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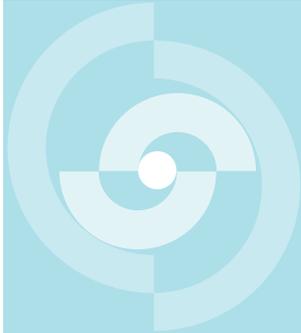
Nationale
Genossenschaft
für die Lagerung
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Cédra

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TECHNICAL REPORT 85-60

FINAL REPORT OF THE BUFFER
MASS TEST
Volume III: Chemical and physical stability
of the buffer materials

Roland Pusch

November 1985

Swedish Geological Co., Lund

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Der vorliegende Bericht betrifft eine Studie, die für das Stripa-Projekt ausgeführt wurde. Die Autoren haben ihre eigenen Ansichten und Schlussfolgerungen dargestellt. Diese müssen nicht unbedingt mit denjenigen des Auftraggebers übereinstimmen.

Le présent rapport a été préparé pour le projet de Stripa. Les opinions et conclusions présentées sont celles des auteurs et ne correspondent pas nécessairement à ceux du client.

This report concerns a study which was conducted for the Stripa Project. The conclusions and viewpoints presented in the report are those of the authors and do not necessarily coincide with those of the client.

Das Stripa-Projekt ist ein Projekt der Nuklearagentur der OECD. Unter internationaler Beteiligung werden von 1980-86 Forschungsarbeiten in einem unterirdischen Felslabor in Schweden durchgeführt. Diese sollen die Kenntnisse auf folgenden Gebieten erweitern:

- hydrogeologische und geochemische Messungen in Bohrlöchern
- Ausbreitung des Grundwassers und Transport von Radionukliden durch Klüfte im Gestein
- Verhalten von Materialien, welche zur Verfüllung und Versiegelung von Endlagern eingesetzt werden sollen
- Methoden zur zerstörungsfreien Ortung von Störzonen im Fels

Seitens der Schweiz beteiligt sich die Nagra an diesen Untersuchungen. Die technischen Berichte aus dem Stripa-Projekt erscheinen gleichzeitig in der NTB-Serie der Nagra.

The Stripa Project is organised as an autonomous project of the Nuclear Energy Agency of the OECD. In the period from 1980-86, an international cooperative programme of investigations is being carried out in an underground rock laboratory in Sweden. The aim of the work is to improve our knowledge in the following areas:

- hydrogeological and geochemical measurement methods in boreholes
- flow of groundwater and transport of radionuclides in fissured rock
- behaviour of backfilling and sealing materials in a real geological environment
- non-destructive methods for location of disturbed zones in the rock

Switzerland is represented in the Stripa Project by Nagra and the Stripa Project technical reports appear in the Nagra NTB series.

Le projet Stripa est un projet autonome de l'Agence de l'OCDE pour l'Energie Nucléaire. Il s'agit d'un programme de recherche avec participation internationale, qui sera réalisé entre 1980 et 1986 dans un laboratoire souterrain, en Suède. Le but de ces travaux est d'améliorer et d'étendre les connaissances dans les domaines suivants:

- mesures hydrogéologiques et géochimiques dans les puits de forage
- chimie des eaux souterraines à grande profondeur
- écoulement des eaux souterraines et transport des radionucléides dans les roches fracturées
- comportement des matériaux de colmatage et de scellement des dépôts finals
- méthodes de localisation non destructive des zones de perturbation de la roche

La Suisse est représentée dans le projet Stripa par la Cédra. Les rapports techniques du projet Stripa sont publiés dans la série des rapports techniques de la Cédra (NTB).

ZUSAMMENFASSUNG

Mit dem Puffer-mass-test bestand die Möglichkeit, die chemischen Aenderungen im Smektit-Anteil beim Heizen bis 125°C während ca. eines Jahres zu untersuchen. Die Veränderungen, die möglicherweise stattfinden könnten, wären eine geringe Aenderung der Ladungen im Kristallgitter mit Ausfällung von Silikat-Verbindungen, und eine Tendenz zur Illit-Bildung. Die Analyse zeigte, dass beide Prozesse auftraten, nicht aber in einem Ausmass, dass die physischen Eigenschaften beeinflusst wurden; dies wurde auch durch die Bestimmung des Quelldrucks und der hydraulischen Konduktivität nachgewiesen.

Der Puffer-mass-test zeigte auch, dass die Erodierbarkeit der auf Bentonit basierenden Verfüllmaterialien kleiner oder etwa gleich den theoretischen Erwartungen ist.

RESUME

Le "buffer mass test" (essai sur la masse-tampon) a offert la possibilité d'analyser si des températures portées jusqu'à 125°C pendant environ un an opéraient des changements chimiques dans la part de smectite. Les altérations que l'on pourrait éventuellement observer seraient une légère modification des charges dans le réseau cristallin avec précipitation de solutions de silicate ainsi qu'une tendance à la formation d'illite. L'analyse a montré que les deux processus se sont déroulés, mais pas au point d'influencer les caractéristiques physiques. C'est ce qu'a également prouvé la détermination de la pression de gonflement et de la conductivité hydraulique.

Cet essai a montré aussi que la propension des matériaux de colmatage à base de bentonite à s'éroder est plus faible ou à peu près égale à ce que l'on peut théoriquement attendre.

ABSTRACT

The Buffer Mass Test offered a possibility to investigate whether chemical changes took place in the smectite component at heating to 125°C for about one year. The alterations that could possibly take place were a slight charge change in the crystal gitter lattice with an associated precipitation of silica compounds, and a tendency of illite formation. The analysis showed that there were indications of both but to such a slight extent that the processes could not have affected the physical properties, which was also demonstrated by determining the swelling pressure and the hydraulic conductivity.

The BMT also showed that the erodibility of bentonite-based buffer materials is less than or about equal to what can be expected on theoretical grounds.

SUMMARY

In addition to the initial objective of the Buffer Mass Test to check the function of bentonite-based materials as barriers, it appeared to be possible to investigate whether chemical changes of the smectite component took place in an experiment where water saturated bentonite was heated to 125°C for about one year. Observations could also be made to find out whether groundwater erosion had taken place in the heater holes and tunnel.

Literature provides data which indicate that two major heat-induced effects can be expected at increased temperature. The first is a charge change caused by replacement of tetrahedral silica by aluminum and associated liberation and precipitation of silica compounds, the second is uptake and fixation of potassium by which the smectite is transformed to illite. The charge change, which is a necessary prerequisite of the illitization, was the only process expected to take place in the heater hole experiment, provided that the temperature was high enough. It cannot be unanimously decided from a theoretical point of view whether the charge change is a true kinetic Arrhenius-type process that takes place at any temperature, or if a critical threshold temperature must be exceeded, but both explanations would yield measurable alterations at about 125°C in one year.

The analyses comprised X-ray diffraction tests and chemical analyses as well as electron microscopy and determination of two characteristic physical properties, namely the swelling pressure and the hydraulic conductivity. The test results showed no clearly identified alterations except for a slight tendency of precipitation of supposed silica compounds and a possible indication of mixed-layer formation. The physical tests gave the same results as those obtained from non-heated samples.

The erodibility of smectite gels emerging from heater holes and propagating into water-percolated fractures was found to be less than expected. The explanation may be that the soft gel front exposed to the flowing water consisted of cohering flocs rather than of discrete, easily disrupted particles. The tunnel backfill consisting of sand/bentonite mixtures did not exhibit any sign of erosion at all, which is partly explained by the filter-type composition of this material.

1 OBJECTIVE

In the course of the Buffer Mass Test it became clear that the understanding of the physical processes involved in the water uptake and redistribution could be greatly improved if the temperature and temperature gradient were raised considerably in the clay overpack of one of the heaters. It was therefore decided to increase the power of heater no 1 from 600 to 1800 W after slightly more than 2 years of operation, which had yielded practically complete saturation. The experiment actually contributed to the physical modeling, although the net power was reduced to 1400 W in the almost 1 year long test due to breakdown of one heater element. The temperature of the clay ranged from about 127°C close to the heater to more than 70°C at the clay/rock interface in this period. As an additional option, the heating was considered to be sufficiently strong to make it meaningful to conduct a detailed analysis of the mineralogy and physical properties which could possibly have been affected. The matter of chemical stability forms the major part of Volume III, while a minor fraction of the report specifies the conclusions of the erodibility of the buffer materials, i.e. the physical stability.

2 CHEMICAL STABILITY

2.1 General aspects

The properties of smectite to serve as an effective permeability barrier and ion exchanger imply that the swelling behavior is retained. There are three major threats to this, the one of greatest concern being the transformation of smectite to illite or some other collapsed phase, such as kaolinite, as has been observed in natural bentonites (1, 2). Silification in the form of precipitation of heat-released silica may also reduce the expandability (3, 4), and a third

mechanism with the same negative effect may be precipitation of non-crystalline Al-rich phases (5). The latter is considered to be less important than the silica precipitation and will not be discussed here.

2.2 Illitization

Transformation of the smectite mineral montmorillonite to illite requires a charge change in the tetrahedral sheet by the substitution of Al for Si (beidellitization), and subsequent uptake and fixation of potassium into the high-charged layers (Fig 1). The matter has been considered in detail by a number of investigators, most of which based their conclusions on geological evidence in the form of observed mineral transformations in smectitic strata of large burial depth. Recently, Pytte concluded from detailed investigations of three American clay-bearing sedimentary rocks, heated by basalt dikes, that the observed monotonic increase in percent illite of mixed-layered clays results from a kinetic controlled conversion of smectite (6).

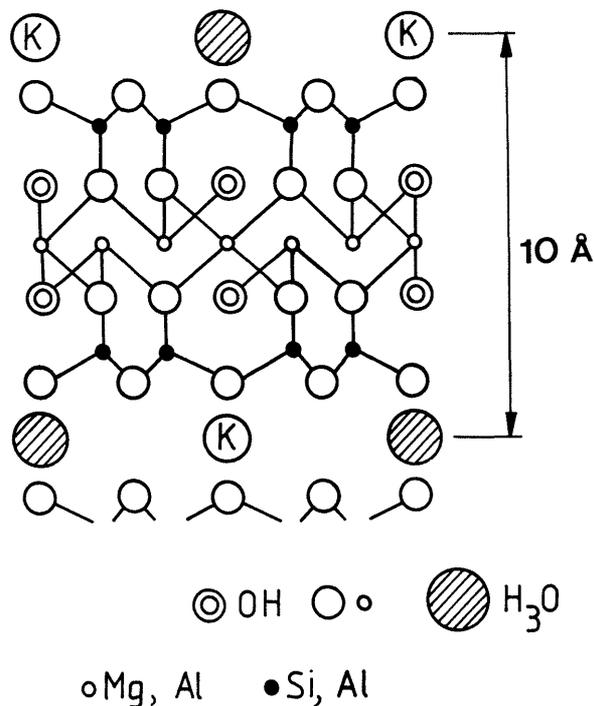


Fig 1. Crystal lattice model of illite ("hydrated mica")

The thermal history of these rocks was modeled and fitted by kinetic reaction models containing parameters for time, temperature, mole fraction of smectite, and pore-fluid chemistry. Reasonable agreement was obtained for activation energies of 27-33 Kcal/mol to yield the transformation and using such kinetic models a general relationship between time, temperature and percent illite was derived, a typical diagram being shown in Fig 2. It indicates that a temperature of 60°C in a closed system with K-bearing minerals would not yield a noticeable illite content in 100 000 years, while 130°C would convert most of the smectite in about 1000 years. At 150°C significant illitization is expected after 10-50 years according to this author.

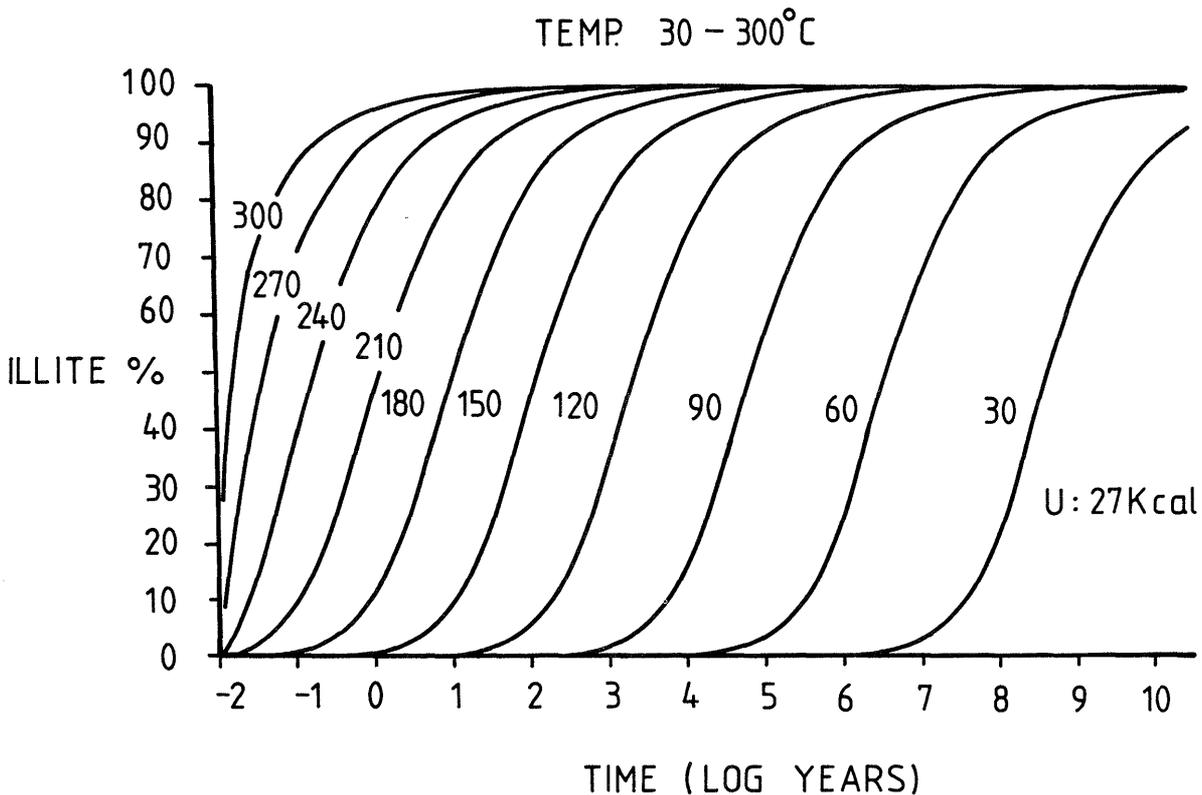


Fig 2. Illitization rates at different temperatures (6)

The rate-governing factor of the smectite/illite transformation is the access to potassium. Only for the special case of all K-ions required for complete S-I conversion being available in the close proximity of the smectite crystals, the rate of the charge change can be evaluated by determining the S/I-ratio of rock samples. In practice, however, external potassium is generally needed for the conversion and the process of K-migration from the surrounding rock is then a determinant of the illitization rate. This means that it is usually not possible to decide how fast the charge change actually took place by using diagrams of Pytte's sort or element analyses of rock samples. In the case of smectite-rich bentonites with a small content of K-bearing minerals, the availability and migration speed of potassium ions certainly governs the rate of transformation, while the charge change may well be rapid.

2.3 The charge change

The only phase of the smectite/illite conversion that can have been reached in the BMT case is a partial or complete charge change yielding beidellite, with possible associated release and precipitation of silica. It should be mentioned here that there was earlier some disagreement of whether potassium is required in the porewater to produce the layer charge or not, but recent experiments indicate that this process takes place also in water very poor in K (5). This finding is of fundamental importance since it excludes an alternative charge change mechanism, i.e. the one produced by substitution of divalent cations, such as Mg, for trivalent aluminum in the octahedral sheet. This would require an increase in positive charge through K-uptake in interlayer positions to preserve electrical neutrality, and thereby an illitization-like process.

The time required to produce the Si/Al replacement, in which diffusion of Si-ions over distances of several thousands of Å to the precipitation sites is involved, depends primarily on the temperature. The exact nature of the Al/Si replacement is not known but it can be assumed that Al ions are not primarily supplied from external sources but migrate from the octahedral layers to the tetrahedral from which Si is released and lost. In this context it should be mentioned that redistribution of structural units for crystal alteration may originate from certain intra-aggregate smectite lamellae, the term being "cannibalization" (4).

It has been suggested, and is still maintained by certain investigators, that there is a critical threshold temperature required to initiate the charge change. Thus, a recent summary of observed reaction sequences in the diagenesis of shales and low-temperature hydrothermal systems suggests 50°C to be a safe lower level, cf. Table 1, (5). Since the exact nature of the charge change is still not known, the critical level - if there is one - may be higher, but it is equally possible that it is an Arrhenius-type process that takes place also at ordinary rock temperatures albeit at a very slow rate. A deeper understanding of the reaction requires systematic autoclave experiments and such investigations have also been frequently reported in the literature. A few studies of this sort, all concerning relatively low temperatures, will be referred to in the subsequent text.

Table 1. Reaction sequence of shale diagenesis (5). K-uptake is assumed to have completed the reaction for each temperature interval

Temperature, °C	Reaction
< 50	None
50-90	Randomly interlayered illite/smectite (65 % illite)
90-150	Regularly ordered illite/smectite ISIS (50-80 % illite)
140-175	IIS-ordered illite/smectite (75-85 % illite)
> 200	Illite

In 1976 Eberl & Hower (7) concluded from autoclave tests using dilute smectite gels that an activation energy of 16-23 kcal/mol is required to break chemical bonds in the tetrahedral sheet so that aluminum can be substituted for silica thereby creating the charge change. They found that at a K/Na-ratio of 0.042 in the porewater, which is actually not too far from that of natural groundwaters, the percent expandable layers of Wyoming bentonite dropped from 100 % to 80 % in 78 days at 152°C and 20 MPa water pressure. According to these authors a transformation of 100 percent expandable smectite to 20 percent expandable I/S may require 10^6 years at 50°C and 10^3 years at 130°C. Extrapolation would give 10^2 years for the same transformation at 150°C, which in turn would yield a measurable change (5-10 %) in expandability at about 130°C after only about one year.

An indication of rapid smectite alteration at 150°C was recently attained by Howard & Roy by use of autoclave tests with Wyoming Na bentonite (8). Clay slurries with a 25:1 water to solid ratio were loaded into gold capsules and exposed to 150°C and 30 MPa for 1 to 6 months. After washing with distilled water the samples were analyzed by X-ray powder diffraction with and without ethylene glycol treat-

ment after which they were saturated with 1.0 N KCl-solution and then washed again by use of water-acetone mixtures. The extent of reaction was estimated from the illite/smectite mixed-layer expandability, which turned out to be very moderate for all the reacted samples. Several of them exhibited small diffuse X-ray reflections which may have originated from quartz formed as a new phase. This is compatible with the investigators' chemical analyses which showed that silica was released from smectite amounting to about 10-30 mmole/liter. Saturation with K gave a recognizable shift in the smectite basal reflections with respect to peak symmetry and spacing from which the percentage of expandable layers was evaluated (Table 2).

Table 2. Estimated percentage of expandable layers in heated Na-Wyoming bentonite (8). Artificial Na- or Ca-rich groundwater

Temperature °C	Duration, days	Average percent of expandable layers
150	30	90-100
150	90	75-90
150	120	75-85
150	180	70-83
250	30	83-90
250	90	74-90
250	120	75-80
250	180	58-65

It was concluded by the investigators that, at 150°C, the release of Si, Al, and Mg was stoichiometric, indicating that sufficient Al can be derived from the smectite alone to produce the charge change. For lower temperatures, i.e. 25-100°C, thermodynamic analyses of granite/bentonite systems suggest that Si release and precipitation is dominant (9).

The most interesting conclusion drawn by Howard & Roy was that the activation energy required for replacing Si by Al may only be a few kilocalories per mole. This suggests that very weak bonds operate in the tetrahedral sheets, which may be taken as a support of the temperature-induced crystal lattice change of montmorillonite that was originally suggested by Forslind & Jacobsson cf. (10). They claimed that the Edelman/Favejee (E/F) crystal concept is the most probable lattice structure at low and moderate temperatures, while the Endell/Hoffman/Wilm (E/H/W) model may be valid for higher temperatures (Fig 3). These investigators proposed a close clay/water interaction which may explain the swelling properties of smectite as well as the heat-induced charge change.

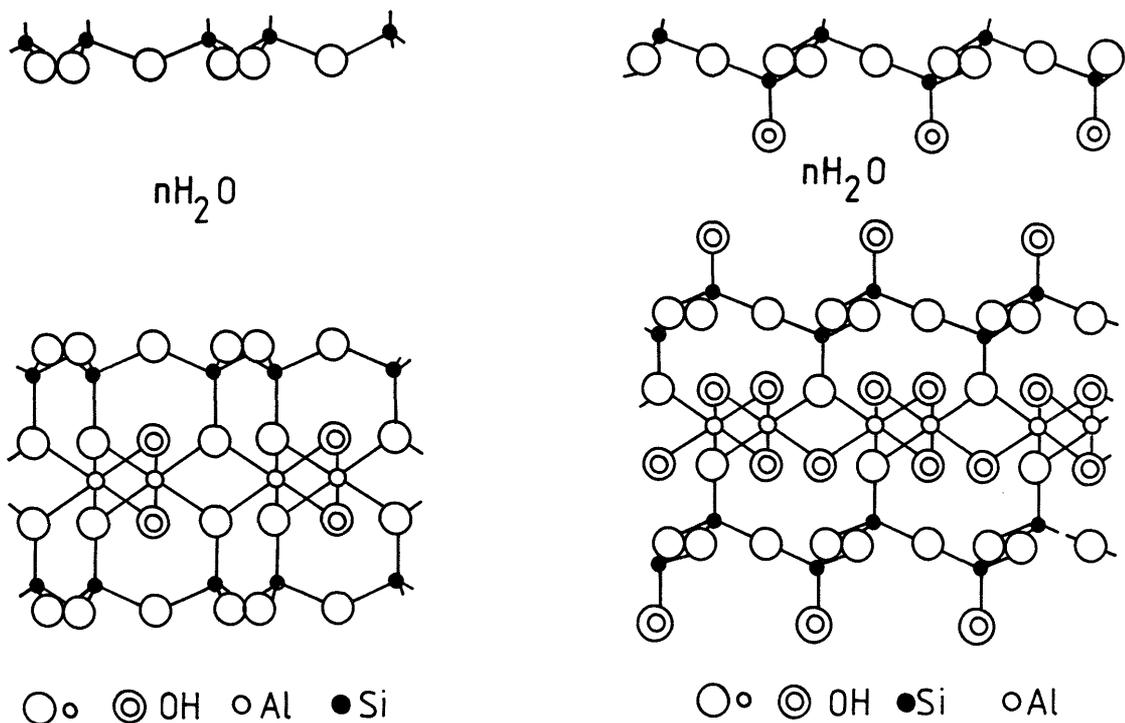


Fig 3. Crystal lattice models of montmorillonite.
 Left: The Endell/Hoffman/Wilm structure
 Right: The Edelman/Favejee structure

The clay-associated water lattice can be assumed to be of a strained ice lattice-type. The basal planes of the montmorillonite crystallites fit together with the water lattice as shown by Fig 4, which exhibits the protruding hydroxyls of the (001) plane with three unit cells marked, and Fig 5, which illustrates the basal molecules of an ice lattice superimposed on these hydroxyls.

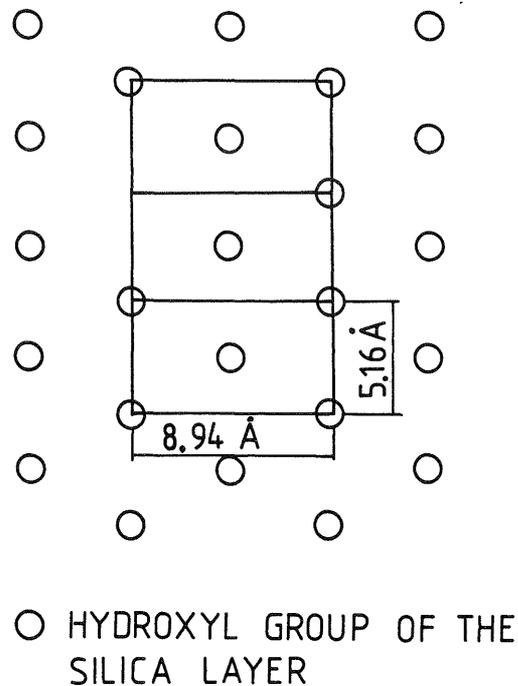


Fig 4. Protruding hydroxyls of (001) plane. The dimensions of one unit cell are marked (After Forslind)

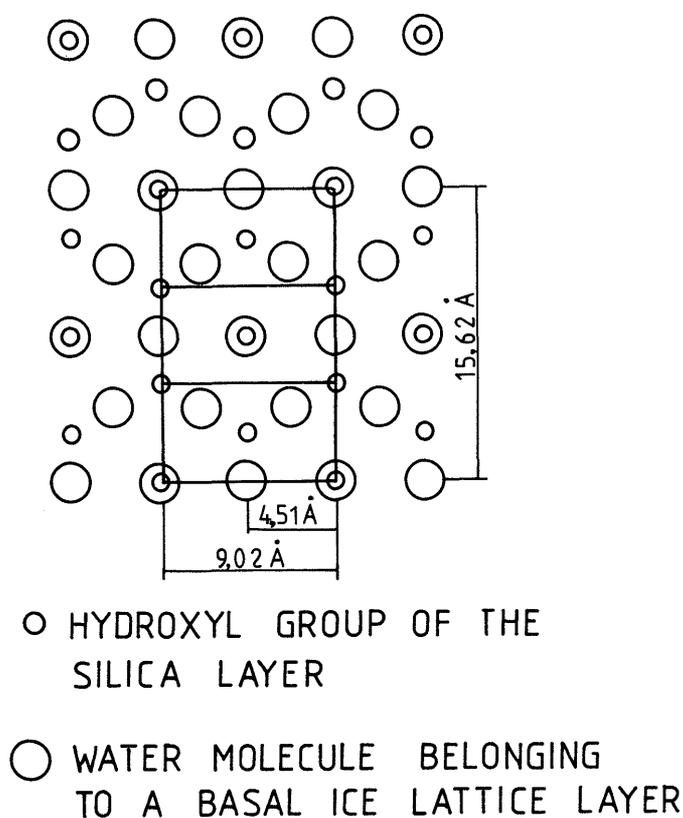


Fig 5. Basal ice lattice molecules superimposed on the hydroxyls of the (001) plane (After Forslind)

The coordination implies that there are only four hydrogen bonds formed between three unit cells and the associated water lattice. The charge of the crystallite will therefore attain a mean value of two-thirds of a charge unit per unit crystal cell, which actually equals the mean value of a large number of literature-reported experimental cation exchange capacity values.

Heating liberates intercrystalline water, which is practically removed at 105°C eventually including some surface dehydroxylation but leaving part of the surface hydroxyls intact even in the temperature interval 150-200°C according to Forslind. The liberated hydroxyls form free water that escapes, and silica, as well as hydrated silica, is then also free to move. Charge equilibrium requires uptake or intralattice migration of other available ion species such as Al, and if K is present these ions sink into the silica layer as the lattice expands at rising temperature whereby tensions and lattice distor-

tions are set up that prevent surface rehydroxylation. For other common cations, dehydration of montmorillonite by heating below 200°C is, as a rule, reversible.

A third study, conducted by Torbjörn Carlsson, Dept Nuclear Chemistry, Royal Institute of Technology, Stockholm, involved autoclave tests of MX-80 bentonite with a bulk density of 1.8 t/m³ at 8 MPa water pressure and 100°C, using distilled water. It lasted for 47 days and comprised X-ray diffraction analyses, scanning microscopy and determination of the spin-spin coherence time T_2 using nuclear magnetic resonance technique. The diffractograms of the heated and non-treated materials turned out to be identical, the appearance of micrographs of the two materials was similar, and the T_2 values were exactly the same. The latter parameter is a rather sensitive measure of the hydration power of minerals and the study thus indicates that the physical state of interlayer water molecules was not affected by the heating.

2.4 Precipitation of silica compounds

The liberated silica precipitates in the form of quartz or silica compounds in the interstitial pore space. Considering the pack-type microstructural features of very dense smectite clay, such as natural bentonites (Fig 6), the liberated silicons will diffuse in the thin interlamellar space towards the edges of the dense aggregates. Assuming the E/F-E/H/W transition to be a true mechanism, the release of Si is largely that of hydroxyl liberation, for which the activation energy is that of the hydrogen bond, i.e. about 3-8 kcal/mole. This would be in agreement with the experimental values derived by Howard & Roy, in which case the outward, interlamellar migration of water associated with lattice collapse in the c-direction through heating, would probably be the main Si-transporting mechanism.

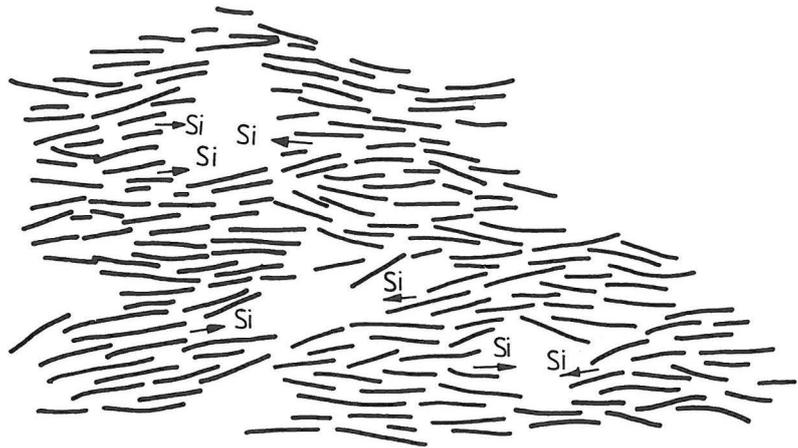


Fig 6. Schematic smectite flake arrangement in natural bentonites. Arrows indicate assumed Si migration at heating

Whatever the detailed Si-migration pattern may be, Si or more generally Si(OH)_n , should be enriched at the edge of the packs of smectite flakes. The Kinnekulle bentonite, which is known to have been heated to $110\text{-}150^\circ\text{C}$ for several hundred years (3) shows evidence of quartz located at particle edges (11) and a close examination of the particle edges in this bentonite bed actually indicates the presence of local "nodules" which may represent precipitated silica compounds (Fig 7).

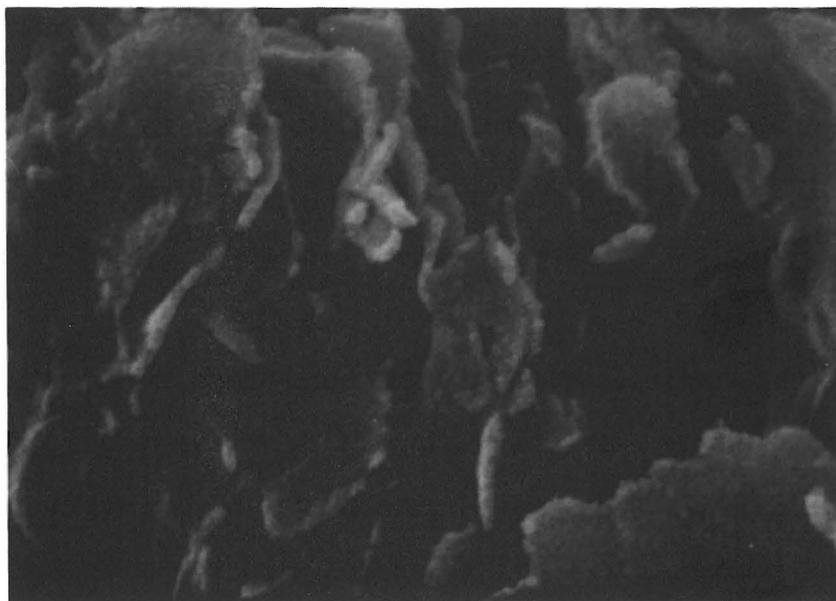


Fig 7. Scanning micrograph of Kinnekulle bentonite. The arrow points at assumed precipitate at particle edges
Magnification 30 000 x.

2.5 Conclusions with respect to possible chemical alterations in the BMT

The literature-derived information of heat-generated alteration of smectite referred to in this chapter suggests that no major changes can be expected in the about 1 year long test. However, possible Si-release and precipitation would be recognizable by applying chemical analyses and electron microscopy with very high resolution power, and X-ray diffraction tests should reveal the replacement of Si by Al. Although physical tests, like determination of the hydraulic conductivity and the swelling pressure, are too insensitive to indicate a change of 100 % smectite to say 95 % purity, they would still serve as a valuable check of the overall integrity and preservation of the original properties of the highly compacted bentonite. A test program was therefore worked out which comprised all these tests.

2.6 Test program

2.6.1 Possible reactions

The possible chemical changes that may have taken place in the dense bentonite in heater hole no 1 are:

- * Charge change through replacement of Si by Al in the montmorillonite tetrahedrons
- * Migration and precipitation of released Si
- * Formation of mixed layer (I/S) minerals by K-fixation
- * Release and migration of Al from the locally corroded heater surface

The heat effects were expected to be most obvious close to the heater and samples were therefore taken in the radial direction at the distances 2, 5, 8 and 17 cm from the heater surface at mid-height of the heater. They had been exposed to the approximate temperatures 125°C, 105°C, 93°C and 72°C, respectively, for about one year (cf. Fig 8).

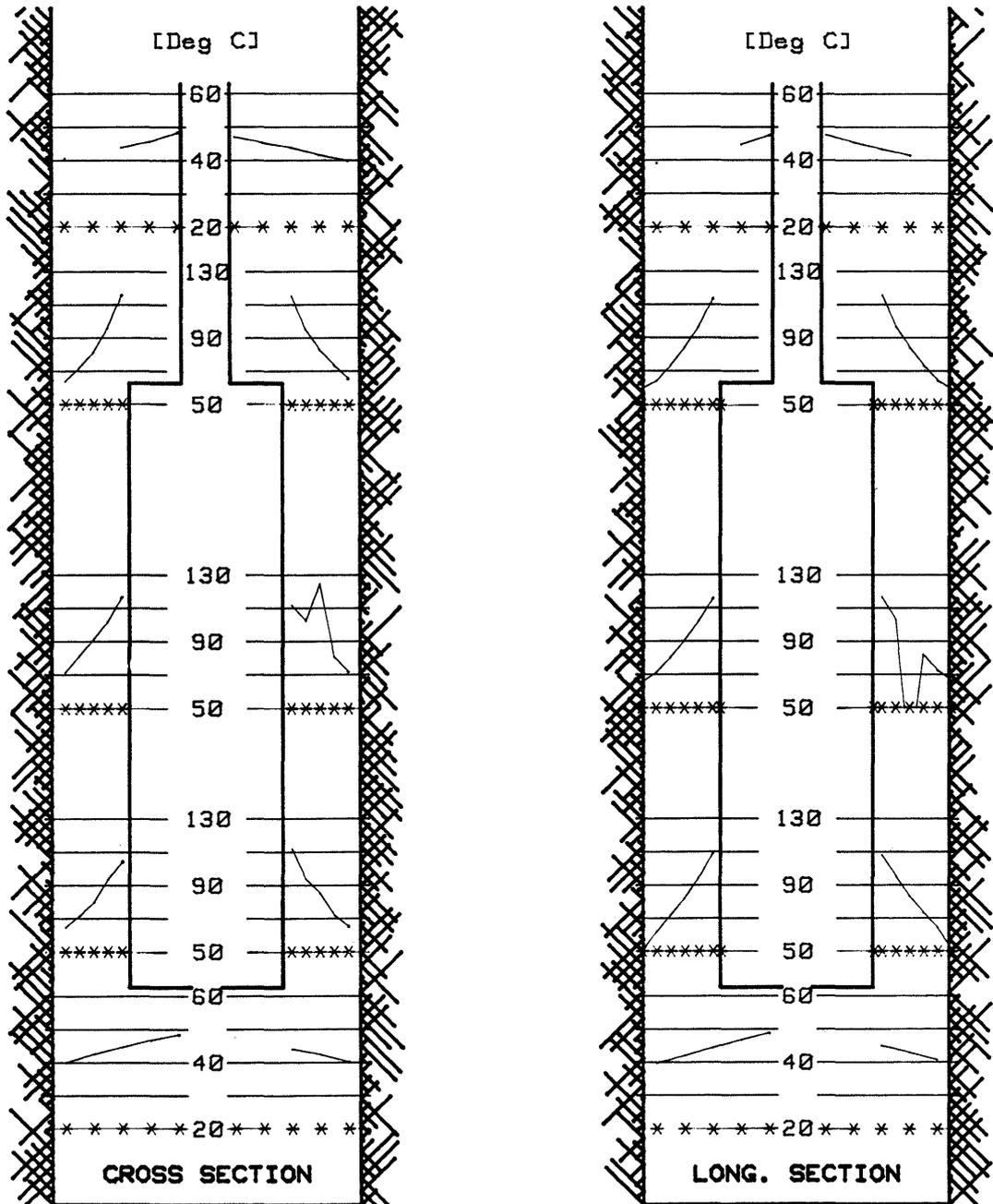


Fig 8. Temperature distribution in heater hole no 1 at the termination of the 1400 W test 0.9 years after the test start

The individual test procedures are described in the subsequent text, the test program being summarized in Table 3 at the end of this chapter.

2.6.2 Charge change

Release of Si is expected to be associated with Al replacing Si in the tetrahedrons of montmorillonite. The process should be recognizable by applying the so-called Greene-Kelley test (11), which involves saturation with Li. After heating to 220-300°C and ethylene glycol treatment (EG), montmorillonite does not show reversible swelling, while beidellite does.

The reason for the different behavior of the two smectite representatives when this test procedure is applied, is supposed to be the following. In both clay types about 1/3 of the possible octahedral cation sites are assumed to be non-occupied. The Greene-Kelley test implies that Li-ions are sufficiently small to enter octahedral positions of the structure and to remain there after heating. This produces neutralization and non-expansion of the montmorillonite, in which tetrahedral substitution is absent. In beidellite, in which such substitution is frequent, Li does not occupy octahedral sites and this mineral retains its swelling properties on EG treatment. The accuracy of the test is said to be better than 20 %. It should be noted that the X-ray diffraction patterns of non-treated EG-saturated montmorillonite and beidellite are identical.

The preparation of specimens for XRD tests involved converting to the Li stage, filtration of a part of the suspensions and drying at 300°C overnight, after which the specimens were submerged in ethylene glycol at 60-70°C for 12 hours. Reference samples that had not been heated, were dispersed in distilled water by ultrasonic treatment.

The analyses were conducted by the Swedish Geological Survey (SGU), Uppsala, Sweden.

2.6.3 Si release and migration

Release and migration of Si down the thermal gradient would be detectable by chemical analyses. Two methods were applied: A) Treatment with 2 M Na_2CO_3 at 90°C for 4 hours and B) Treatment with 0.2 M NaOH at 60°C for 2 hours, and subsequent neutralization with 0.5 M H_2SO_4 . The determination of Si is made by use of ammonium molybdate.

The accuracy of the chemical analysis, the concentration being expressed in terms of percent of "amorphous SiO_2 ", is better than 0.1 percent units. The analyses were made by SGU.

2.6.4 Si precipitation

A safe identification of precipitated Si compounds requires high resolution electron microscopy. Two techniques have been applied in the present study, the main one being transmission electron microscopy (TEM) using a JEOL 100 cx microscope with a theoretical line resolution of 3 Å, the other being scanning microscopy (SEM) for which a Hitachi S-800 field emission microscope with an estimated resolution power of about 15 Å was utilized.

In addition to the samples from heater hole no 1, core samples from the aforementioned Kinnekulle bentonite were also investigated to double-check the earlier finding of Si edge-coatings.

The TEM study was made by professor Anders Andersson, Department of Histology, University of Lund, Sweden, and M. Olivier Touret, Centre de Sedimentologie et de Geochimie de la Surface, Strasbourg, France, while the SEM work was conducted by Dr Ethan Goldberg, Gentrionix

Laboratories, Inc., Maryland, USA. All three investigations were made using specimens prepared in the author's laboratory or according to his specifications. The major preparation steps were as follows:

TEM One series of tests was made of dispersed material for identification of precipitated matter at smectite flake edges. The preparation involved very slight mechanical agitation of samples submerged in distilled water. A drop of the low concentration clay suspension was transferred to the grid of the specimen holder which was inserted in the microscope after slow drying in room atmosphere.

A second series, intended for identification of precipitated matter as well as possible indications of restricted swelling due to such matter, was conducted using samples embedded in low viscous epoxy resin (Epo-tek, MPE Microtech AB, Sweden) and Spurr (Balzers Union, France).

SEM This study was made for identification of precipitated matter at smectite flake edges. The preparation involved gradual shaving to form sticks of the clay approximately 10 mm long x 2.5 diameter. The specimens were then frozen in liquid freon and fractured with a chilled knife. Immediately thereafter they were placed in liquid nitrogen and then into "vacuum" to remove any internal (frozen) water. After sublimation, the stick was mounted to a SEM specimen stub with the fracture face upwards. Several tape pulls were performed to remove attached debris and the specimens were then evaporatively coated with gold.

2.6.5 Mixed-layer formation

XRD was applied to check whether mixed-layer intercalations had been formed by possible charge change and uptake of K from the porewater in the heater hole. Also, K-treated samples were tested to see whether the ion exchange properties had been altered. The latter samples were prepared by repeated treatment of the dispersed clay with 1 mol KCl solution. To remove excess KCl washing with distilled water was made before mounting the specimens for XRD. The investigations were made by SGU.

2.6.6 Al release and migration

Release of Al from the corroded surface of the heater should be recognizable through spectrometer element analyses. Such tests were conducted after leaching the samples with 1 M Na-tartrate solution at 60°C overnight. After heating, the samples were centrifuged, decanted and analysed. The study was conducted by SGU.

2.6.7 Determination of physical properties

The swelling ability and hydraulic conductivity of a sample extracted close to the heater were determined by applying KBS standard techniques. The 32 mm high sample was trimmed to fit in the \emptyset 50 mm space of a swelling pressure oedometer (Fig 9). Synthetic groundwater with an electrolyte composition close to that of natural Forsmark water (chlorides with 2530 ppm Na, 950 ppm Ca, 22 ppm Mg and 5 ppm K, total salt content about 10 ‰) was added to the sample to make it expand to a density of 2.05 t/m³ and become homogeneous. After the determination of the swelling pressure, percolation with synthetic Forsmark water was made, and the sample was then allowed to swell to a bulk density of 1.95 t/m³. The swelling pressure measurement and the percolation test were repeated and the sample again allowed to swell,

the bulk density now being 1.80 t/m^3 . The same physical tests were repeated also for this expanded version.

All the physical tests were conducted at the SGAB laboratory in Lund.

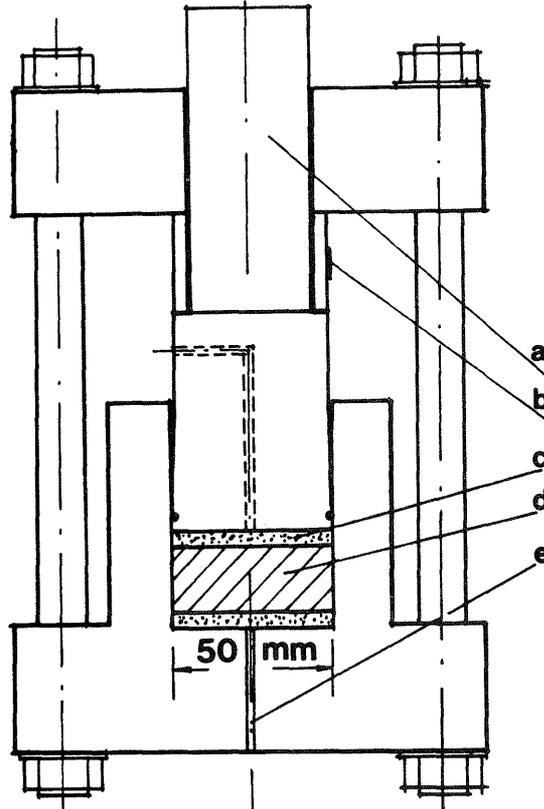


Fig 9. The swelling pressure oedometer. a) is piston loaded to make the ring b) stress-free, which means that the swelling pressure is balanced by the external load. c) is filter and d) clay sample. e) is inlet for percolation of water or gas

2.7 Test results

2.7.1 Charge change

The Greene-Kelley tests gave no consistent results. Thus, beidellite reaction was noticed for all the samples in a first test series. The heating in the oven was found to have been insufficient, however, and a very careful second series was therefore conducted. It showed no beidellitization for the samples located close to the heater and close to the rock, but reactions for the other two heater hole samples and the non-heated reference samples (Fig 10). It was concluded that the applied technique was inadequate to bring the specimens in the intended, defined state with respect to Li saturation and heat-

ing.¹⁾ Since practically all the other tests indicated that charge changes through Al replacing Si had not taken place to a measurable degree, further Greene-Kelley testing was not made.

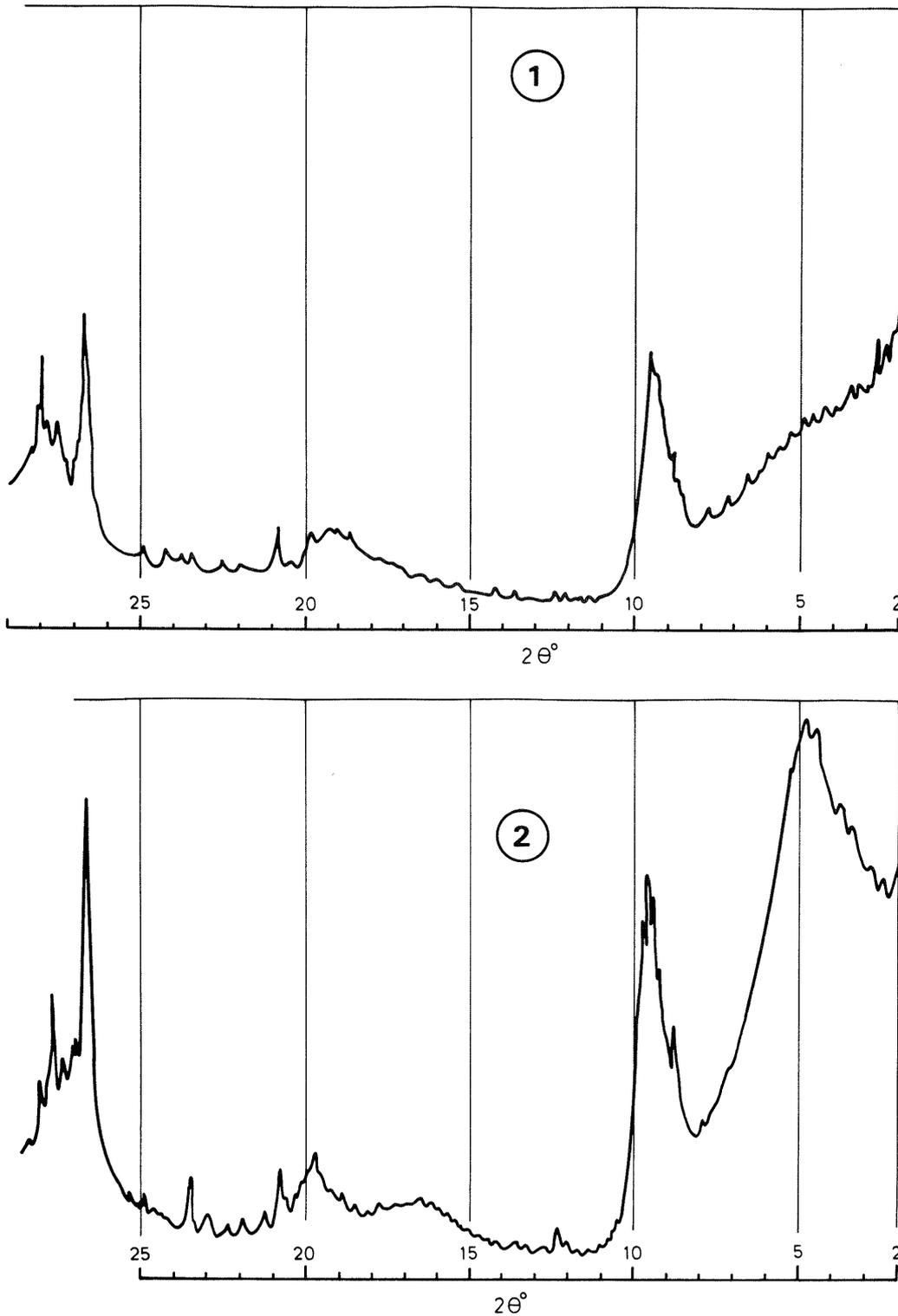


Fig 10. XRD diagrams of the EG-treated samples no 1 (upper diagram) and no 2, respectively

1) Pers comm Dr Ann Marie Brusewitz (SGU)

2.7.2 Si release and migration

The results of the chemical analyses for determination of Si in the form of amorphous silica are reported in Table 3.

Table 3. Distribution of amorphous SiO₂ in the heated bentonite

Sample no	Distance from heater, cm	Amorphous SiO ₂ , weight percent		Note
		Method A	Method B	
1	2	0.49	1.07	Heater samples
2	5	0.33	1.13	
3	8	0.52	1.17	
6	17	0.61	0.99	
7	-	-	2.10	Reference, "non-heated" samples
8	-	-	1.02	
9	-	-	1.87	

The analyses, which gave a systematic difference between results obtained by use of Methods A and B, show that only insignificant amounts of SiO₂ may have been released and that the sample close to the heater did not differ from the less strongly heated material in this respect. Separate tests were made using non-heated granite specimens with a content of crystalline SiO₂ of 61 %, and this material gave off about 0.6 % SiO₂. The fact that also crystalline silica was leached indicated that all the recorded values for bentonite were actually close to or below the detection limit.¹⁾

2.7.3 Si precipitation

The scanning microscopy yielded high quality micrographs with magnifications ranging between 20 000 and 100 000 times of specimens from the heater/bentonite interface and the rock/bentonite interface.

1) Pers comm Dr Ann Marie Brusewitz, SGU

The freezing process in the preparation caused the interlaminar water to migrate to larger interstitial pores where freezing took place. The concomittant contraction of the domains gives the image of rather thick particles in these micrographs (Figs 11-13), but they are actually condensed packs of 10 Å smectite flakes. The gold sputtering yielded a uniform, very finely nodular cover of all particles, which, however, is distinguishable from those larger nodules at the domain edges, that were previously observed in micrographs of the Kinnekulle bentonite and that were interpreted as precipitated silica compounds (cf Fig 7). Larger nodules indicating Si precipitation were not observed in the presently investigated samples, with the exception of a few pictures of specimens extracted from the heater/bentonite interface (Fig 14). They showed a small number of such features which called for additional electron microscopical investigations with even better resolution. For this purpose, transmission electron microscopy (TEM) was applied.

The application of preparation techniques using organic substances, such as the various resins that are usually used in soil preparation for electron microscopy (12), is known to produce swelling through interlamellar uptake of organic molecules. This yields TEM images that are expected to be fairly representative of the actual microstructure but different from the SEM micrographs of freeze-dried specimens in which the originally expanded aggregates are shown in a collapsed state.

Ultramicrotome-cut specimens turned out to give largely structure-free pictures, indicating almost isotropic, fully expanded smectite, with no restraint due to cementation of samples from the heater/bentonite interface as well as from the rock/bentonite interface (Fig 15). The expected appearance if edge-located, cementing Si-compounds had been present, is that of Fig 16, and this

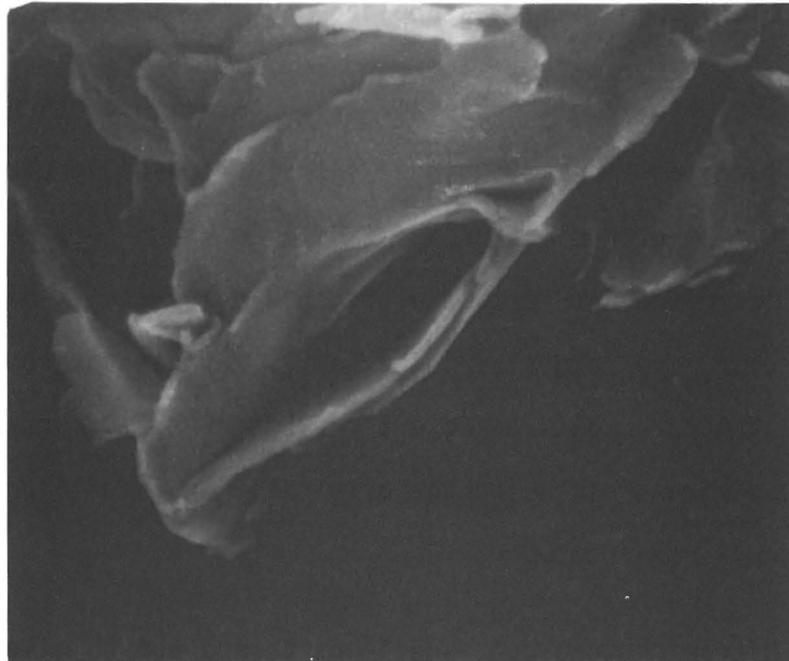
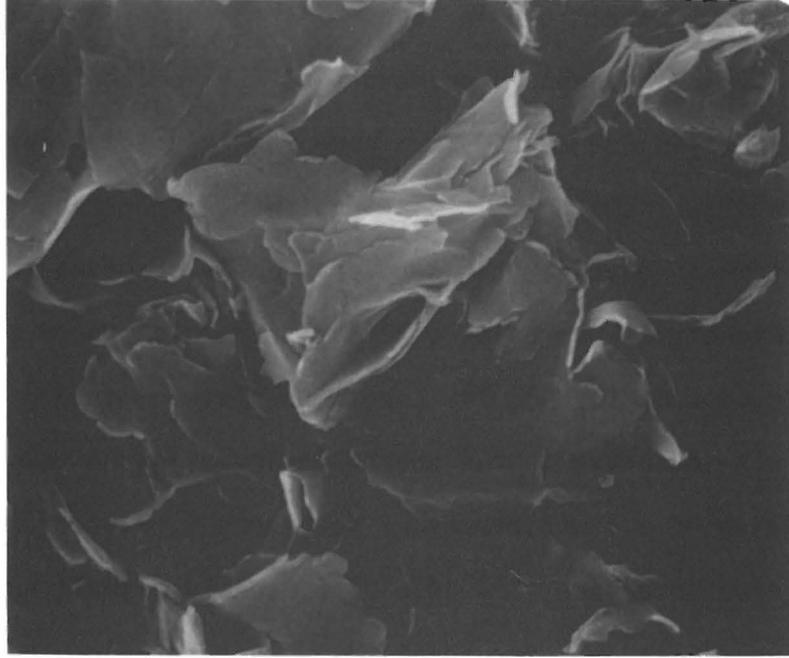


Fig 11. Scanning micrographs of samples that were located at the rock/bentonite interface. Magnifications 20 000x (upper micrograph), and 50 000x, respectively

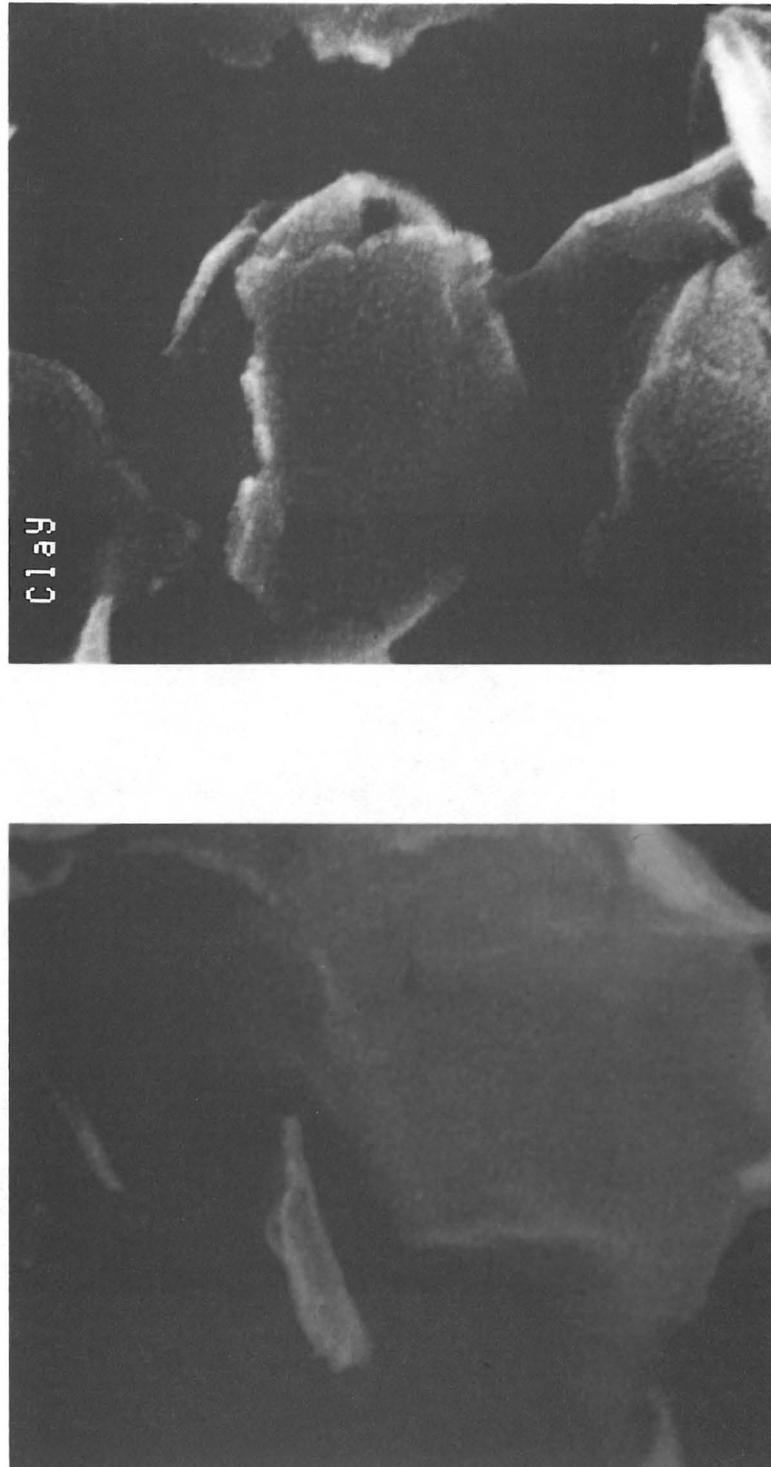


Fig 12. Scanning micrographs of samples that were located at the rock/bentonite interface. Magnification 100 000x

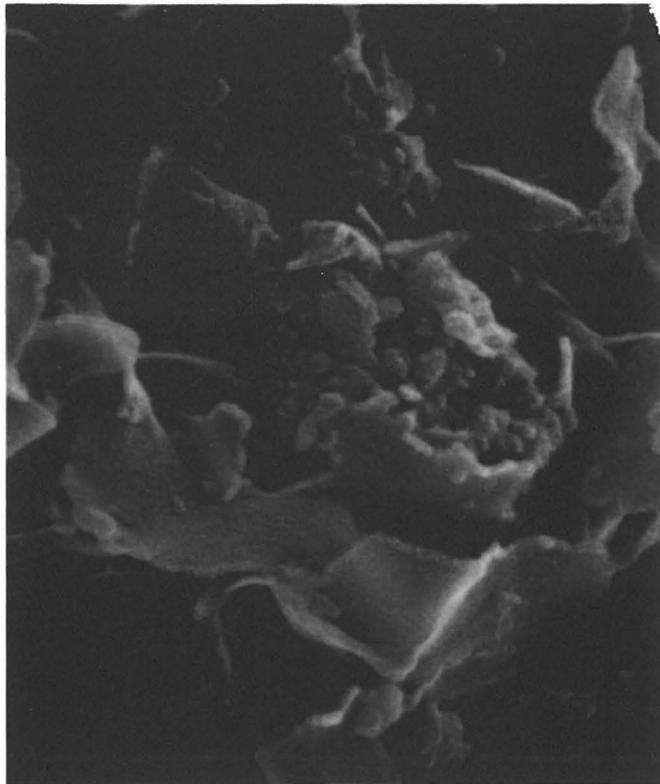


Fig 13. Scanning micrograph of sample located at the heater/bentonite interface. Magnification 50 000x

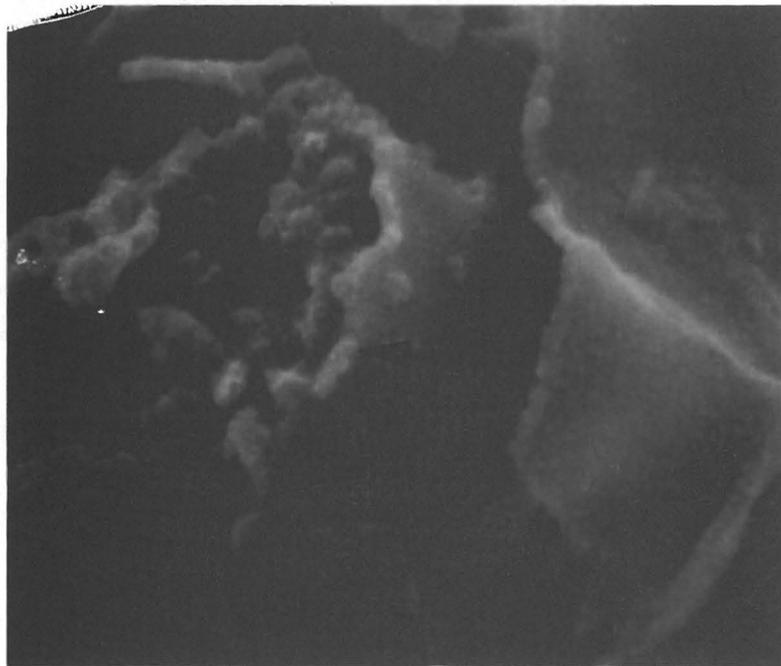
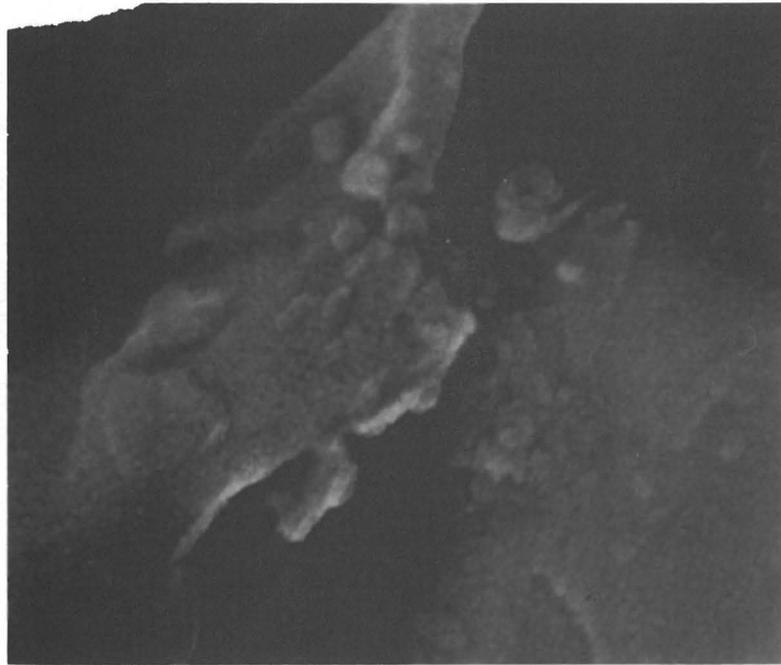


Fig 14. Scanning micrograph of sample located at the heater/bentonite interface. The lower picture shows possible precipitates (arrowed). Magnification 100 000x

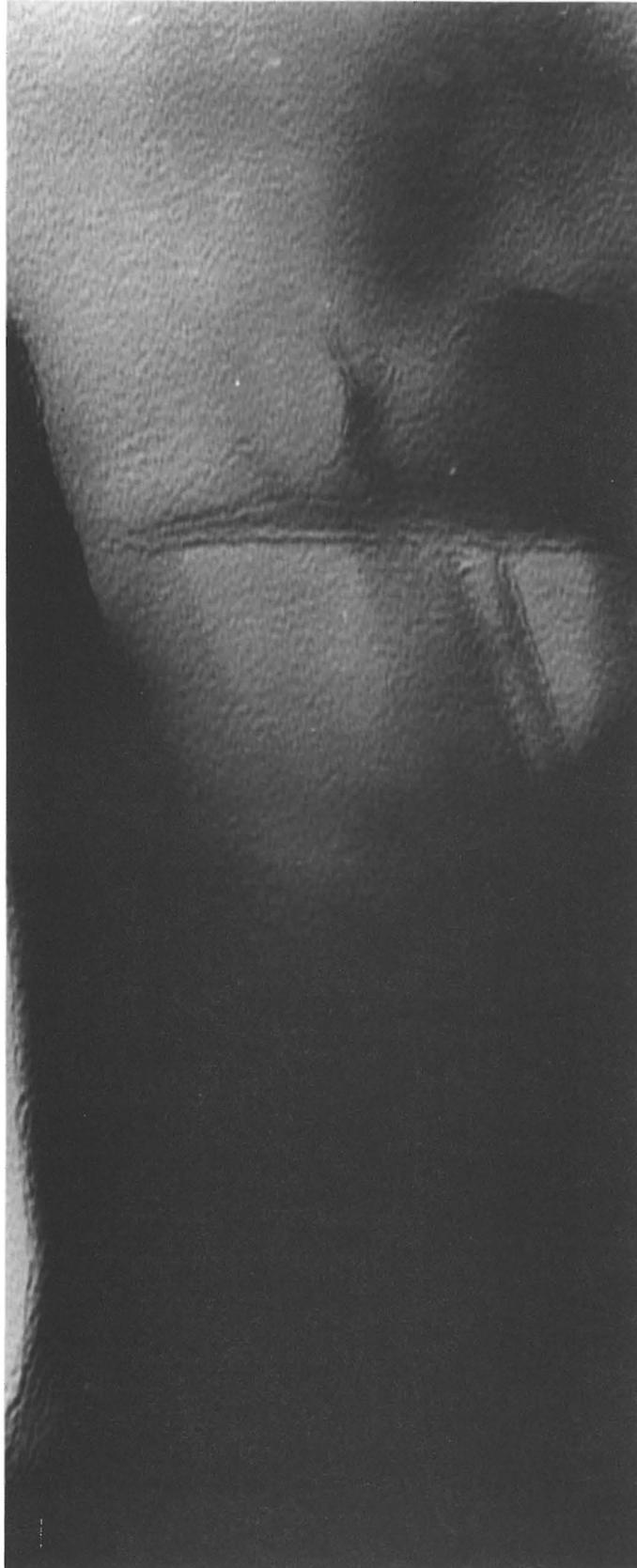


Fig 15. TEM micrograph of ultrathin section of sample from the heater/bentonite interface. Magnification 50 000x



Fig 16. Expected appearance of smectite flake domains that are silica-cemented at the edges. Interlamellar absorption of organic molecules produces swelling but not at the edges

is more or less what we see from TEM-micrographs of the Kinnekulle bentonite (Fig 17). It should be noticed that although the "point-welding" caused by the precipitation of edge-cementing agents reduces the swelling power, it is only partly eliminated.

An additional attempt was made to identify minute edge-precipitates by applying TEM to dispersed clay specimens. Since these had a thickness of only a few tens of Å units, the high resolution power of the microscope could be fully utilized. About 20, randomly chosen micrographs were analyzed and none of them showed any sign of precipitates at the periphery of the smectite flakes, neither at the heater/bentonite interface nor at the rock/bentonite region (Figs 18-20). The expected appearance of precipitates would be dark (electron-absorbing) objects adhering to the periphery of the condensed groups of smectite flakes. As demonstrated by the micrographs, the high resolution power made it possible to identify the individual laminae of suitably oriented domains while there was no sign of precipitates. A

corresponding study of dispersed, smectite-rich Kinnekulle material showed approximately the same pattern (Fig 21) with the exception that a number of very small, electron-dense objects appeared near the edges of tapered domains in some pictures. It was not clear whether these are precipitations or simply fragments of larger minerals produced in the course of the dispersion and additional tests were therefore made of ultrathin sections of epoxy-embedded Kinnekulle material. The preparation of these samples involved even less mechanical disturbance and since the dark objects appeared also in these micrographs, they may represent true precipitations (Fig 22).



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Fig 17. TEM micrograph of ultrathin section of smectite-rich Kinnekulle bentonite



Fig 18. TEM micrograph of dispersed heater hole bentonite. Notice the uniform character of the thin domain of smectite flakes
Magnification 235 000x

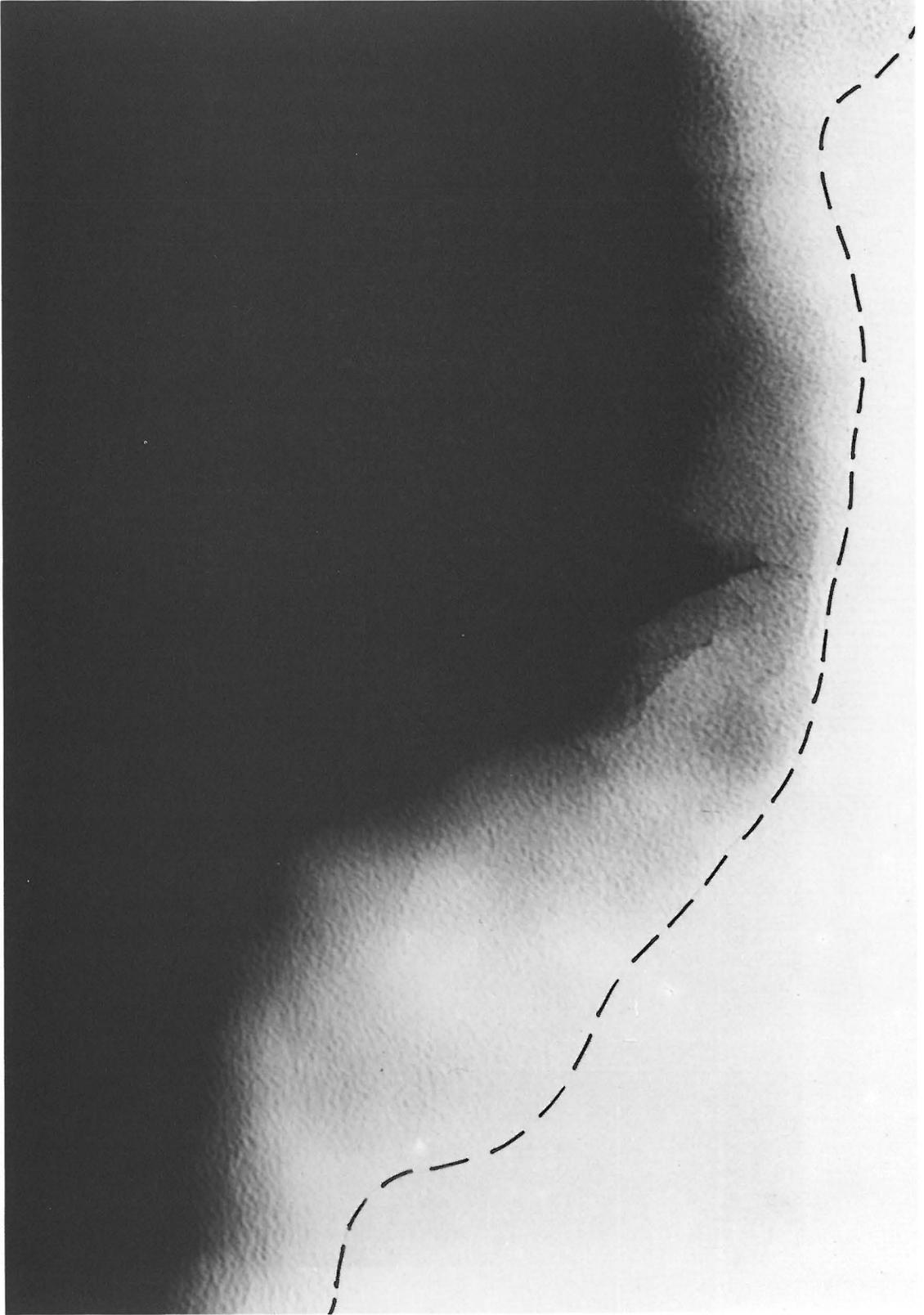


Fig 19. TEM micrograph of dispersed heater hole bentonite. The electron beam passed through a few parallel smectite flakes oriented in the plane of the micrograph, the outer contour being the broken curve. Notice the uniform greyish appearance with no indication of precipitations. Magnification 235 000x



Fig 20. TEM micrograph of dispersed heater hole bentonite. Overlapping thin domains of uniform character. Magnification 305 000x



Fig 21. TEM micrograph of dispersed smectite-rich Kinnekulle bentonite of a supposedly non-cemented kind. Magnification 590 000x

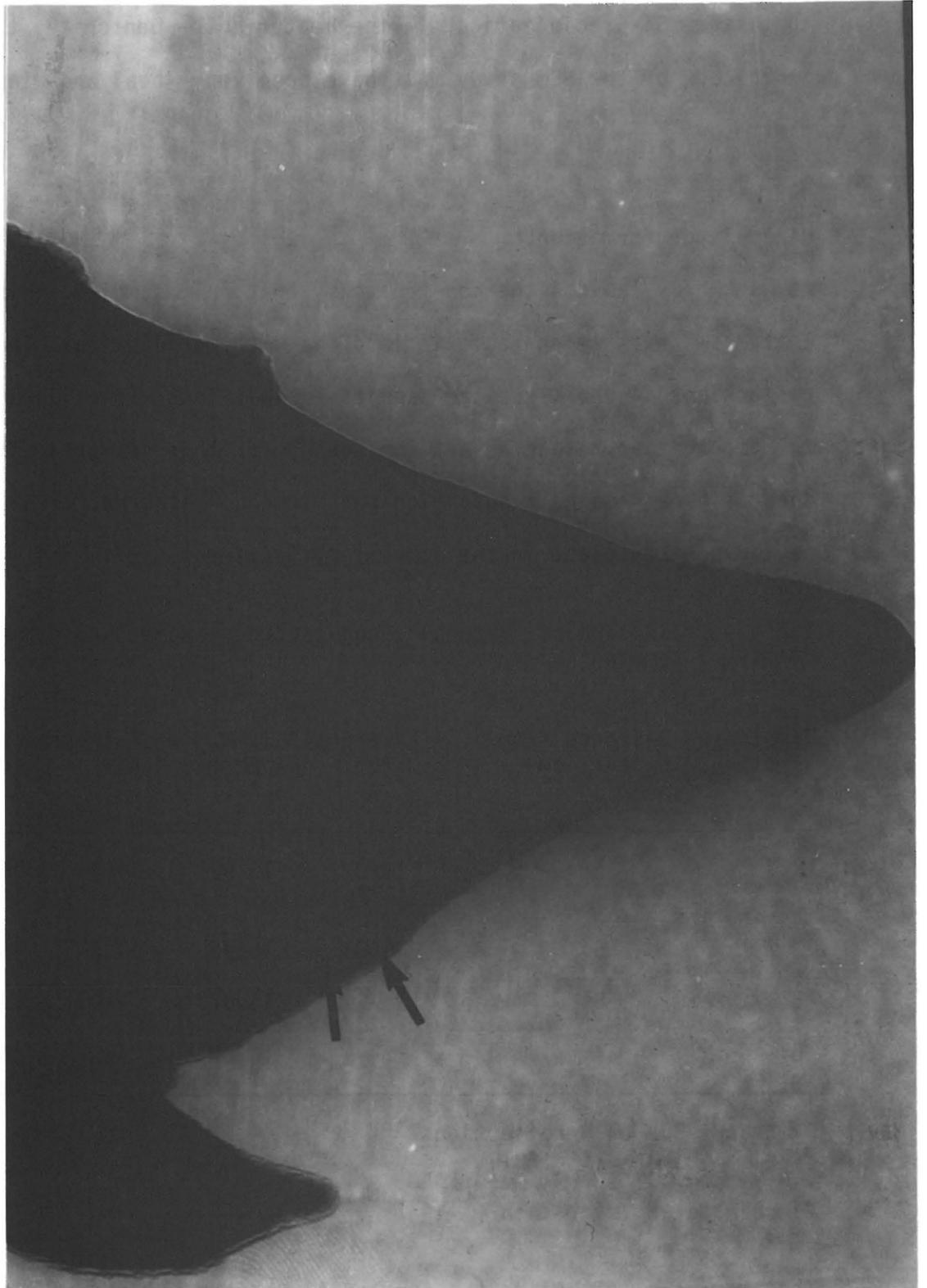


Fig 22. TEM micrograph of ultrathin section of epoxy-embedded smectite-rich Kinnekulle bentonite. The electron beam passed through a pack of aligned smectite flakes that were oriented in the plane of the micrograph. Notice the discrete electron-dense objects which may represent Si precipitations (arrowed). Magnification 380 000x

The general conclusion of the various microscopical investigations is that the Si-precipitation in the heater hole bentonite that was indicated by the SEM study must have been very local and insignificant.

2.7.4 Mixed-layer formation

As shown by Table 4 the non-treated, "natural" samples exhibit XRD patterns that are characteristic of Na montmorillonite. The basal reflections of the K-treated samples exhibit less expansion after EG-treatment than expected but the reason for this is not altogether clear. There remains a suspicion that some collapse due to K-fixation may have taken place in the laboratory treatment.

Table 4. XRD data of "natural", non-treated samples and K-saturated samples

Sample no	Distance from heater, cm	"Natural"		K-treated	
		Air Å	EG ¹⁾ Å	Air Å	EG Å
1	2	13.0	16.92	11.9	15.5 14.7
2	5	13.0	16.95	11.5	16.7
3	8	-	16.97	-	14.5
6	17	-	16.85	-	16.0

1) Mean value of 8 reflections

2.7.5 Al release and migration

The analysis of Al in the form of Al_2O_3 showed no concentration profile and no enrichment of Al close to the corroded heater surface (cf. Table 5). This suggests that Al-compounds formed by the chemical interaction of bentonite and metallic aluminum remained close to the clay/heater interface.

Table 5. Distribution of non-lattice Al_2O_3 in the heated bentonite

Sample no	Distance from heater,	Al_2O_3 weight percent	Note
1	2	0.060	Heater samples
2	5	0.051	
3	8	0.060	
6	17	0.053	

7	-	0.066	Reference, "non-heated" samples
8	-	0.083	
9	-	0.085	

2.7.6 Physical properties

Swelling and swelling pressures

The sample that was extracted from the close vicinity of the heater and transferred to a swelling pressure oedometer for equilibration with Forsmark water with subsequent, controlled expansion gave the swelling pressures shown in Table 6. These values are also plotted in the diagram in Fig 23 in which earlier data from the author's swelling pressure determinations using porewaters with a salinity of 30 %- 100 % of sea water are collected. It is concluded that the recorded swelling pressures are at least as high as those obtained for non-heated MX-80 bentonite saturated with salt water. Even relatively slight cementation would be expected to have produced significantly lower pressures.

Table 6. Recorded swelling pressures of bentonite sample from the close vicinity of the heater

Bulk density ¹⁾ ρ t/m ³	Swelling pressure MPa
2.05	50
1.95	4.6
1.80	0.5

1) Water saturated

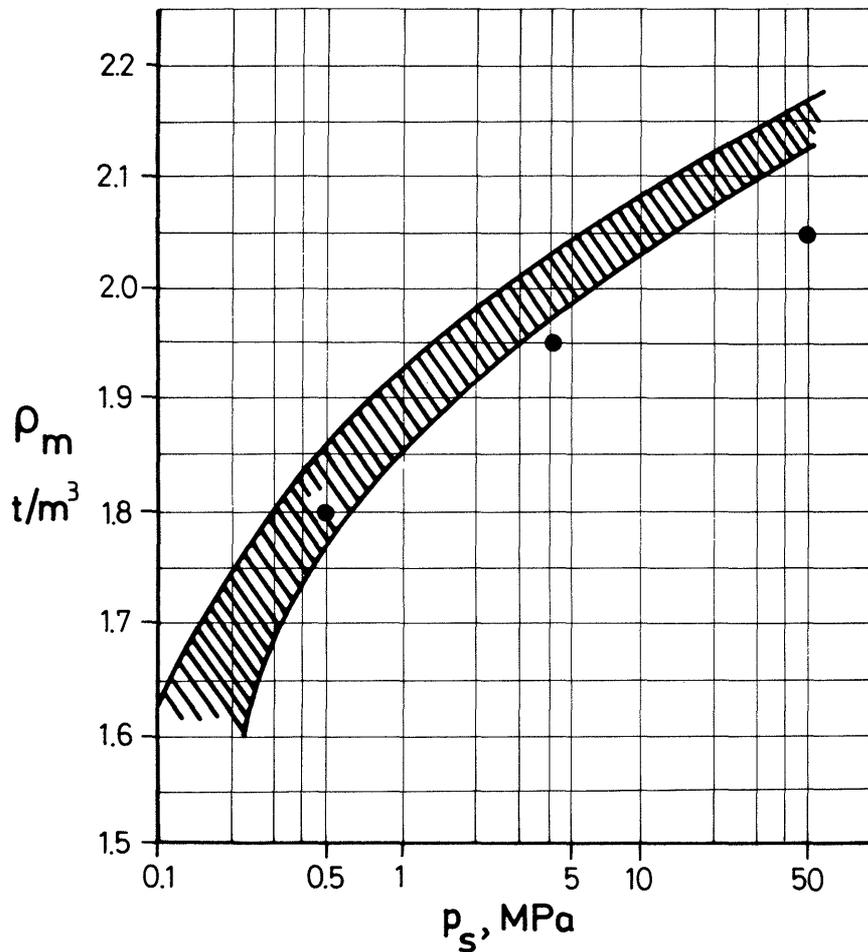


Fig 23. Swelling pressure (p_s) versus bulk density of MX-80 bentonite with high porewater salinity. Dots represent the samples from the heater hole

Hydraulic conductivity

At each expansion stage, which involved at least two weeks time for swelling and homogenization under drained conditions, the hydraulic conductivity was measured by applying a controlled water pressure using nitrogen as a driving force. The evaluated hydraulic conductivity k is given in Table 7 as a function of the hydraulic gradient and the time after onset of each test. The values are plotted in the diagram in Fig 24 in which earlier data from percolation tests of MX-80 clay with low salinities ("Allard water") have been collected. It is clear that the permeability is in the same order of magnitude as that of non-heated bentonite, although the more saline Forsmark water was expected to yield stronger flocculation and significantly higher k -values especially for the most expanded stage. The conclusion is that no heat-induced microstructural changes or influence on the water-adsorbing properties of the heater hole bentonite had taken place. It should be added here that the recorded k -values for the highest bulk density are probably too high due to non-complete homogenization.

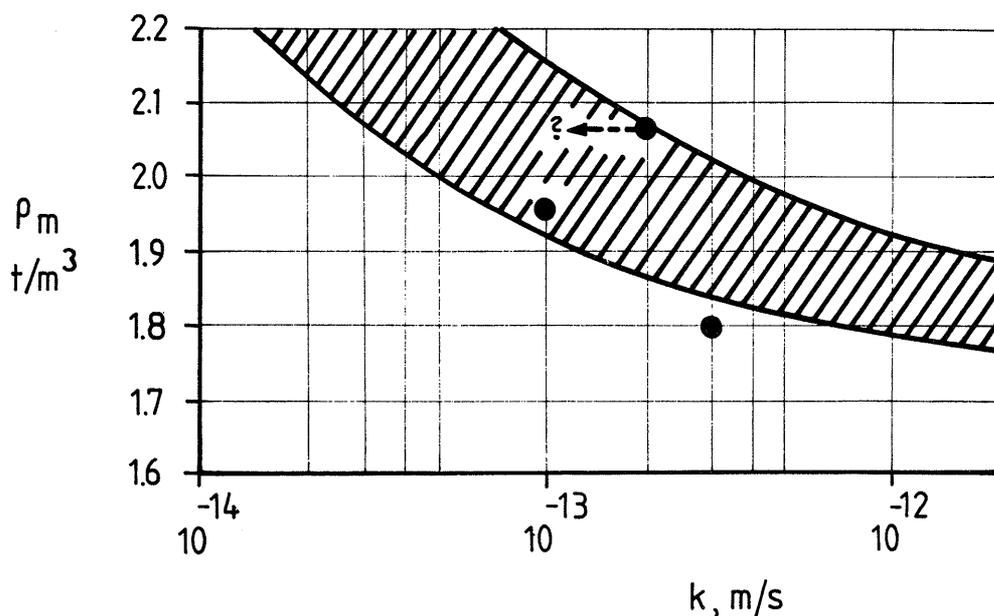


Fig 24. Hydraulic conductivity (k) versus bulk density of MX-80 bentonite. The band represents low salinities while the three dots refer to the heater bentonite

Table 7. The hydraulic conductivity k of bentonite sample from the close vicinity of the heater

Bulk density, t/m^3	Time after onset of test or change of gradient, hours	Hydraulic gradient	k , m/s
2.05	17	2000	$5 \cdot 10^{-13}$
	28		$4 \cdot 10^{-13}$
	100		$2 \cdot 10^{-13}$

1.95	22	400	10^{-13}
	45		10^{-13}
	69		10^{-13}
	141		10^{-13}

1.80	24	4000	$2 \cdot 10^{-13}$
	48		$3 \cdot 10^{-13}$

2.8 Conclusions

2.8.1 General

The results of all the tests concerning the possible effect on the chemical integrity of water saturated bentonite by heating to about 125°C for about one year are summarized in Table 8.

It is concluded that the influence of heating was insignificant and that the important physical properties, swelling and permeability, were not altered. The key reaction, i.e. that of Al replacing Si in the tetrahedrons, may have taken place to a very limited extent, which supports the idea that this process is very slow at temperatures of about 100°C . Actually, the results do not invalidate the alternative concept, namely that of a critical temperature of approximately 100°C . The matter cannot be settled without further detailed studies.

Table 8. Summary of chemical analyses and physical tests. X denotes observed effects, the parenthesis indicating that there is no statistical certification

Test	Effect		Nature
	At heater	At rock	
Charge change (Greene-Kelley)	-	-	
Si release and migration	-	-	
Si precipitation	(x)	-	Possible tendency of precipitation at domain edges
Mixed-layer formation	(x)	-	Slightly reduced swelling ability
Al release and migration	-	-	

Swelling properties	-	-	
Hydraulic conductivity	-	-	

3 PHYSICAL STABILITY

3.1 General aspects

The main question with respect to the physical stability of buffer materials is that of erosion. This effect has been considered in the KBS 3 concept for which laboratory tests as well as some preliminary theoretical studies were conducted a few years ago (13, 14). Some of the findings are of interest in connection with the BMT study and they are therefore summarized here.

3.1.1 **Highly compacted bentonite**

One beneficial effect of highly compacted Na bentonite contacting rock is that it tends to enter fractures by which they get sealed off. This has been demonstrated in several series of laboratory tests and was very obvious also in the BMT heater holes. The laboratory investigation formed the basis of a simple physical model for the penetration of expanding bentonite into rock fractures, the basic mechanism being that the penetrating clay expands by absorbing water which is sucked through this same clay (Fig 25). The model implies that the rate of penetration is governed by the hydraulic gradient, caused by the suction power, and the permeability. The first-mentioned drops due to the reduced density of the expanding clay, while its permeability increases. The resulting, successively altered physical state of the penetrating clay is illustrated in Fig 26.

In practice, there is a retarding effect on the penetration rate by the friction and adhesion forces that are developed at the fracture walls so a more complete flow model, at least for fracture apertures of less than a few millimeters, must also contain a "Poiseuille" term. However, the general character of a clay film with a dense "root zone" and a soft front would be valid also for such an improved model version. A mathematical analogy of general form that has been

found to agree relatively well with laboratory-determined clay penetration rates has the following appearance:

$$x = A d^2 \log (t+1) \quad (1)$$

where x = displacement of the film front

d = aperture

t = time after the onset of flow

A = constant depending on temperature, clay type and fracture surface characteristics

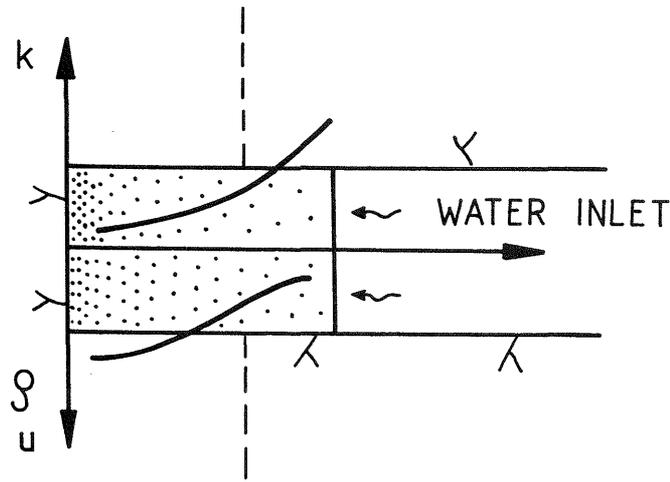


Fig 25. Schematic picture of "free expansion", i.e. swelling of a dense clay body resulting in a drop in density ρ towards the advancing front. k - and u - changes are discussed in the text

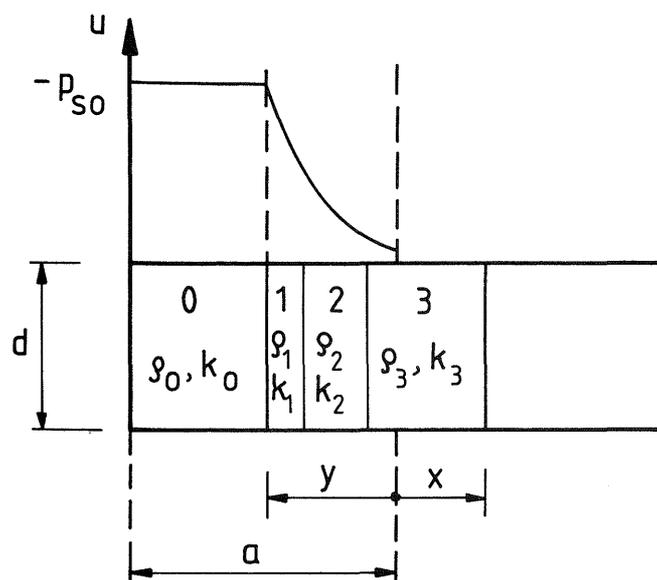


Fig 26. Model for stepwise calculation of the expansion process in a clay zone with the original density ρ_0 , negative pore pressure u_0 , and permeability k_0 . x represents the expansion of the zone, while y represents a softened part

According to laboratory studies as well as to the theoretical, physical model, the film front has the form of a very soft clay gel and this was also confirmed at the excavation of the BMT heater holes. The possible erosion of the film front is therefore a matter that must be considered.

Dispersion of expansive clays is known to be largely affected by the so-called sodium adsorption ratio (SAR) of the pore fluid which is defined as $SAR = Na / (\sqrt{\frac{Ca+Mg}{2}})$, where the chemical symbols refer to the concentrations of the designated cations in milliequivalents per liter. The dispersibility of such clays is also related to the $ESP = (Na/CEC) \times 100$ where Na stands for the sodium concentration in the exchange complex and CEC is the total exchange capacity, both expressed in milliequivalents per 100 g dry clay. Very low SAR and ESP values (5) are typical of non-dispersive clays, while higher values than 15 characteristically point to strong slaking. Experience from pinhole tests shows that clays tested with sodium ions occupying more than about 10 percent of the exchange complex exhibit dispersion provided that the electrolyte concentration of the fluid has a normality that is lower than 10^{-3} . This suggests that the Na bentonite that propagates through joints and fractures in rock should be very sensitive to erosion. However, the experience from dam construction concerns flow velocities that are very much higher than those expected in rock joints, and a more relevant estimation of the erodibility is required. It can be derived by considering the involved physics and such an approach was made a few years ago, the intention being to calculate the drag forces caused by flowing water and comparing them with the interparticle bonds of the very dilute gel front in the fractures (14). The average particle bond strength derived from viscometer tests was found to be in the order of $10^{-13} - 10^{-12}$ N for porewaters of low salinity, which is also in agreement with data reported by other investigators (15).

The drag forces exerted on the particles constituting the clay gel were estimated by applying hydrodynamics (cf. Fig 27), the average force being in the range of 10^{-11} to 10^{-15} N at flow rates varying from 10^{-3} m/s to 10^{-6} m/s (14). This analysis was based on the assumption that the particles have a spherical shape and that the median value and the lower and upper quartiles of the Stoke diameter of MX-80 particles are $0.15 \mu\text{m}$, $0.07 \mu\text{m}$, and $0.18 \mu\text{m}$, respectively.

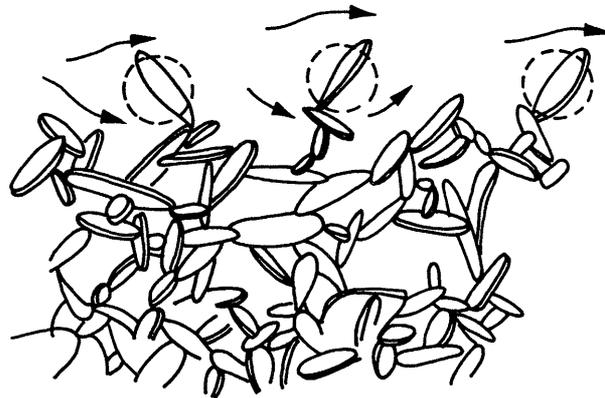


Fig 27. Schematic view of clay gel front exposed to drag forces produced by flowing water

3.1.2 The pore pressure state in the bentonite and related phenomena

The uptake of water and swelling of clay gels in fractures (cf. Fig 25) is rapid at first and then successively retarded as implied by Eq. (1). This is compatible with the general conclusion from the BMT that, under real repository conditions as well as in the BMT heater holes, bentonite confined in the holes and fractures initially absorbs water more rapidly than the rock gives off water, while later the opposite condition prevails, which gives rise to the build-up of water pressures in the fractures. Schematically, the pore pressure development at different stages of moistening in the system will be that in Fig 28. The occasion at which external overpressure appears is when the water flow rate ($\text{m}^3/\text{m}^2, \text{s}$) through the percolated, advancing clay in the fractures drops below the water inflow rate into

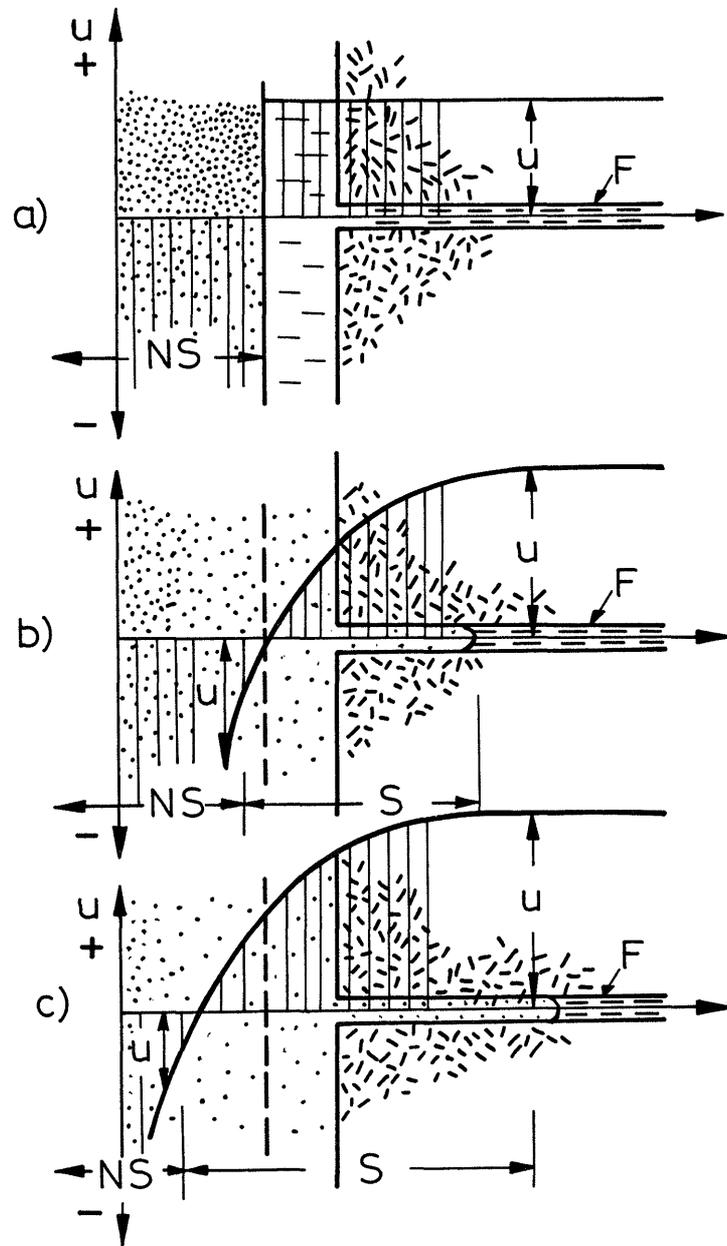
the fractures. As discussed in Volume II this happened already after a few days in the strongly water-bearing holes no 1 and 2 while it took more than 2 months in the remaining holes. Actually, no over-pressure at all appeared in hole no 6 which shows that the water absorption rate in the bentonite exceeded the inflow rate in the entire 2 year long testing time in the latter case.

Theoretically, the early development of high water pressures in the fractures in wet deposition holes, which are most critical, may have the following deteriorating effects on soft clay gels in the fractures and on the dense bentonite in the large holes:

- 1) The clay gel is pushed back in the fractures towards the holes. This would require a largely non-saturated condition of the gel except at its outer front
- 2) Water penetrates the gel and displaces the bentonite blocks
- 3) Water penetrates the bentonite and forms channels at the rock/bentonite interface in the holes

As to the behavior of soft clay gels developed from the highly compacted bentonite we see from Fig 28a that they are developed in the water phase and thereby are largely saturated from the start. Positive water pressures therefore prevail in the fractures and even in the bentonite close to the rock surface. Due to the almost complete saturation of the soft gels in the fractures, they cannot be compressed by any water pressure.

It is implicitly given by Fig 28b that the hydration power of the dense bentonite in the deposition holes yields very high negative pore pressures which prevent splitting or opening of preferential flow passages.



LEGEND: F = FRACTURE
 S = SATURATED
 NS = NON - SATURATED
 ROCK
 BENTONITE
 FREE WATER

Fig 28. Schematic pore pressure distribution in bentonite in "wet" hole. a) Application of bentonite blocks with high negative pore pressures, b) Clay gel penetrating fracture at an early stage with dominant negative pore pressures in the bentonite. c) Late stage of clay penetration with reduced suction power

The matter of erosion and formation of channels in the clay requires that the bulk density of the clay close to the rock in the deposition holes be considered. A possible critical situation may be at hand relatively early, i.e. before high swelling pressures are built up in the holes and before the backfilling of the tunnels have taken place with concomitant reduction of the hydraulic gradient in the axial direction of the deposition holes. There should be some similarity between the gas penetration at high pressures and sudden "piping" at sufficiently high, rapidly built-up hydraulic gradients. This would mean that the latter effect appears when the water pressure approaches the swelling pressure, the development of which is well known from the BMT study. Thus, in the wet holes, the swelling pressure rose to 1-1.5 MPa in about 4 months, while the maximum recorded water pressure did not exceed 0.75 MPa. It is therefore concluded that piping conditions cannot have existed in the BMT with the possible exception of the first weeks or months. If break-through and channeling took place, complete healing occurred later in the tests when high swelling pressures were built-up.

3.1.3 Evaluation of erosion processes in the BMT

The flow rate in the water-bearing fractures that intersect the heater holes in the BMT can be roughly estimated by considering the pressure gradient that existed from the base of the holes to the tunnel floor. This gradient was particularly high in heater hole no 5 in which a steeply oriented fracture with an estimated actual aperture of 0.01-0.2 mm was exposed to the highly compacted bentonite. The fracture must have served as a major discharge for the pressurized water at the base of the hole which yields an opportunity to check the proposed relationship between flow rate and erodibility. This is because the fracture was intersected by the Kovari boreholes

in which water sampling and measurements took place regularly. These holes had been drilled parallel to the heater hole and were located close to the large heater hole, the distance between the two hole peripheries being only about 5 cm (Fig 29). Applying elementary hydraulic relationships for flow in a plane slot the average flow rate can be expressed as:

$$v = \frac{d^2 \cdot g \cdot \rho_w \cdot i}{12\eta} \quad (2)$$

where v = flow rate, m/s

d = slot aperture, m

g = gravity, m/s²

ρ_w = bulk density of water, kg/m³

η = viscosity of water $5 \cdot 10^{-4}$ Pas

i = hydraulic gradient

For $d = 0.01$ mm the flow rate is about $0.0002 \cdot i$ m/s, while for $d = 0.2$ mm the rate is about $0.07 \cdot i$ m/s. The recorded water pressure at the base of the hole amounted to about 700 kPa during the major part of the test and since the water flowing through it is expected to have been discharged into the Kovari holes at about 2 m distance from the base, the average hydraulic gradient must have been in the order of 35. For an assumed fracture aperture of 0.01 mm this gradient would have caused a rate of water flow of about 0.007 m/s, while 0.2 mm aperture would have caused a flow rate of about 2 m/s. The estimated minimum rate of flow of the water that percolated the fracture into which bentonite penetrated can safely be taken as 0.001 m/s, which yields drag forces ranging between 10^{-12} to 10^{-11} N for particle or floc diameters between 0.1 and 2 μm (14). As an average this range of drag forces is one to two orders of magnitude higher than the estimated average single interparticle bond and considerable erosion and clay particle migration should therefore have taken place in the Stripa case. The sampling in the Kovari holes actually showed

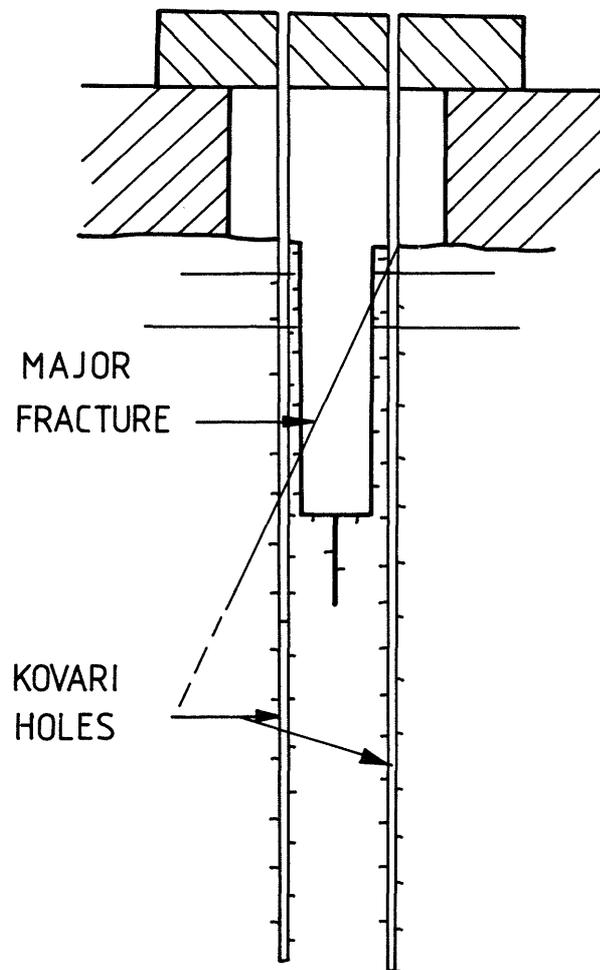


Fig 29. Heater hole no 5 and parallel Kovari holes intersected by a steeply inclined fracture

fine-grained debris suspended in the water soon after test start and X-ray diffraction tests were made to analyse the material. It turned out to be derived from the cement grouting of the metal casings and had no content of smectite. The fact that the debris appeared at all is an evidence that water was driven through the fracture in the way assumed.

The absence of eroded clay material is probably due to the fact that flocculation takes place spontaneously in natural groundwater, the coupling of particles to form flocs being obvious already at low salinities (Fig 30). These larger units of particles interact mutually and with the fracture walls which largely reduces the erodibility even of smectite clays.

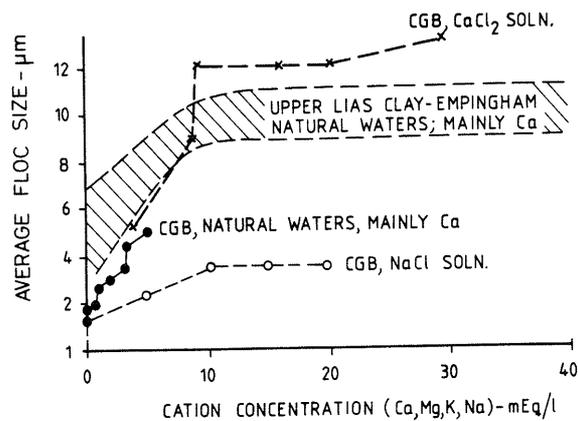


Fig 30. Variation in clay floc size as a function of the water chemistry (16). CGB is termed Cow Green Boulder Clay

In contrast to the conditions in heater hole no 5, there was no simply evaluated case of erosion in the backfilled tunnel. As long as the suction power of the tunnel backfill was stronger than the ability of the surrounding rock to give off water, there was of course no eroding effect of water flowing from the rock into the backfill. At a rather late stage the degree of saturation had become so high in the lower half of the backfill that water in the tunnel floor started

flowing in the axial direction of the BMT tunnel, because of the hydraulic gradient that had been built up. This gradient was in the order of 0.3 (cf. Vol. II, Chapter 2.4.2.2) and is assumed to have been the main cause of the increase in inflowing water into heater hole no 3 after excavating in June 1984. The corresponding average axial water flow in the tunnel floor was about 1 liter per hour and assuming this discharge to have taken place uniformly through the upper 0.3 m of the 4 m wide tunnel floor we arrive at an average hydraulic conductivity of about $5 \cdot 10^{-7}$ m/s assuming the rock to behave as a porous medium. In practice, the flow took place through interconnecting individual fractures and the actual flow velocity must therefore have been in the range of $10^{-3} - 10^{-5}$ m/s. This would not have been sufficient to erode the soft bentonite gel located in the pores of the ballast grains constituting 90 % of the lower tunnel backfill and no eroding effects were therefore expected. The fact is also that no visible change in appearance or behavior of the backfill contacting the tunnel floor was observed at the excavation, i.e. no fines had been washed out. Nor could effects of this sort be observed at the tunnel walls and roof where the bulk density of the backfill was lower than at the tunnel floor.

4 ASPECTS OF THE USE OF THE BUFFER MATERIALS IN OTHER TYPES OF ROCK WITH SPECIAL REFERENCE TO THE CHEMICAL AND PHYSICAL STABILITY OF THE BUFFERS

4.1 General

The BMT buffer materials should be useful in a large variety of rocks but certain points need consideration. They are:

- 1 Hydrology
- 2 Temperature conditions
- 3 Rock stress and strength conditions
- 4 Chemical interaction of rock and bentonite

The major rock types considered here are non-weathered igneous or metamorphic rock and sedimentary rock, while salt will not be discussed.

4.2 Hydrology

4.2.1 **Fracture conditions in general**

It is concluded from the Buffer Mass Test that also virtually fracture-free rock is sufficiently pervious to yield continuous, albeit slow, water uptake of highly compacted bentonite used as canister overpack. The opposite extreme, i.e. very richly fractured rock, will yield rapid saturation by which the intermediate period of cyclic vaporization/condensation of water close to hot canisters is at minimum. This is favorable with respect to canister corrosion and temperature conditions in the canister overpack and does not present any risk of significant loss of bentonite into the fractures as long as their apertures are smaller than about one or a few millimeters. Wider fractures need to be sealed in order to prevent such loss and to reduce erosion of buffer materials in the construction and deposition periods when the hydraulic gradients will still be high. Formation of new, stress-induced fractures or widening of pre-existing

fractures may be unfavorable, and this requires that the stress conditions in the repository area should be considered.

4.2.2 Igneous rock

Granite is known to exhibit strong variations in water flow. As in Stripa, large dry volumes are separated by long range, richly water-bearing zones which usually originate from tectonic movements. The same properties are characteristic of crystalline rock in general with the exception of alkaline rock, such as gabbro, which usually has a low frequency of open fractures and a low average hydraulic conductivity. This is due to the self-healing ability of the crystal matrix, which is rich in sodium feldspar and magnesium-containing silicates like hornblende and olivine. The practical consequence of this would be that "dry" near-field conditions, i.e. the presence of only very few water-bearing joints or fractures, around drilled deposition holes and tunnels can be obtained in any type of crystalline rock. However, the hydraulically active zone formed by the blasting of a repository tunnel is expected to interact less extensively with remote, strongly water-bearing zones in gabbro-type rock than in granite. This difference is of minor importance for the water uptake and saturation of the buffer materials but may be important for the chemical composition of inflowing water and therefore of the chemical integrity of smectite-based buffer materials.

4.2.3 Metamorphic rock

The major metamorphic rock types of interest for the location of repositories are gneiss and shales. Both are usually characterized by a low average hydraulic conductivity with the exception of long range, water-bearing zones of tectonic origin. Structural anisotropy yields a corresponding difference in hydraulic conductivity and this

becomes very obvious when the stress conditions are altered in shales. Thus, reduced stresses normal to the stratification may generate flow passages along the interface between certain strata or laminae, which represent the major fracture set in such rocks. Stress-related changes in hydraulic conductivity is not so significant in gneiss with a low degree of schistosity.

The major consequence of locating deposition holes or tunnels in metamorphic rock is that the number of water-bearing joints per unit area of exposed rock is larger than that of granite. Thus, although the average gross hydraulic conductivity of granite may be higher than that of gneiss or micaceous shale, water would tend to be more accessible and flow more uniformly through the latter rocks. This is expected to be even more pronounced by the influence of stress changes. From a practical point of view the resulting higher rate of wetting of the buffer materials would be valuable.

4.2.4 Sedimentary rock

The high average hydraulic conductivity of most sedimentary rocks make them unsuitable for repositories. Possible exceptions are claystone and very stiff clay sediments that are unconsolidated in the geological sense. They are both expected to behave mechanically in a similar way as shales. The major problem in excavating caverns in them concerns the mechanical stability of excavations at large depths and since this will probably require expensive stabilization, such rocks will not be considered here. Another reason for this is the frequent intercalation of pervious strata or laminae which bring these rocks into the same group as sandstones and siltstones.

4.3 Temperature conditions

The key factor that determines the near-field temperature conditions is the quartz content and the porosity of the rock. Granite and other quartz-rich types of rock yield the lowest temperatures and gneiss with a quartz content similar to that of the Stripa granite will have the same average heat conductivity as this igneous rock (about 3.6 W/m,K). Some anisotropy is of course expected and this is the case also for water saturated shales. If the latter rocks have a quartz content of 10-20 % and a porosity of 5-10 % they exhibit heat conductivities that are approximately 60-80 % of that of granite. This percentage would drop to about 50 for saturated very stiff unconsolidated clayey sediments if their quartz content is not lower than about 20 %. For comparison it should be mentioned that water saturated limestone with a porosity of 10-20 % and no quartz content has a heat conductivity of at least 2.2 W/m,K.

It is concluded from this that almost any type of water saturated rock is sufficiently thermally conductive to let the heat produced by canisters be dissipated so effectively that the temperatures of the bentonite and canisters will only be slightly higher than when granite constitutes the host rock.

4.4 Rock, stress and strength conditions

4.4.1 Structure

In contrast to igneous rock like granite, metamorphic rocks exhibit large structural variations and varying stress/strain properties, which has a bearing on the stress state. An exception is the rather common homogeneous granite gneiss, which shows rock mechanical and hydrological properties that are similar to those of granite which typically has two steeply oriented fracture sets and one that is

subhorizontal. Usually, most gneiss outcrops as well as most shales are structurally anisotropic and typically have one fracture set parallel to the planar elements and one set perpendicular to the fold axis. Their regularity and extension is usually less obvious than in granite and the general appearance of gneiss is often that of a wavy, dense brickwork. The anisotropy of gneiss usually has the character of coarse-grained irregular bands in which the schistosity is poorly defined because of the preponderance of quartz and feldspar over micaceous minerals. Very often different minerals dominate in the various strata and laminae, quartz, biotite and chlorite often being major constituents of adjacent structural elements. This yields a considerable variation in elasticity and strength as well as in thermal and creep properties also within a rather small rock volume. Usually, the extension of fractures that are perpendicular to the planar elements is much less than in granite, which has a strong bearing on the hydraulic properties as mentioned earlier in the text. Shales of the slate and schist types have a perfect cleavage which yields far-reaching propagation of fractures that can develop along the layering.

4.4.2 **Rock stress conditions**

Low tangential stress conditions at the periphery of deposition holes and tunnels with a circular cross section prevail for a certain range of the ratio of the maximum and minimum primary stresses in planes that are perpendicular to the axis of symmetry. Usually, one of these stresses, which are often principal stresses, is considerably higher than the other one which means that the tangential stress varies over the periphery. When the ratio is 3 the tangential stress is zero in certain parts. If a radial pressure is exerted onto the periphery by the hydration of highly compacted bentonite contained in such holes

or tunnels, tension stresses are generated in the rock which may cause failure in the form of axially oriented tension fractures that propagate radially from the periphery. Such fractures are developed when the tension strength has been fully mobilized and since this strength is at least 5-10 MPa, we find that the maximum allowable swelling pressure is of the same order of magnitude. This is demonstrated by Fig 31 which shows the tangential stress conditions at the periphery of a deposition hole before swelling of its content of highly compacted bentonite, and after the development of a swelling pressure of 10 MPa. The stress conditions at larger distances are also demonstrated in this figure. In practice, this means that when the critical primary stress ratio is equal to or higher than 3, a swelling pressure of 5-10 MPa may trigger failure in tension. This reasoning is very conservative because the tension strength of unfractured crystalline rock is usually considerably higher than 10 MPa. Since the stress ratio referred to is often in the range of 2-3 in practice it is recommended to choose a suitable, not too high bulk density of the bentonite and to determine the general primary stress to make sure that there is no extreme stress anisotropy in the rock mass. It is concluded that the density of the highly compacted bentonite used in the Buffer Mass Test was very suitable in this respect.

The risk of development of tension fractures should not be overstressed, however, since it is not critical to the use of highly compacted bentonite as canister overpack. Thus, if such fractures should be formed they will have a very small aperture and will tend to be self-sealed by bentonite that propagates into them.

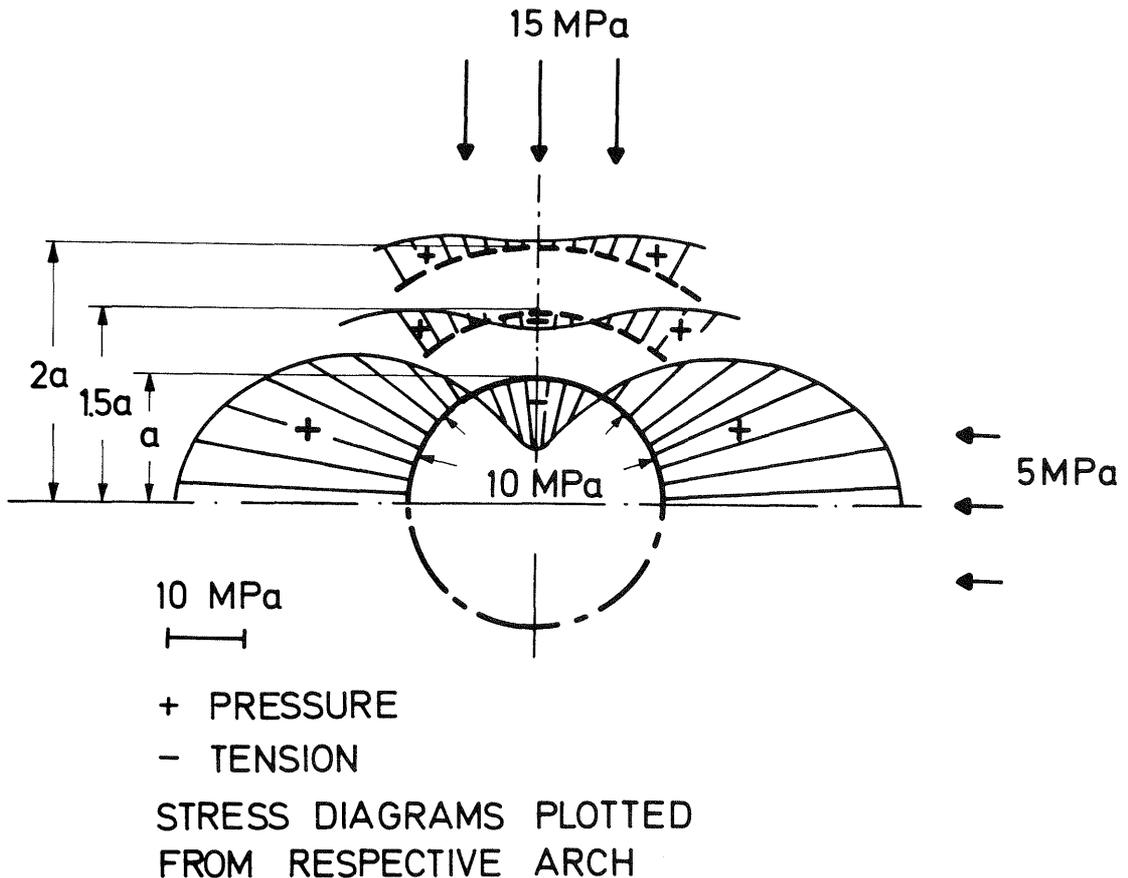


Fig 31. Tangential stress conditions at the periphery of a deposition hole with its axis oriented in the direction of the minor principal stress σ_3 of the rock. The ratio of the major and intermediate principal stresses is 3, while the internal swelling pressure is taken as 10 MPa

Strong anisotropy with respect to the structure should be associated with a strongly anisotropic primary stress state. This suggests that the afore-mentioned critical stress ratio conditions should be abundant and that high swelling pressures exerted by highly compacted bentonite may frequently trigger tension fracture in deposition holes and tunnels. The impression is, however, that the occurrence of flexible planar structures, like mica-rich laminae yield creep properties of gneiss and mica-rich shales that tend to reduce high

deviatoric stresses and there are reasons to believe that the principal stress ratio in such tough rock should be lower than that in brittle granite.

The risk of opening up series of laminae in shales and shaly gneiss by blasting is wellknown from ordinary foundation engineering which suggests that metamorphic rock with very well developed schistosity is unsuitable. Gneiss of the vein type with interfingering structural components would do well, on the other hand.

4.5 Chemical interaction of rock and bentonite

Two major chemical effects on bentonite have to be considered, one being the integrity of the smectite crystal lattice, the other one being ion exchange processes.

Smectite alteration leading to non-expanding minerals is generally assumed to require heat-induced charge changes of the smectite crystal lattice and the fixation of potassium that is offered by the ground water or by K-bearing minerals in the rock that surrounds the bentonite buffer materials. This means that the charge change, being an intralattice phenomenon, takes place regardless of the type of ambient rock. The rate and degree of such changes depend on the temperature but as described previously, the difference in temperature will be very slight also if a large range of rock types is considered.

The major potassium source in metamorphic rocks is the same as in igneous rock, i.e. K-bearing feldspars and micas and to that comes dissolved potassium moving from remote sources with flowing groundwater.

Ion exchange effects in highly compacted bentonite will be moderate. They can appear when the groundwater salinity is very high or when the calcium concentration is high. The main effect is aggregation of weakly associated clay particles in interaggregate positions in the dense bentonite, which increases the hydraulic conductivity by one or two orders of magnitude. In soft bentonite gels, such as those in the pores of the ballast in bentonite/ballast mixtures, ion exchange to calcium or the influence of a largely increased salinity has a corresponding strong influence on the hydraulic conductivity (Figs 32 and 33). These negative effects can be partly compensated for by increasing the bentonite content in bentonite/ballast mixtures.

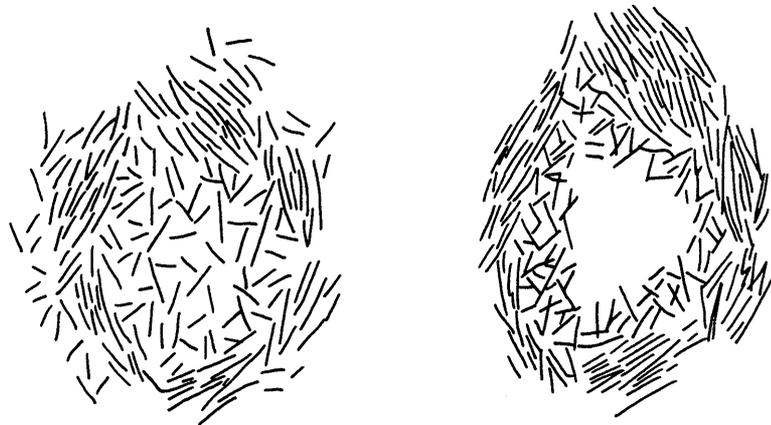


Fig 32. Influence of pore water salinity on the clay particle arrangement at low bulk density. Left: low salinity; Right: high salinity.

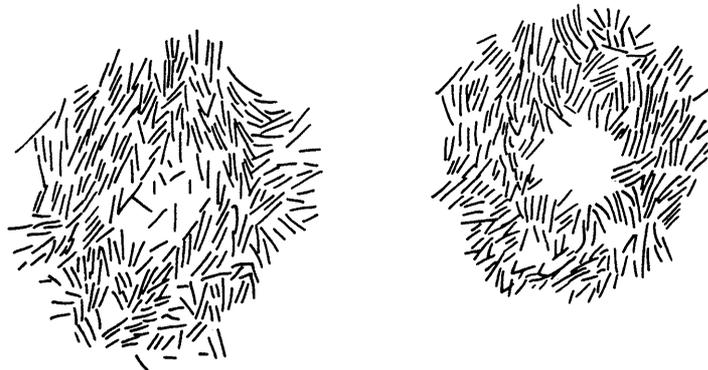


Fig 33. Influence of pore water salinity on the clay particle arrangement at high bulk density. Left: low salinity; Right: high salinity.

GENERAL CONCLUSIONS

The main conclusions that can be drawn from the BMT with respect to the chemical and physical integrity of the bentonite component are the following:

- 1 No unexpected change in crystal lattice constitution of the smectite minerals took place in the 1 year long test with water saturated bentonite at temperatures up to 125°C. The only sign of a possible alteration was a slight indication of Si-precipitation, and a tendency of reduced swelling on EG-treatment, possibly caused by embryonic mixed-layer formation. No change in physical properties like the swelling ability and the hydraulic conductivity was observed.
- 2 The erodibility of soft Na-bentonite gels exposed to flowing water in fractures is less than or similar to what can be expected on theoretical grounds. Thus, the very soft gel front of bentonite propagating into fractures from one of the heater holes was not noticeably affected by the water percolating them. The estimated rate of water flow was 10^{-3} m/s, which is assumed to exceed the flow rate in fractures intersecting deposition holes in a true repository soon after the application the backfilling.
- 3 No erosion leading to wash-out of clay particles from the sand/bentonite tunnel backfill could be observed. The rock fractures adjacent to it were percolated at flow rates that were in the same order of magnitude as those in the heater hole. The filter-type composition of the tunnel backfill offered an effective resistance to erosion and no wash-out of fines was expected even if the flow rates had been considerably higher.
- 4 Provided that the temperature can be kept at an acceptable level, almost any type of rock - salt not being considered - can be accepted for hosting repositories as far as the physical and chemical integrity of the bentonite-based buffer materials are concerned. The only major point that requires attention is the presence of and potential for development of wide fractures. With access to suitable fracture sealing substances and techniques the spectrum of potentially useful host rocks can be very wide.

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