Grinding, drying	Tunnel & shaft backfill	2-3
Illitic, late-glacial	100	Grinding, drying *	"Overpack", comp. blocks	3
Ca smectite, low content	100	Grinding, drying	Tunnel & shaft backfill	1-2
Ca smectite, low content	100	Grinding, drying *	"Overpack", comp. blocks	2-3
Ca smectite, high content	10-50	Commercial bentonite	Tunnel & shaft backfill	1-2
Ca smectite, high content	100	Commercial bentonite *	"Overpack"& plugs comp. blocks	2
Na smectite, moder. content	20-100	Grinding, drying	Tunnel & shaft backfill	1
Na smectite high content	10-30	Commercial bentonite	Tunnel & shaft backfill	1
Na smectite high content	100	Commercial bentonite *	"Overpack"& plugs comp. blocks	1

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* stands for heating to remove organics

7. ACKNOWLEDGEMENTS

The present report summarizes a number of experimental investigations which have been run at the Division of Soil Mechanics, University of Luleå, under the guidance of the author, who wants to express his gratitude to the entire staff for their devoted work.

Special thanks are due to Kerstin Pousette who was responsible for most of the experiments and who made a very careful job.

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