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Clay colloid formation and release from MX-80 buffer

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December 1999

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ABSTRACT

Flowing groundwater can tear off clay colloids from buffer clay that has penetrated into fractures and transport them and bring sorbed radionuclides up to the biosphere. The colloids are 2-50 μm particle aggregates that are liberated from expanded, softened buffer if the water flow rate in the fractures exceeds a few centimeters per second. Except for the first few months or years after application of the buffer in the deposition holes the flow rate will not be as high as that. The aperture of the fractures will not hinder transport of colloids but most of the fractures contain clastic fillings, usually chlorite, that attract and immobilize them. This condition and the flow rate criterion combine to reduce the chance of radionuclide-bearing clay colloids to reach the biosphere to practically zero except for certain cases that need to be considered.

LIST OF CONTENTS Page

SAMMANFATTNING

De minsta smektitpartiklarna som transporteras av strömmande vatten är aggregat som innehåller hundratals till tiotusentals packar av smektitkristaller. Strömmande grundvatten can riva loss sådana kolloider från buffertlera som trängt in i bergsprickor och transportera dem med sorberade radionuklider till biosfären. Kolloiderna är 2-50 μm stora partikelaggregat som frigörs från den i sprickorna expanderade och uppmjukade bufferten om flödeshastigheten överskrider några mm per sekund. Det är dock normalt endast under de första månaderna eller åren efter buffertens anbringande i depositionshålen som hastigheten kan vara så hög.

Buffertlera kan penetrera sprickor med större öppning än ca 100 μm. De flesta naturliga eller av spänningssituationen nybildade sprickorna har mindre vidd än så men vissa spänningsförhållanden och sprickorienteringar kan orsaka expansion till betydligt mer än 100 μm varigenom lera kan vandra in. Bergets spänningssituation och struktur bestämmer sålunda lerinträngningen, som kan uppskattas till högst några få meter. Strömmande grundvatten kan erodera lerans lösa front och ge upphov till kolloider. Emellertid innehåller de flesta av sådana sprickor klastiskt material av klorit som kan adsorbera och fixera kolloiderna. Detta förhållande i samband med strömningshastighetskriteriet gör att möjligheterna för radionuklidbärande kolloider att nå biosfären är praktiskt taget inga alls utom för vissa speciella fall.

SUMMARY

The smallest smectite clay particles transported by flowing water are aggregates containing hundreds to tens of thousands of individual stacks of lamellae. Moving groundwater can tear off such colloids from buffer clay that has penetrated into rock fractures and transport them and bring sorbed radionuclides up to the biosphere. The colloids are 2-50 μm aggregates that are liberated from expanded, softened buffer if the water flow rate in the fractures exceeds a few mm per second. Except for the first few months or years after application of the buffer in the deposition holes the flow rate will normally not be as high as that.

Buffer clay can penetrate fractures that have an aperture exceeding about 100 μm. Most natural or new fractures formed by critical stress conditions are tighter than that but certain stress conditions and fracture orientations may cause expansion to much more than 100 μm by which buffer clay will move in. The rock stress and structural conditions hence determine the extent to which buffer clay can move into the rock, an estimated maximum depth being a few meters. Flowing groundwater may erode the soft front of the clay that has penetrated into fractures and produce colloids. However, most of these fractures have clastic fillings of chlorite that attract and immobilize the colloids. This condition and the flow rate criterion combine to reduce the chance of radionuclide-bearing clay colloids to reach the biosphere to almost none except for a few important cases.

1 DEFINITIONS

Clay particles and aggregates of colloidal size may be released from the dense clay buffer and move into fractures in the rock surrounding the deposition holes. If radionuclides are sorbed on the colloids and these are transported far by the moving clay they may reach the biosphere in practically important quantities. This matter is dealt with in the present report, focus being on the physical state of the fractures with special respect to the stress state and the fracture fillings, as well as on modeling the clay migration and colloid formation processes.

The term colloidal refers to ”dispersed suspensions” [1] and implies that only particles smaller than 0.1 μm can serve as radionuclide carriers. In practice, also bigger ones can have this function as outlined in the present report.

2 CRITERIA

Radionuclide-bearing colloidal particles may be released from the dense buffer and move from the deposition holes into rock fractures. The following conditions apply for making clay colloid transport from deposition holes of radionuclides important:

1. Released clay particles and particle aggregates must be small as compared to the rock fracture aperture

2. The front part of fracture-penetrated clay must be very soft with very low cohesion for making release of particles and particle aggregates possible

3. There must be either spontaneous or erosion-induced release of clay particles or aggregates

4. Groundwater flow along and further through intersected fractures to the biosphere must be sufficient to transport released clay

5. Sorption of particles and particle aggregates to fracture surfaces or clastic fracture fillings must be insignificant

6. The propagating clay must not meet an obstacle in the fracture, like constrictions or clastic fillings, at which it can accumulate and consolidate.

3 PARTICLE SIZE CONSIDERATIONS

3.1 Clay particle size

The size of montmorillonite particles emanating from dispersed MX-80 clay is illustrated by Figure 1, which shows the distribution of the maximum diameter (crystallographic a/b plane) of stable stacks of lamellae. The clay in the experiments yielding these data was ultrasonically dispersed and maintained disintegrated by adding $Na_4P_2O_7$ [2]. One finds that about 5 % of the particles are smaller than 0.1 μm while 40 % of the particles are smaller than 0.15 μm. This latter fraction, which is taken to be colloidal with some approximation, has a specific surface area of at least 2000 m^2 per gram solid mineral mass and hence represents the most surface-active part of MX-80 clay.

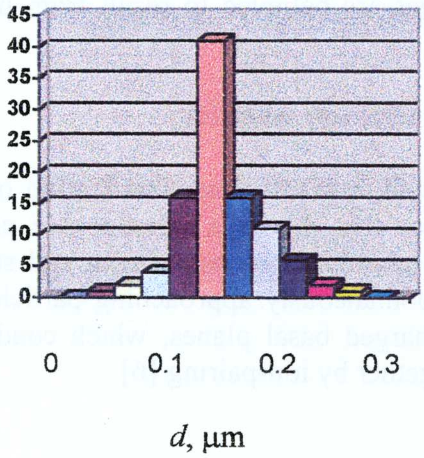

Figure 1. *Frequency in % (vertical axis) of maximum diameter d of small particles of effectively dispersed Na montmorillonite as evaluated by transmission electron microscopy [3]. 0.1 μm=100 nm.*

3.2 Clay particle aggregates

3.2.1 General

A major question in the present context is whether discrete particles of colloidal size can really appear in the groundwater keeping aggregation and microstructural features in mind. Aggregates of colloidal clay particles result from coagulation of discrete stacks of lamellae or small aggregates of such stacks, or represent particles released as such from the buffer. In the first case the aggregates result from successive concentration of particles to a size that yields mechanical stability, while in the latter case they originate from successive expansion and disintegration of initially denser clay. The size and strength of either aggregate type may or may not be the same.

3.2.2 Aggregation of dispersed particles released from the buffer clay

Processes

Dispersed particles move by Brownian molecular motion and easily form gels if the charge is favorable. The motion is both rotational and translatory and the highest probability is that particle contact is established edge-to-edge [4]. However, the charge distribution over the particle surfaces and also the conditions for bringing particles in close contact affect the way in which adjacent particles make and remain in contact.

The most simple structure model implies that coagulation of dispersed particles takes place by Coulomb-type attraction of nearby particles, i.e. through bonding of the commonly positively charged part of one particle and negatively charged parts of an adjacent particle. There is strong evidence that the edges of the stacks are positively charged in the neutral and acid pH ranges (pH <7-8), cf. Figure 2, while the basal planes carry a negative charge, hence yielding edge-to-face aggregation.The positive charges are believed to result from processes of the type indicated by Eq.1 [5].

$$\text{Al-O-Al} \dashrightarrow (\text{Al-O} + \text{Al}) + (\text{H OH}) \dashrightarrow \text{Al-(OH)} \qquad (1)$$

Negative edge charge is believed to result from dissociation of structural OH. However, it has been proposed that this is a questionable process since the hydroxyls are bound to lattice aluminum or magnesium and stay in the lattice but in protonated form [5]. Spontaneously approaching particles with negative edge charge and negatively charged basal planes, which conditions prevail at pH>8, can become bonded together by ion-pairing [6].

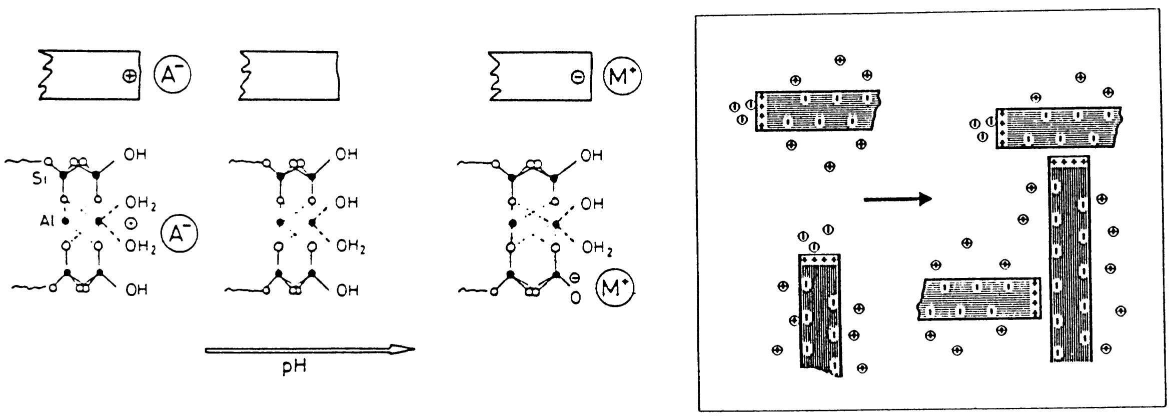

***Figure 2**. Edge charge conditions as a function of pH in the porewater [5].*

Smectite clay particles with the same surface charge in suspensions can also associate in a face-to face fashion by electrical double layer interaction [6]. The coupling is weak, however, except at low and moderate electrolyte contents. Such association may take place spontaneously in suspensions but it commonly emerges from compressed ("consolidated") or sheared states. It is assumed that association in edge-to-edge (reticulate) form primarily evolves from the rotationary motion of dispersed particles with ion-pair bonding being responsible for this type of interparticle coupling [4]. Edge-to-face coupling caused by Coulomb attraction of different edge and basal plane charges may in fact be face-to-face contacts since the edges are often very thin and curved and micrographs may hence give the same impression as a true edge-to-face coupling. Similarly, edge-to-edge contacts may in fact be of edge-to-edge or edge-to face type.

During the course of face-to-face coupling, lateral growth leading to large, i.e. micrometer-sized, flat aggregates may occur. Light scattering and viscosity measurements on montmorillonite in the pH range 6-13 have been interpreted as ribbons of edge-to-edge flocs, but cardhouse-type edge-to-face association may also be formed [6]. The latter is believed to dominate at neutral and acid pH.

Structure and size

Dispersed particles in suspensions are affected by Brownian motion that provides opportunities for interparticle coupling of both equally and differently charged crystallites if the concentration of solids is sufficiently high. Thus, in principle, suspensions are not stable but undergo coagulation through formation of particle aggregates. Both edge-to-edge and edge-to face coupling take place.

Aggregation by coagulation of discrete montmorillonite particles and small aggregates have been investigated by use of transmission and scanning electron microscopy [7]. The experiments had the form of mixing finely powdered montmorillonite-rich bentonite clay (SWY-1) with water and freeze-drying specimens after homogenization. Figure 3 illustrates the arrangement of particles in very soft gels (<1060 kg/m^3) of cardhouse type formed spontaneously from clay sols. The micrographs show many apparent edge-to-face contacts and edge-to-edge couplings as well as of the face-to-face type but they may in fact all be of the same type.

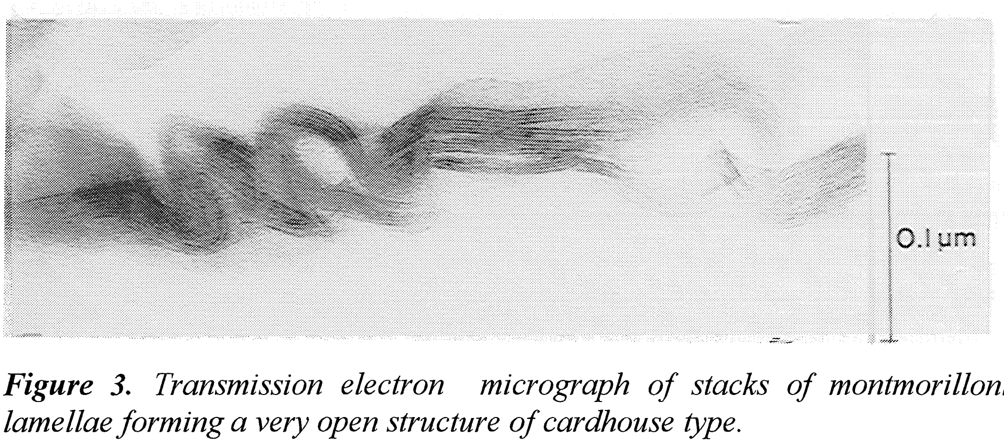

Figure 3. *Transmission electron micrograph of stacks of montmorillonite lamellae forming a very open structure of cardhouse type.*

Aggregates forming "Schweeb-stoffe" in fresh-water rivers have been investigated by light and electron microscopy [8]. Very soft, porous ones are assumed to result from spontaneous coupling of discrete particles or small aggregates while denser ones are expected to be soil fragments torn off from the sediments by the rivers. Figures 4 and 5 represent typical soft aggregates with a typical diameter of 10-50 μm.

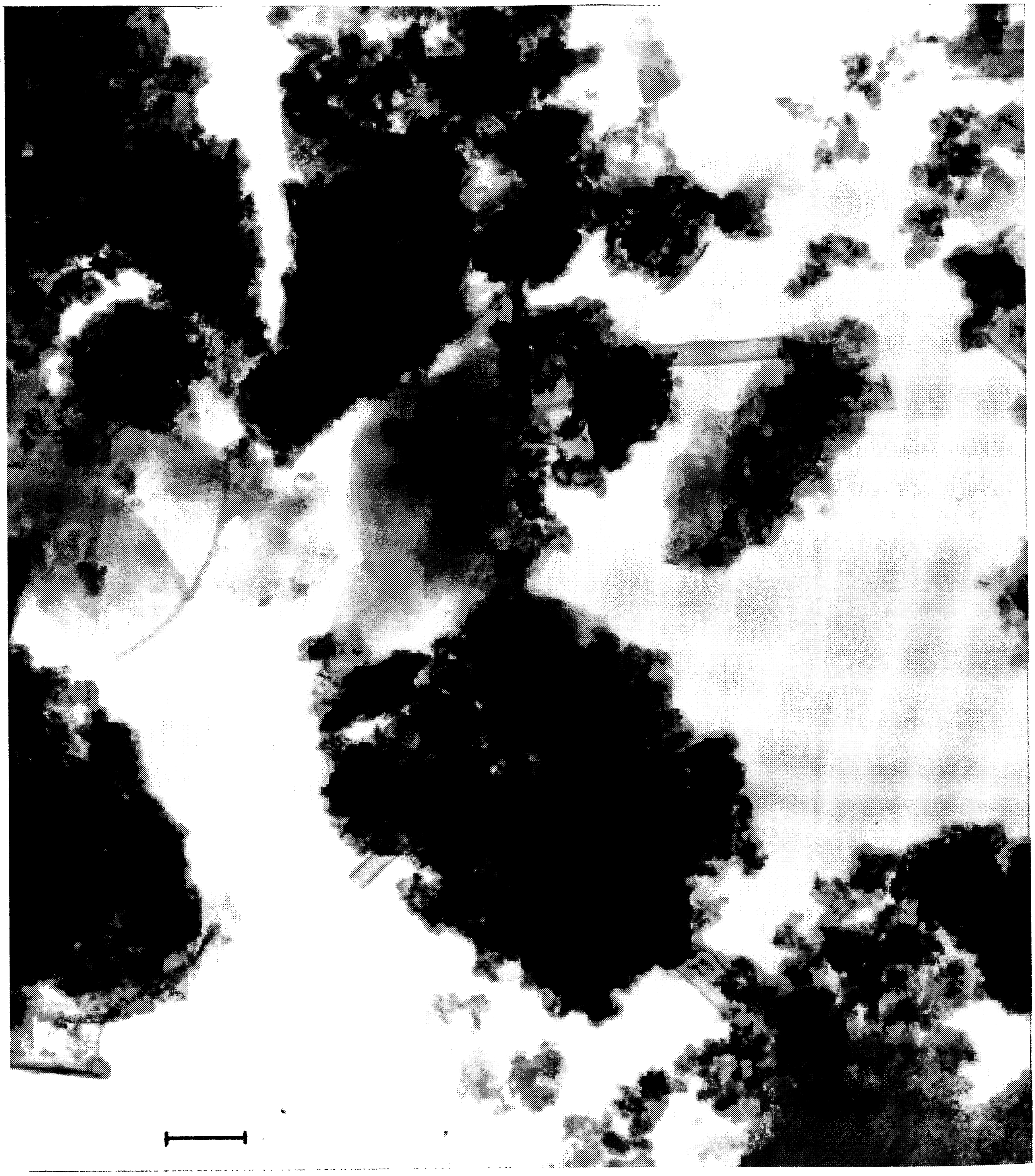

Figure 4. *Transmission electron micrographs of smectite particles forming aggregates in the river Elbe. The diameter is about 10-20 μm [8].Bar=10 μm.*

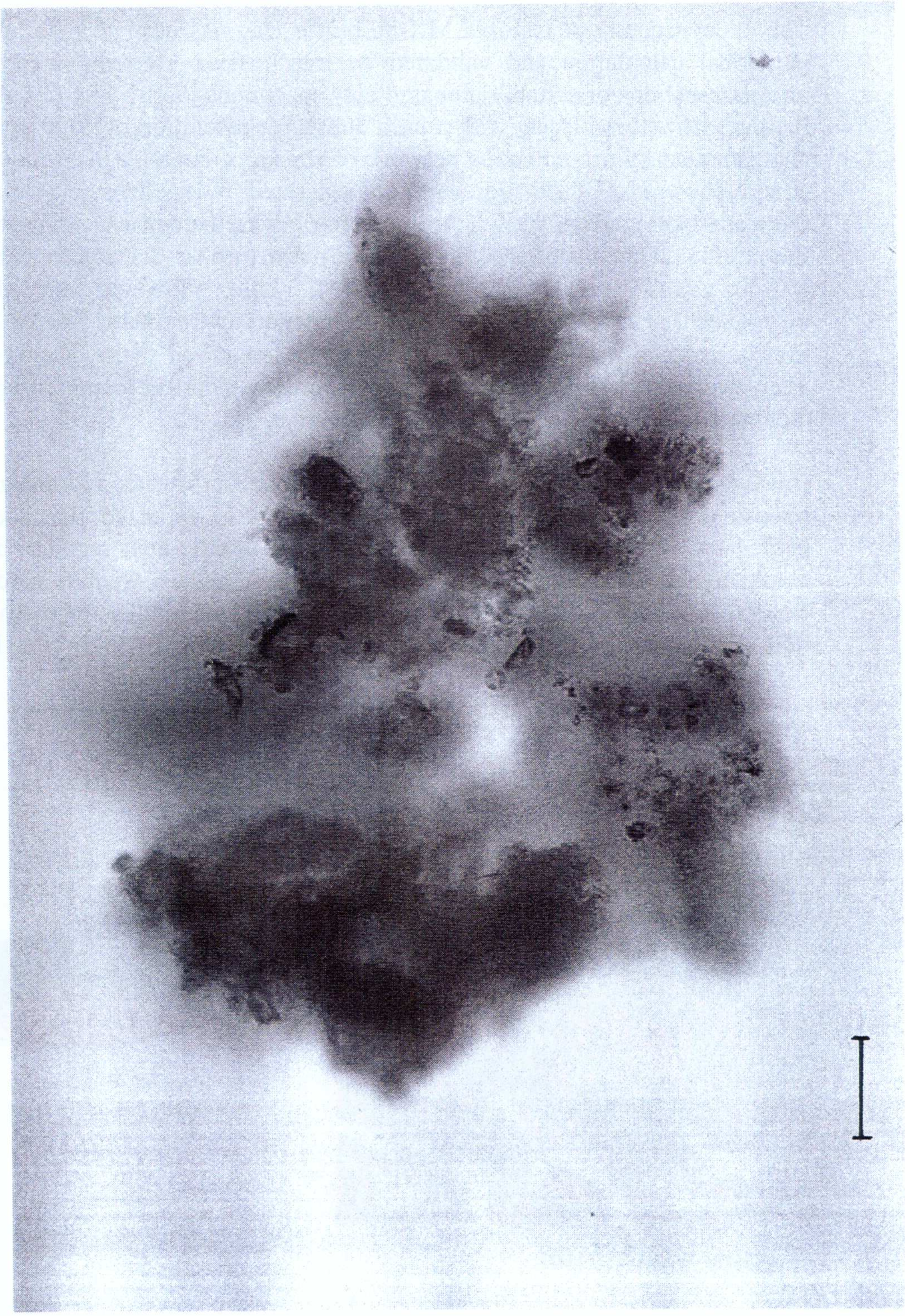

Figure 5. *Light-microscope picture of smectitic mixed-layer particles forming aggregates in the river Oder. The diameter is about 20-50 μm [8].Bar=20 μm.*

3.2.3 Aggregates released as such from the buffer clay

Expansion phase

The microstructure of matured MX-80 buffer clay has been modeled by use of numerical calculation and validated by transmission electron microscopy of ultrathin sections of suitably prepared clay specimens [9,10]. Figure 6 illustrates the microstructure of clay with a bulk density at saturation of 2000 kg/m^3. It is characterized by a continuous network of stacks of lamellae with more or less continuous voids. Dense aggregates are separated by less dense clay matrix and there are local voids as well. To some minor extent the density variations mirror the internal structure of the original grain structure of the compacted MX-80 powder grains, which have a size of 0.1 to 1 mm. However, since the dense aggregates in micrographs like Figure 6 have an approximate size of only 1-3 μm the original powder grains are split in the course of the hydration process. The remaining aggregates cannot expand further under the confined conditions that prevail in the buffer clay in deposition holes.

If water saturated clay of high density is free to absorb water and expand into a fracture in the rock surrounding the buffer, it is expected that the denser aggregates expand while the already swollen soft matrix does not. The net result would be a uniform, regular network of stacks of lamellae, which is exactly what experiments have shown. Thus, Figure 7 illustrates the regular microstructure of fully or almost fully expanded MX-80 clay with a density of 1050-1100 kg/m^3.

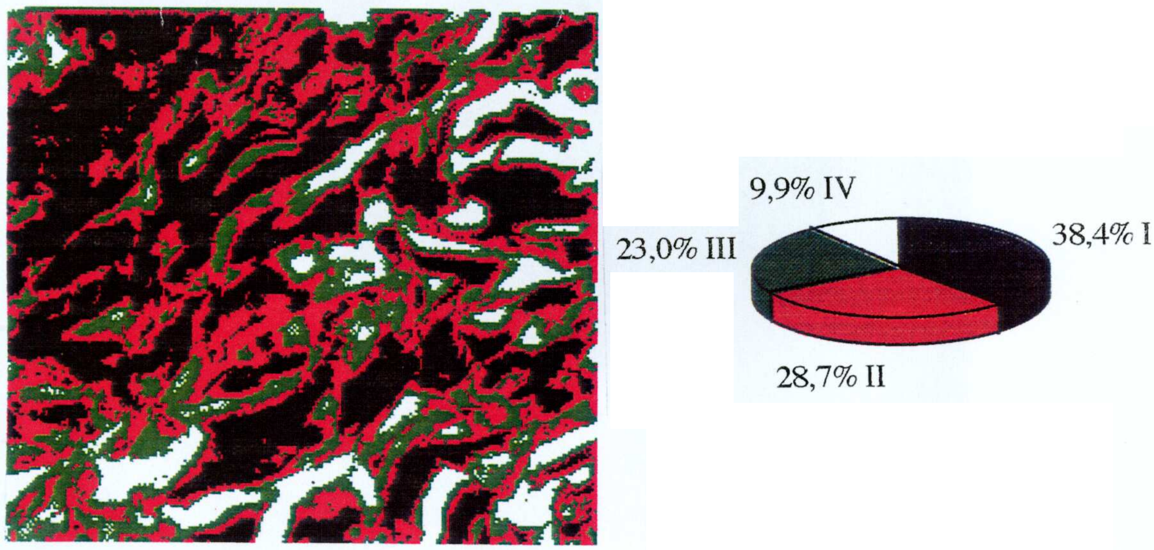

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Figure 6. *Scanning electron micrograph of MX-80 clay with a bulk density of 2000 kg/m^3 [10]. The micrograph has been digitalized to make density variations obvious: Black: Most dense parts (density exceeding 2000 kg/m^3), Red: Second densiest parts (about 2000 kg/m^3), Green: Medium dense parts (1500-2000 kg/m^3), and White: Open voids.*

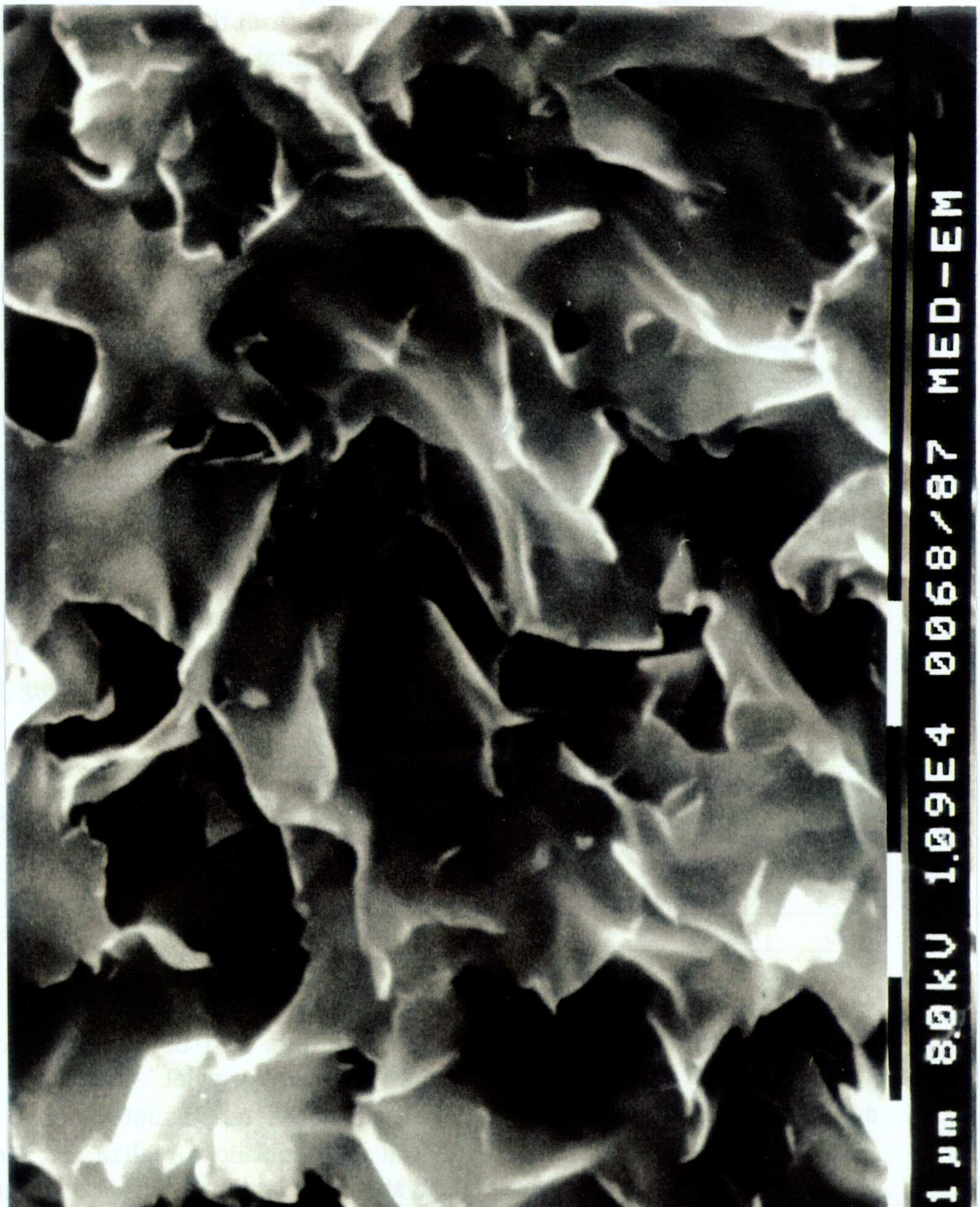

Figure 7. *Scanning electron micrograph of MX-80 clay expanded in distilled water from about 2000 to 1100 kg/m*^{3}.

Studies of this sort indicate that sponanteous breakage and disintegration of fully expanded smectite gels do not take place even in distilled water. Instead, disintegration appears to require external impact like erosion by water flowing along the gel front, or gas-induced water flow from the central part of the buffer out through the soft clay gel. Strong thermal agitation like boiling may also yield disintegration.

Release phase

Erosion of uniformly expanded clay takes place if the drag forces induced by flow along the gel front exceeds the particle bond forces. This matter has been investigated in a comprehensive earlier study [3], in which the drag force on particles/aggregates of various size was determined by applying Stoke's law neglecting gravity, and calculating the interparticle bond strength by dividing the bulk strength by the number of bonds evaluated from microstructural estimates as summarized by Table 1. The average density of the smectite gel was taken as 1050 kg/m^3, corresponding to the soft front of the buffer moving into fractures.

Table 1. *The ratio F_d/F_b of the drag force (F_d) and bonding force (F_b) as a function of particle/aggregate size d and water flow rate v at 15°C. The ratio is given in the respective columns. The average interparticle bonding force was taken as 4E-13 N [3].*

v, m/s	**D, 0.1 μm**	**D, 0.5 μm**	**D, 1.0 μm**	**D, 2.0 μm**	**D, 10 μm**	**D, 50 μm**
E-3	2	9	15	30	150	900
E-4	0.2	0.9	1.5	3	15	90
E-5	0.02	0.09	0.15	0.3	1.5	9
E-6	0.002	0.009	0.015	0.03	0.15	0.9

Unity or higher figures mean that the flow rate will be sufficient to tear off aggregates and make them move with flowing water. One finds that for aggregates smaller than 0.5 μm the critical flow rate is about E-4 m/s, while for 50 μm aggregates it is about E-6 m/s. It is important to realize that the primary particles, i.e. the non-associated basic stacks of lamellae with a diameter of 0.1-0.2 μm (Figure 1), are less apt to be torn off than bigger aggregates. For larger particles, gravity causes frictional resistance that contributes to the interparticle bond strength. The critical flow rate hence has a minimum for a certain particle size interval, 50-500 μm, as manifested by several sedimentological investigations. However, river bed studies suggest that the critical flow rate for erosion of particles of this size is much higher than found in this and similar investigations [11].

For the present purpose it is essential to estimate the aggregate size and flow rate. The firstmentioned matter will be considered in some more detail in the subsequent section while the flow rate issue is focused on in a subsequent chapter on rock hydraulics.

3.3 Experiments with MX-80

3.3.1 Pinhole tests

Pinhole tests for investigating the size of particle aggregates torn off from MX-80 clay have given data on the critical flow rate and on the clay mass released per time unit under constant flow rate conditions [3].

The experiments were performed by use of a cylindrical permeameter cell filled with clay gel through which a 1 mm hole had been made by a pin. The sample was then exposed to a hydraulic gradient for determining the critical gradient and the concentration of the discharged clay material for the flow rate that gave breakthrough, cf. Table 2.

Table 2. *Critical gradients, flow rates and transported clay expressed in terms of the concentration of solids in the discharged solution [3].*

Water content, %	Critical gradient	Av. flow rate at crit. g.,m/s	Discharge conc., %	Permeating solution
435	E-1	3E-3 to E-2	0.07 to 0.09	Dist. water
994	E-2 to 4E-2	5E-3 to E-2	0.01 to 0.04	Dist. water
1000	E-2	7E-4 to 6E-3	0.03 to 0.05	Weak brackish
2000	E-2 to 3E-2	2E-3 to 4E-3	0.03	Weak brackish

One concludes from these tests that the critical hydraulic gradient is about E-2 for clay gels with a density of 1030 to 1080 kg/m^3 while it is appreciably higher for denser clay. The average flow rate at break-through is about E-2 m/s for the softest gels, i.e. 10 mm per second, and the concentration of solids in the discharged solids 0.05 % at maximum.

Referring to the theoretically derived data in Table 1 one concludes that released particle aggregates in the experiments had an average size of at least 20-50 µm.

3.3.2 Piping tests

The experiments involved percolation of an initially homogeneous clay gel with a density of 1060 kg/m^3 in a small cell for photographic recording of piping processes and associated erosion and transport of particle aggregates [12]. The critical flow rate was found to range between E-5 and 5E-4 m/s, yielding aggregates with a size ranging between 2 µm for percolation with distilled water through a clay gel density of 1020 kg/m^3 to 70 µm for percolation with Ca-rich water (Forsmark) through a clay gel density of 1140 kg/m^3. The majority of the aggregates in the experiments had a size of 10-50 µm (Figures 9 and 10).

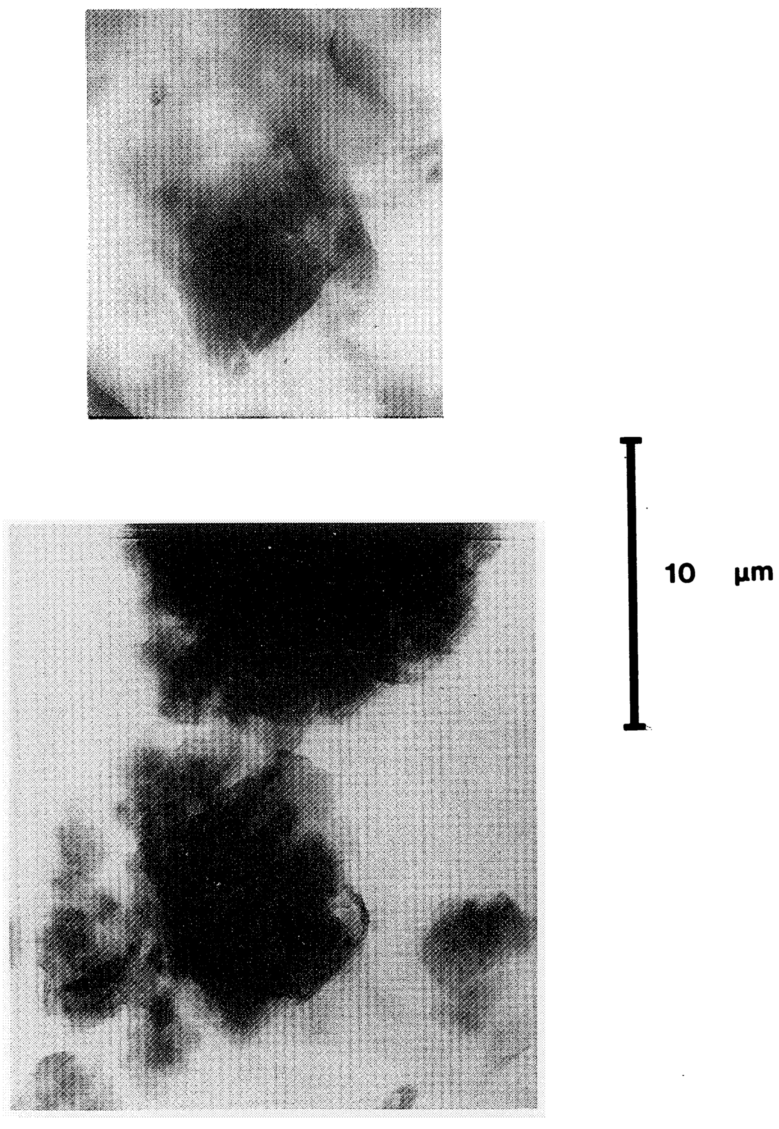

Figure 9. *Characteristic smectite particle aggregates in distilled water observed by use of "Humid Cell" High Voltage Microscopy [7].*

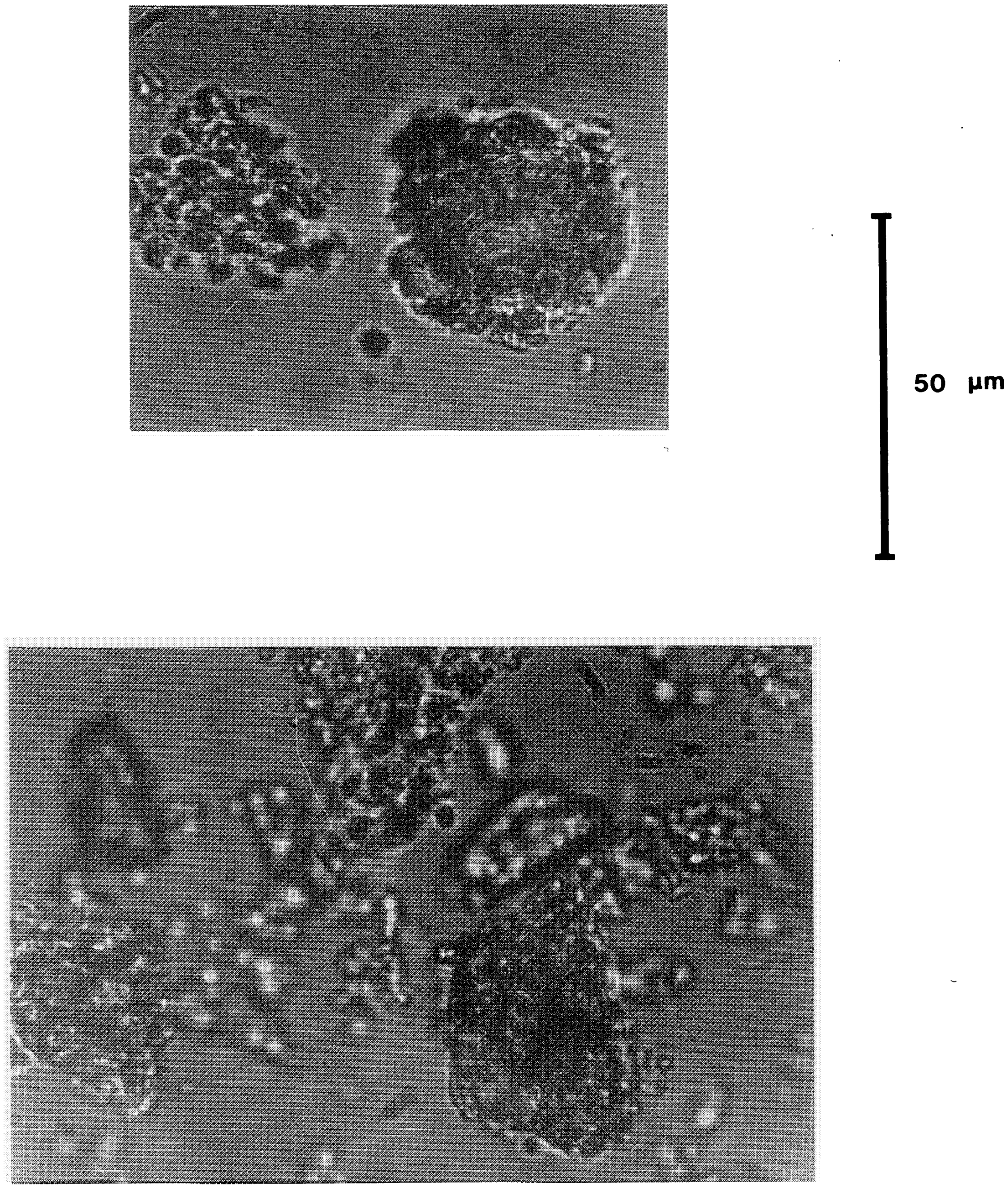

Figure 10. *Characteristic smectite particle aggregates in 35 000 ppm NaCl solution as observed by light microscopy.*

4 PENETRATION OF BUFFER CLAY INTO ROCK FRACTURES

4.1 Processes

Migration of clay particle aggregates consisting of minute smectite stacks emanating from the buffer clay requires that it is eroded by flowing groundwater or expelled by some other mechanism like gas-generated water flow. They all presuppose that the buffer has softened by expansion into rock fractures. A primary criterion is hence that such penetration can take place.

4.2 Penetrability

Penetration of clay into fractures is a function of the expandability of the clay, which is a function of the smectite content, density, and porewater chemistry. The most important parameters are, however, the aperture of the fractures, the nature of the fracture surfaces and the presence of clastic fracture fillings as outlined in the subsequent sections.

4.3 Fracture aperture

4.3.1 Rock discontinuities

The discontinuities of a rock mass can be characterized in various ways, a simplified mode of categorization being shown in Table 3.

Table 3. *Simplified categorization of discontinuities in rock [13]. L=typical length and a=typical spacing in meters.*

Order	Function and typical dimensions	SKB defin.
1st	Very large hydraulically and mechanically active fracture zones (L>E+4 m, a>E+3)	D1
2nd	Large hydraulically and mechanically active fracture zones (L=E+3 to E+4 m, a=E+2 to E+3 m)	D2
3rd	Hydraulically and mechanically active fracture zones (L=E+2 to E+3 m, a=E+1 to E+2 m)	D3
4th	Hydraulically and mechanically active discrete fractures (L=E+1 to E+2 m, a=E+0 to E+1 m)	D4
5th	Potentially active discrete weaknesses (L<E+1 m)	_

4.3.2 Rock structure

The only practically important discontinuities listed in Table 3 that will intersect deposition holes are those of 4^{th} (D4) and 5^{th} orders. According to the definitions, the latter type is neither significantly weak or water-bearing but can be activated by critical stress states and become water-bearing. The firstmentioned are long-extending fractures that are assumed to have evolved from smaller breaks in the evolution of the earth crust and been percolated by hydrothermal solutions on one or several occasions, usually in Ordovician, Permian and Tertiary time [13,14].

A number of investigations have shown that deposition holes are commonly intersected by 3-5 breaks of 4^{th} order type, two or three being steep, one inclined and one or two almost horizontal [13,14], (Figure 11). It is noticeable that such discontinuities have a strongly varying aperture and that the major water-conductive parts are their crossings. Figure 12 illustrates such a case, which in fact can be taken as a standard pattern of permeable paths in the rock surrounding deposition holes. Using bulk conductivities of E-10 to E-9 m/s, which are typical of moderately and acceptably water-bearing nearfield rock, one finds that the hydraulic fracture aperture is 10-100 μm. The true size is a bit larger but still on the same order of magnitude. Exposure to permeation of hydrothermal solutions has lead to the typical chloritic coatings and fillings [13].

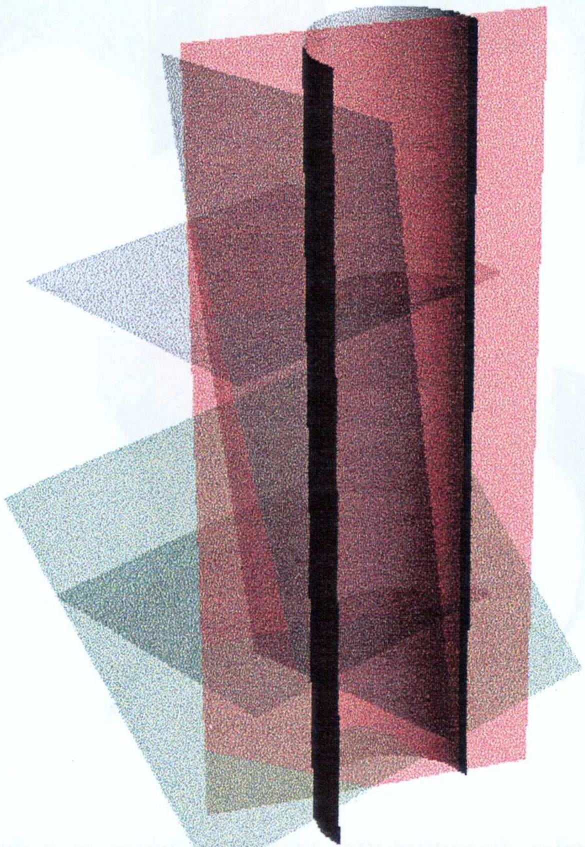

Figure 11. *Typical constellation of 4^{th} order discontinuities in the form of water-bearing fractures in a sectioned deposition hole.*

By definition, discontinuities of 5th order have a negligible water-bearing capacity but they can be activated, propagate and widen under critical stress conditions. Such conditions may be generated in the vicinity of the deposition holes that extend from the floor of TBM-drilled tunnels [13] if the primary rock stress field is strongly anisotropic.

The most critical cases with respect to significant buffer clay penetration are represented by 1) Steep fractures intersecting deposition holes such that wedges are formed (Figure 12), 2) Steep fractures intersecting the holes as in Figure 13 in rock with the maximum primary rock stress being horizontal and higher than 3 times the minor primary stress in the horizontal direction.

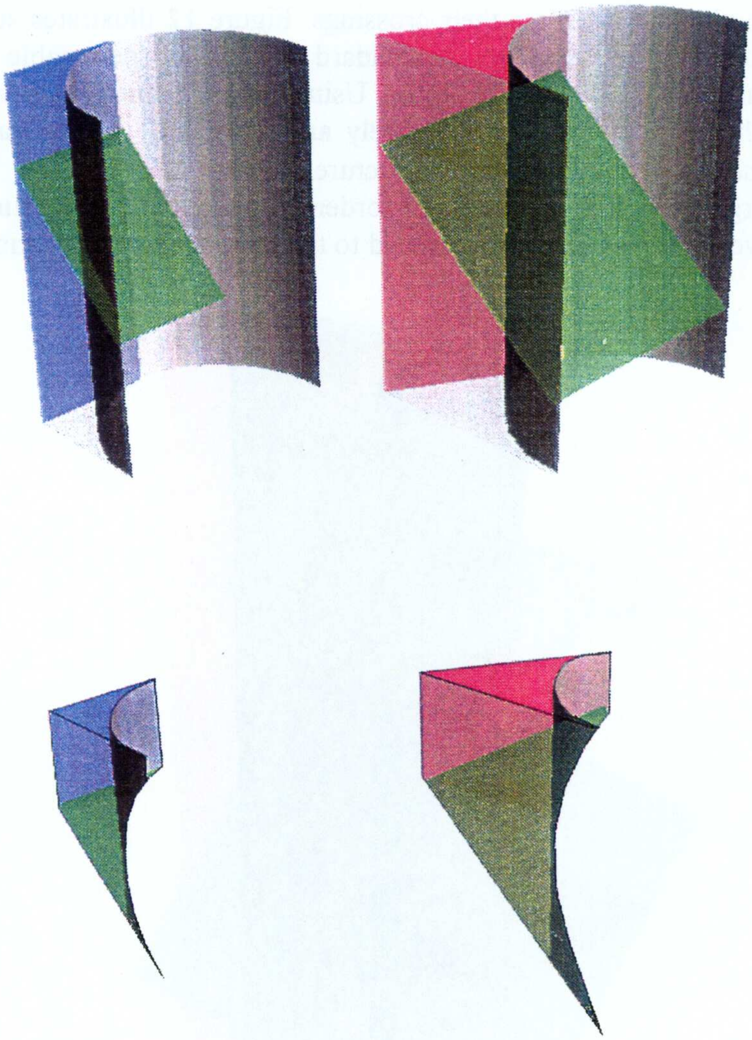

__Figure 12.__ 4^{th} order discontinuities yielding wedges in a deposition hole. The discontinuities forming the boundaries of wedges that do not fall from the rock but remain in principle in position, expand and get an increased water-bearing capacity.

Also, the special case with a primary rock stress ratio higher than 2 and buffer clay exerting a high swelling pressure can yield this state (Figure 14). However, in all these cases widening or neoformation of such steep fractures proceeds to a very moderate distance from the rock wall, i.e. a few decimeters. The large majority of critical stress conditions or practically all of them can be avoided by orienting the tunnels parallel to the major primary principal stress as illustrated in Figure 13 and by selecting positions for the deposition holes so that they will not be *diametrically* intersected by steep discontinuities of 4th order. Since the spacing of such fractures is commonly on the order of 5 m this latter criterion does not significantly limit the usability of the deposition tunnels.

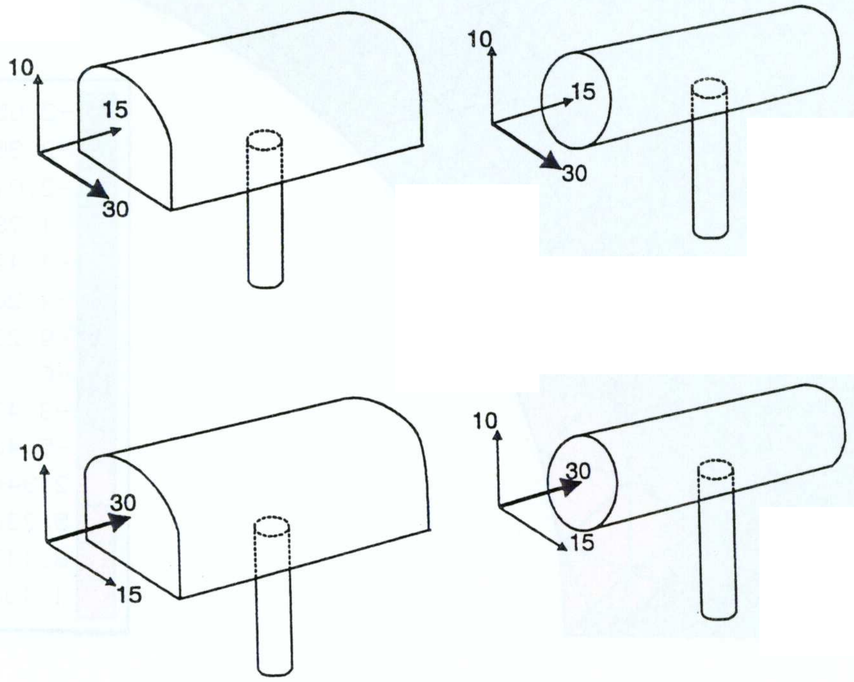

Figure 13. *Non-critical and critical tunnel orientations for avoiding tension in the rock around deposition holes.The primary principal stresses are 10 (vertical), 15 and 30 MPa, respectively. Upper: Orientation with highest risk Blasted and TBM-shape. Lower: Orientation with lowest risk. Blasted and TBM-shape.*

Figure 14 shows an example in which the principal stresses result from the nearness of the overlying tunnel that is assumed to be oriented perpendicularly to the the major primary horizontal stress. The maximum tension stress will cause tension failure in the rock yielding a vertical fracture, or activation and propagation of an already existing vertically oriented 5th order discontinuity, or expansion of a 4th order discontinuity located and oriented in this same fashion. The aperture of such discontinuities is estimated to be 500 μm at maximum.

Using normal average bulk hydraulic conductivities of granite rock in which deposition holes can be bored, i.e. E-10 to E-9 m/s, for estimating the average hydraulic aperture of the 3-6 4th order discontinuities that normally intersect the holes, one arrives at the interval 20-100 μm [15].

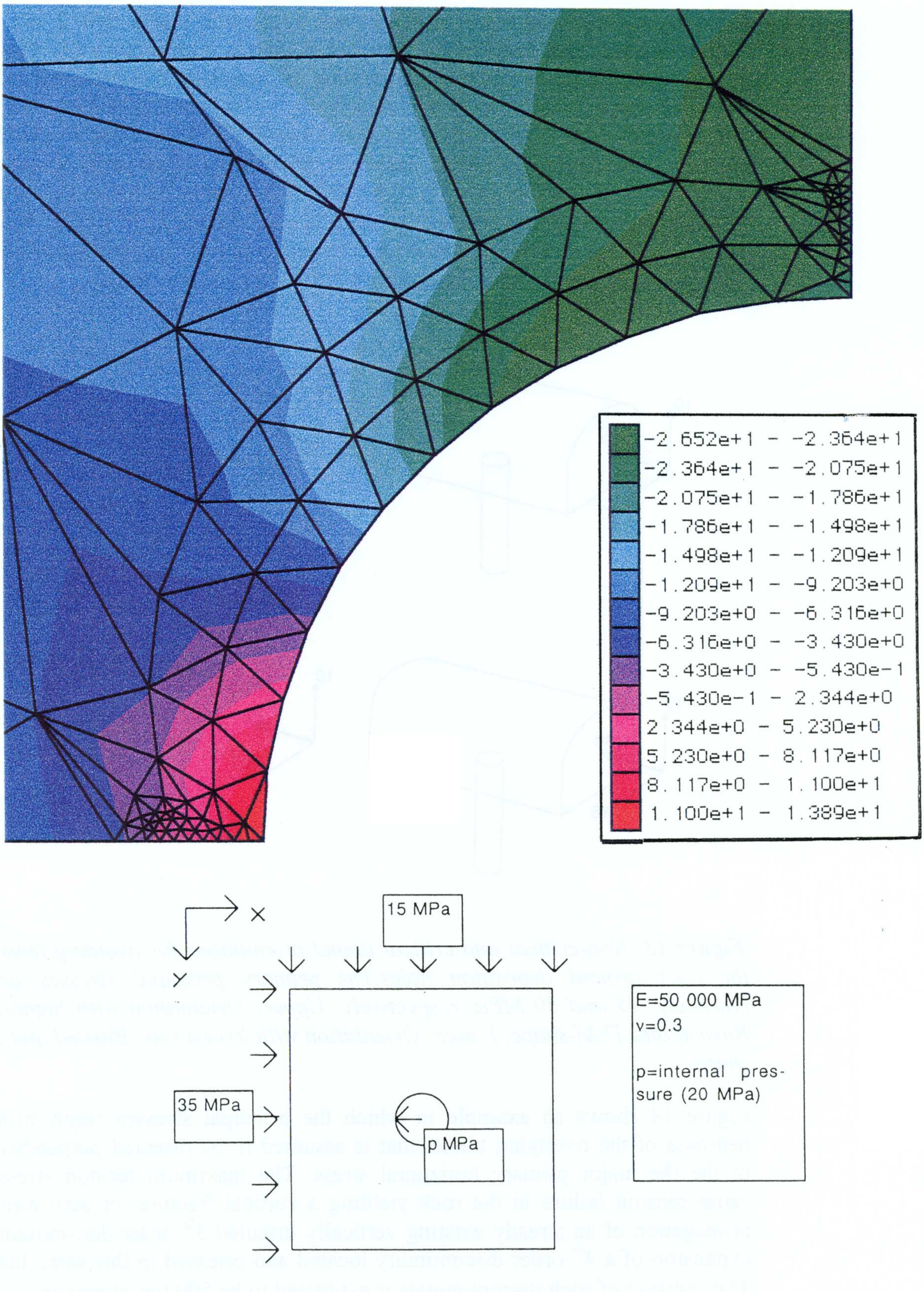

Figure 14. *Tension stresses developed in the nearfield of a deposition hole in rock with the common principal stresses 35 and 15 MPa and buffer clay with a swelling pressure of 20 MPa. The maximum (horizontal) tension stress is 1.4 E+1 MPa. (Compressive stresses have negative sign).*

4.2 Penetration process

The major issue is now to find out what fracture openings that expanding buffer clay can move into. This has been investigated by laboratory experiments and analytical calculations using simple material models [3], and more sophisticated calculations using the ABAQUS code and a general material model for MX-80 bentonite

4.2.1 Laboratory experiments

142 mm diameter granite discs with a 39 mm central hole containing MX-80 bentonite with 1900 kg/m^3 density at saturation were separated by metal foils with 100 to 1000 μm thickness and effectively confined. The discs were submerged in distilled water and examined after 2-3 months with respect to the depth to which the clay had penetrated and to the density and physical constitution of the penetrated clay [3]. The major results are summarized in Table 3. The inspection of the penetrated clay showed that there was a denser part near the bentonite core in the hole and a softer outer part with a density of 1050 to 1100 kg/m^3. Also, a rather distinct boundary between the soft part and the water was found, indicating that no spontaneous dispersion of smectite particles had taken place (Figure 15).

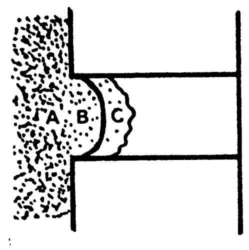

Figure 15. *Schematic picture of the appearance of penetrated clay. A) is the core bentonite, B) fairly stiff part, and C) very soft region [3].*

Table 3. *Recorded data in 2-3 months long penetration tests [3].*

Aperture μm	**Distance of front (soft clay) from hole, mm**	**Thickness of soft clay part, mm**	**Average density, kg/m^3**
100	Around 10	5-10	-
200	Around 10	5-10	-
300	Around 10	5-10	1240
400	10-15	5-10	1150
500	15	8	1140

One finds from Table 3 that the penetration was about the same for apertures ranging between 100 and 300 μm, which is explained by the fact that the resolution was limited. A very important observation was that the extension of the soft outer part was not very different for the various apertures.

Preceding pilot tests had shown that the rate of penetration was retarded. Thus, for the aperture 150 μm the penetration in 42 days was 2 mm and remained at 2 mm also after 70 days. For the aperture 300 μm the penetration was 2 mm in 10 days and 4 mm in 70 days. For 500 μm aperture the penetration depth was 4 mm in 14 days and 6.3 mm in 70 days.

4.2.2 Intial modeling

The penetration recorded as a function of time suggested that the front of the penetrating clay moved forwards according to a log time law [3]. Assuming that the process is of Poiseuille type, i.e. with the viscosity as major physical property of the clay and no slip at the clay/fracture contact, and the swelling pressure gradient as driving force, the penetration depth can be expressed as in Eq.2.

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

where: x=Penetration depth of the denser part of the clay (meters)
A=Viscosity-related parameter (5E+4 to E+6 MPas)
d=Aperture (meters)
t=time after onset of penetration (years)

This model, which is believed to be a better approximation to the real conditions in 4th order fractures with their common chlorite coatings than a simple expansion model based on swelling retarded only by the hydraulic conductivity of the clay [3], yields the theoretical penetration depth 15 cm of the denser part of the penetrated clay after 1000 years and 20 cm after 10 000 years.

4.2.3 Recent modeling

Application of a general reological model worked out for MX-80 [15] has been made in numerical calculation of the porosity distribution in fracture-penetrating clay into a 4 mm wide slot representing a fracture with a dead end located 44 mm from the fracture opening. The calculation showed that the denser part of the penetrating clay would move into the fracture by about 20 mm in 23 days and that it would yield a rather homogeneous clay fill with a density of about 1500 kg/m^3 after 90 days. The density distribution across the slot after 23 days is clearly in support of the Poiseuille flow approach in the initial modeling. Applying the initial model to the case with a fracture aperture of 4 mm it would yield a penetration depth of about 300 mm in 10 years and about the same depth as obtained by the sophisticated numerical calculation, i.e. about 20 mm, in 23 days.

4.3 Tentative conclusions on the clay penetration issue

It can be concluded from the present study that the aperture of the majority of hydraulically active fractures intersecting deposition holes is in the interval 20-100 μm. They are commonly coated with chlorite, which is a surface-active mineral with properties similar to those of illite and mixed-layer smectitic minerals, meaning that the fracture surfaces adsorb not only cations but also clay particles and clay particle aggregates. The following findings with respect to clay penetration are pertinent:

1. The penetration depth of MX-80 clay into fractures intersecting deposition holes is normally expected to be only a few centimeters in the first 100 years and up to a decimeter after several thousand years.

2. Under special conditions, i.e. critically oriented deposition tunnels with respect to the rock stress conditions, certain fractures may expand or be neoformed with an aperture of several hundred micrometers, which would increase the penetration of clay buffer to several decimeters in a few thousand years.

5 MIGRATION OF CLAY COLLOIDS IN ROCK FRACTURES

5.1 General

It has been demonstrated that the front part of MX-80 clay penetrating into fractures will be very soft, its density being estimated at 1050 to 1100 kg/m^3. Clay particles are not spontaneously released into the water in the fracture even if it has a very low electrolyte content, which means that erosion by flowing water is required for dispersion. One therefore has to determine whether groundwater flow can produce sufficient drag forces to tear off clay from the soft front. A second major issue is to what extent clay particles moved by flowing groundwater can be caught and sorbed on fracture coatings.

5.2 Groundwater flow rate in fractures intersecting deposition holes

5.2.1 Factors

The flow rate in fractures intersecting deposition holes is controlled by the local hydraulic hydraulic gradients and the hydraulic aperture.

5.2.2 Hydraulic gradients

At complete restoration of the original piezometric conditions the local and regional hydraulic gradients are assumed to be of similar magnitude, i.e. about E-2 neglecting the influence of thermally induced pressure differences, which may have some influence in the first few hundred years. Taking the thermal effects into consideration it is believed that the local hydraulic gradient in the nearfield rock during the first 10-20 years after emplacement of buffer and canisters may be E-1 at maximum.

5.2.3 Groundwater flow rates

Using the conservative figure E-9 m/s for the bulk hydraulic conductivity of the about 200 m^3 rock cylinder with 30 m^2 base surrounding each deposition hole and assuming that it is exposed to a vertical hydraulic gradient of E-1 yielding only axial flow, one finds the flux to be 3E-9 m^3/s. Assuming that this flow takes place through *n* steep fractures each with more than 10 m length (height), *b* m width, and a hydraulic aperture of *d* µm, one finds the average flow rate to be as summarized in Table 4. The actual width of this sort of discontinuities is believed to be 5-10 m but the fact that the permeable part of the fractures is of channel type justifies lower width values and various estimates have led to the conclusion that from the point of hydraulic performance the channels have an approximate width of 1 cm and a spacing of 10-20 cm [15]. These spacings mean that a fracture with 5-10 m total width has a hydraulic width of 25-100 cm.

Table 4. *Flow rates in generalized nearfield rock with steep fractures and an average hydraulic conductivity of E-9 m/s.*

Number, *n*	Width, cm	Aperture, μm	Flow rate, m/s
2	100	20	7.5E-3
	100	50	3E-3
	100	100	1.5E-3
	25	20	3E-2
	25	50	1.2E-2
	25	100	6E-3
3	100	20	5E-3
	100	50	2E-3
	100	100	E-3
	25	20	2E-2
	25	50	8E-3
	25	100	4E-3
4	100	20	3.8E-3
	100	50	1.5E-3
	100	100	7.5E-4
	25	20	1.5E-2
	25	50	6E-3
	25	100	3E-3

The data in Table 4 suggest that the average flow rate in fractures intersecting deposition holes may be as high as a few centimeters per second for an average aperture of 20 μm, which is sufficient to erode and transport dispersed particles according to the experimental data in Tables 1 and 2. The maximum size of particle aggregates that can be transported is 10-20 μm. For the case of 4 fractures with 100 μm size and 100 cm hydraulic width one finds that the average flow rate is slightly lower than one millimeter per second, which is close to the critical figure according to the MX-80 experiments reported in Table 2. Naturally, nearfield rock with a vertical hydraulic conductivity of E-10 m/s or less strongly reduces or eliminates buffer particle transport.

For the lower hydraulic gradients that will prevail after the saturation period it is estimated that neither erosion or transport of clay particles emanating from the buffer will be very significant.

5.3 Influence of adsorption of clay on fracture surfaces

The majority of the fracture surfaces of 4^{th} order discontinuities in virgin granitic rock are coated with chlorite and micas although calcite may dominate in certain cases. Chlorite crystals have an appreciable surface charge due to vacancies in the lattice, which provides a cation exchange capacity of around 30-40 meq/100 g, and an ability to adsorb clay particles through ion-pair and hydrogen bonding.

Chlorite coatings are usually clastic due to tectonically induced shearing in orogenetically active periods and ubiquitous creep caused by deviatoric stresses in the earth crust. Fracture fillings of chlorite are permeable but the size of the numerous voids and fissures is mostly on the micrometer scale and hence smaller than the hydraulic aperture.

In summary, it is believed that physico/chemical interaction of dispersed clay particles and many fracture coatings and fillings significantly reduces the possibility of the particles to be transported by flowing water over larger distances in the nearfield rock.

6 DISCUSSION AND CONCLUSIONS

6.1 Conditions for clay particle migration

Release of clay particles aggregates from the buffer implies that the density of the part exposed to the water in the fracture is very low. This requires that the buffer clay has expanded into the fracture, which means that swelling is the most important prerequisite of buffer clay for colloid formation. The present study shows that penetration of buffer clay into fractures intersecting deposition holes will take place if the true aperture exceeds about 100 μm. The depth will, however, not exceed a few decimeters provided that no major tectonical events take place. The front part of the penetrating buffer will be very soft but spontaneous disintegration and release of clay particles will not take place even if the groundwater salinity is very low. The buffer clay will hardly move into more narrow fractures, which means that it will stay dense and offer greater resistance to erosion than when the aperture exceeds 100 μm.

For release and transport of particles from the soft front of clay penetrated into fractures, erosion by flowing water is required and the flow conditions are therefore determinants of particle migration in the rock.

6.2 Flow considerations

Bulk hydraulic conductivities exceeding about E-8 m/s may yield unacceptably strong inflow into the deposition holes in the buffer application phase (>>2 l/h) and sealing by grouting may be required [17]. In practice, the nearfield rock will therefore probably have an average bulk conductivity of E-9 m/s ore less. A deposition hole with 8 m depth and 1.75 m diameter is expected to be intersected by 1-2 steep hydraulically and mechanically active fractures, belonging to the total set of 2-4 steep fractures that are assumed to appear in the 200 m^3 nearfield rock of each hole. The situation is expected to be as shown in Figure 16, implying that the flow paths are partly formed by the intersection of differently oriented 4^{th} order fractures. It is important to realize that once migrating particles have reached the blast-disturbed zone they are expected to move easily in this zone to intersecting major fractures that can bring them further to the biosphere.

A special case is represented by holes drilled from deposition tunnels with unsuitable orientation with respect to the rock stress field. Under certain stress conditions and with a buffer with high swelling pressure, tension can form new fractures or expand existing ones, primarily along the upper part of the holes. They may be significantly wider than ordinary fractures and hence play a role in particle transport. This case can be avoided by taking the rock stress conditions and rock structure into consideration when the repository is being planned.

The disturbed zone of properly oriented TBM tunnels is not believed to contain fractures with an aperture exceeding 100 μm that are interconnected over longer distances. Hence, clay particles that have been moved up to the tunnels will be stuck in the backfill if it contains smectite as planned. It is therefore advantageous to excavate the tunnels by TBM technique compared to blasting.

6.3 Nature of migrating clay and implications on its motion

Discrete clay particles of colloidal size are not expected to remain dispersed in the groundwater. Instead, it is believed that they form aggregates and that these have a size of 10-20 μm. The present study has shown that in the intial phase after applying the buffer clay and the tunnel backfill, the flow rate in fractures intersecting deposition holes may be a few centimeters per second, which can erode the soft front of penetrated buffer clay. The maximum size of particle aggregates that can be transported is 10-20 μm.

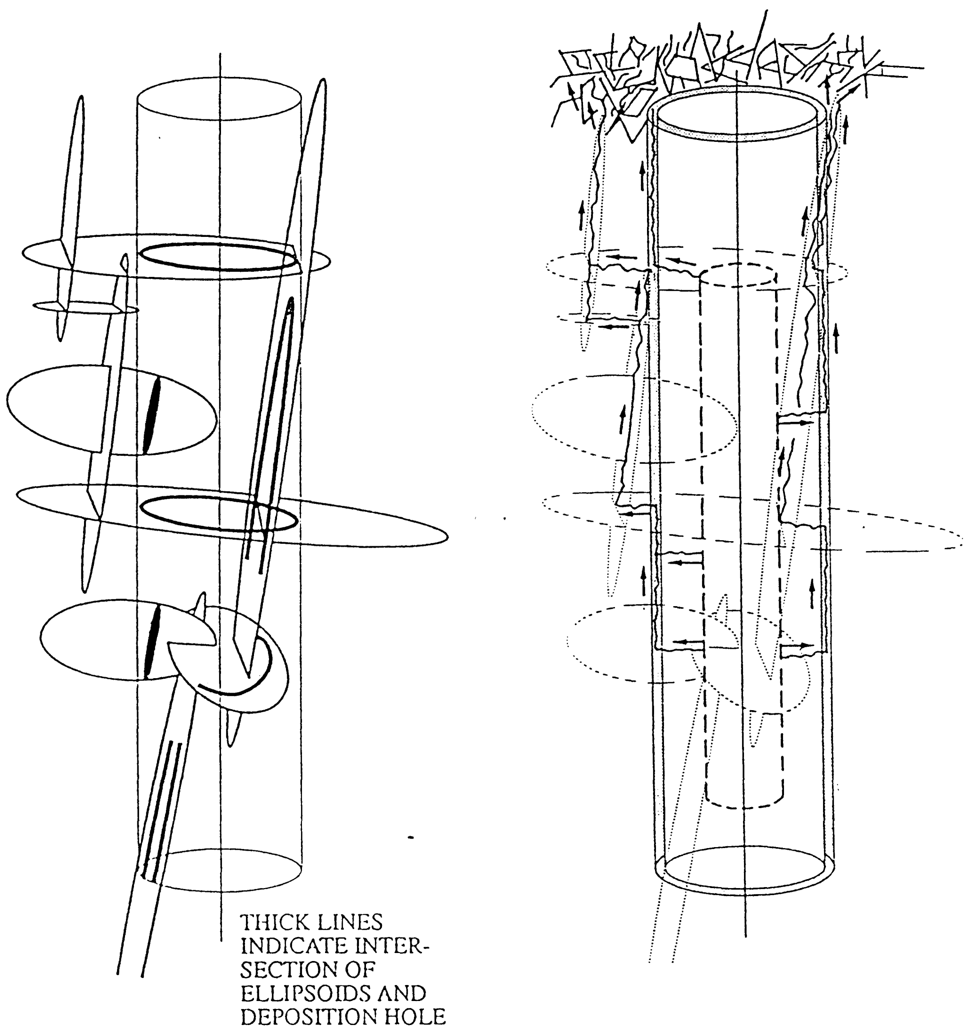

Figure 16. *Schematic picture of KBS3 deposition hole. Additional steep fractures, not shown, belong to the nearfield. Left: Generalized rock sructure. Right: Channel-type paths for water and particle transport.*

The presence of surface-charged clastic chlorite coatings and fillings in hydraulically active fractures means that smectite particle aggregates will be adsorbed and form plugs, especially at constrictions. This is believed to effectively reduce further particle transport

For the lower hydraulic gradients that will prevail after the saturation period and when the temperature gradients are largely evened out it is estimated that neither erosion or transport of clay particles emanating from the buffer will be practically important.

6.4 Mass transport capacity

The average mass and surface area of released clay particle aggregates, which typically consist of E+3 to E+6 particles are estimated at E-18 kg to E-15 kg, and E-10 to E-7 m^2, respectively. Knowing the amount of sorbed radionuclides per aggragate surface area and the number of aggregates that are released from the buffer per time unit one can estimate the transport rate of radionuclides from the nearfield. For the highest expected flow rate, i.e. about 3E-2 m/s according to Table 4, no 20 μm aggregates will pass through for geometrical reasons, but for flow rates around E-2 m/s in fractures with 50 μm aperture, one finds the mass transport along the deposition holes to the tunnel floor to be roughly 10 grams of solids in 100 years, assuming that the transported aggregates form a single continuous train. The subsequently reduced hydraulic gradient will yield slower transport of solids, i.e. probably on the order of a few tens of grams per thousand years.

If constraints like physico/chemical interaction of clay particle aggregates and fracture coatings and fillings are considered, the true solid transport of particles is believed to be smaller and the supply of radionuclides to the biosphere negligible. On the other hand, if fractures with larger aperture are formed due to to tensile failure of the nearfield rock, such supply may have to be considered.

6.5 General conclusions

The following major conclusions were drawn from the present study:

- Penetration of buffer clay into rock fractures will be negligible except when the aperture is larger than 100 μm. In such fractures the clay will move in and the front part be sufficiently soft to make erosion and particle transport possible.

- Aggregation of discrete smectite particles and flocs will take place and the aggregates will have a diameter of 10 –50 μm, with 20 μm as a probable average measure. This means that moving clay particles will be stuck where there are constrictions in the fractures. This condition and the fact that physico/chemical interaction with fracture coatings and fillings will take place is believed to cause significant reduction of clay particle transport, except in wide fractures caused by tensile failure in the nearfield rock if critical stress conditions prevail.

- The local hydraulic gradients and flow rates determine whether erosion will take place and yield practically important transport of radionuclide-bearing solids. With common rock structure models and probable hydraulic gradients this transport is not expected to be significant.

- The structure and stress conditions of the rock are most important for clay particle transport. If the tunnels from which the deposition holes extend are produced by TBM technique the disturbance will not yield fractures and fissures with sufficient interconnectivity and aperture to make particle migration possible. If the tunnels are unsuitably oriented and the stress conditions very anisotropic, and the stress level also high, fractures with substantial aperture may be formed and they can serve as particle transport paths.

- If the tunnel orientation is appropriate and no major tectonic events take place, and the holes drilled in rock with a bulk hydraulic conductivity of E-9 m/s or lower, it is concluded that migration of radionuclide-bearing clay particles emanating from the buffer is not of importance.

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