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Dense smectite clay used as overpack of deeply buried metal canisters with highly radioactive wastes

Argile dense de smectite utilisée comme tropballot pour des boîtes métalliques profondément enterrées avec des déchets extrêmement radio-actifs

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SYNOPSIS The Swedish KBS concept for the disposal of highly radioactive reactor wastes makes use of highly compacted Na bentonite as near-field isolation of canisters emplaced in boreholes in crystalline rock. The clay is applied in the form of compressed blocks of air-dry bentonite powder which take up water from the rock so that the clay eventually swells to fill the space between the rock and the canisters. The final bulk density of the completely saturated clay will be about 2 t/m^3 which yields a practically impermeable barrier.

The homogeneity of the moistening of the clay and the associated swelling are of fundamental importance for the clay/rock and clay/canister interactions. Recent field experiments serve to illustrate the involved processes.

INTRODUCTION

The Swedish multibarrier concept for the disposition of highly radioactive, unprocessed spent nuclear fuel, originally termed KBS 2, implies that copper canisters containing radioactive material be surrounded by dense Na bentonite clay (KBS, 1980). The repositories typically consist of tunnel systems at a minimum depth of 500 m in crystalline rock, the canisters being located in deposition holes with about 6 m spacing, drilled from the tunnel floor. A more recent, similar concept, KBS 3, has formed the basis for fuelling permission for the two latest Swedish reactors.

The clay is applied in the form of well fitting blocks of highly compacted, granulated bentonite powder. They are not water-saturated to start with and take up water from the surrounding rock which makes them swell and form a tight contact with the rock and the canisters. After saturation the clay will ultimately be in equilibrium with the surroundings with respect to the effective stresses as well as to the pore pressure. It then forms a medium with a very low hydraulic conductivity and a low ion diffusivity. In addition to these valuable properties the dense bentonite is also self-healing in the sense that voids or local passages in the clay, caused by possible slight rock or canister displacements, will be sealed by the swelling power of the clay.

Early theoretical predictions based on laboratory experiments suggested that the rate and uniformity of the water uptake by the compacted bentonite from the rock should be largely determined by the distribution and aperture of the joints and fractures exposed in the rock walls of the deposition holes. This has been tested in a large scale field test in Stripa, Sweden, which shows the importance of the joint-sealing capacity of the swelling clay, as well as of the temperature gradient for the water migration.

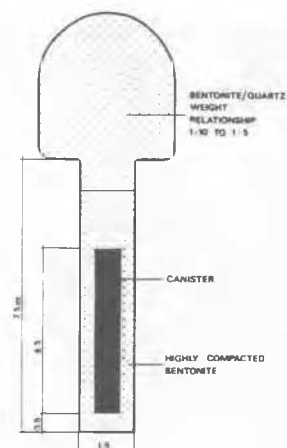


Fig. 1. Cross section of repository tunnel.

The blocks can be prepared on an industrial scale by applying cold isostatic compaction technique with pressures in the range of 50-110 MPa.

The bentonite used in the experiments referred to is the commercial Volclay MX-80, which has a clay content ($<2\mu\text{m}$) of approximately 85 % and a montmorillonite content of 80-90 % of this fraction. The specific surface area is about 700 m^2 .

WATER UPTAKE AND SWELLING

Mechanisms of hydration

Smectite minerals have a strong affinity to water which is primarily due to hydrogen bonding and van der Waals effects, and to the

hydration of adsorbed cations. The latter effect is less important in Li- and Na-saturated smectite than when Ca or Mg occupies the exchange sites. In the latter case, hydration leads to very moderate expansion of a sample that is free to swell. Na smectite takes up water and swells to several times its volume in water of very low salinity.

The bentonite blocks, which suitably have a bulk density of about 2.1 t/m^3 and a water content of 8-14 % depending on the humidity of the air, have a degree of saturation that is of the order of 60 %. Their microstructure is characterized by a domain-type arrangement of the clay platelets, which tends to be increasingly homogeneous in the course of the saturation process (Fig. 2). At a constant dry density of this magnitude the face-to-face grouping is largely preserved in the process, while expansion to a higher water content than about 50 % produces considerable microstructural rearrangement leading to a very open edge-to-face aggregated state after strong swelling. At very low salinities spontaneous dispersion may even take place.

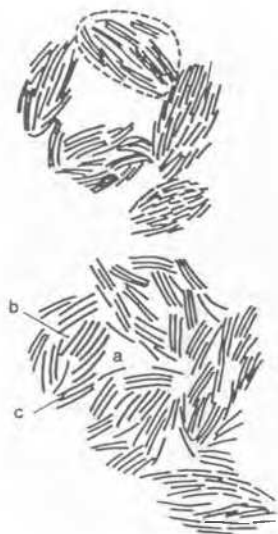


Fig. 2. Microstructure of highly compacted Na bentonite. Upper picture represents the freshly compacted, non-saturated clay. Central picture illustrates the saturated, matured state, while the lower picture represents a strongly expanded clay gel. a is large void ($\geq 10 \mu\text{m}$), b is small void ($\leq 10 \mu\text{m}$), c is interlamellar space.

PHYSICAL REACTIONS

Swelling pressure

Since the blocks are confined in the deposition holes the expansion of the clay is very limited, the major swelling being in the vertical direction in which the overlying on site-compacted sand/bentonite backfill can be displaced and compressed. These effects are expected as a consequence of the swelling pressure exerted by the dense bentonite. This

pressure, which has been found to be practically independent of the chemical composition and salt concentration of the pore water at higher bulk densities than about 2 t/m^3 in a fully water-saturated state, is approximately 10-50 MPa if the bulk density ranges between 2.00 and 2.15 t/m^3 . The corresponding hydraulic conductivity interval at 25-75°C is $5 \cdot 10^{-14}$ to $5 \cdot 10^{-13} \text{ m/s}$ at hydraulic gradients of 10^3 to 10^4 , while the diffusion coefficients of relevant radionuclides is $2 \cdot 10^{-13}$ to $2 \cdot 10^{-11} \text{ m}^2/\text{s}$ (Pusch, 1982). Some of these laboratory-derived relationships, which are also theoretically accounted for, were tested in the large scale field test as shown later in this article.

Uniformity of water uptake

The question of how uniformly water enters the bentonite annulus is of profound importance since a non-uniform wetting may produce a large variation in swelling pressure over the canister surface, leading to bending moments and tensile fractures in the metal. Also, a variation in contact pressure at the clay/rock interface may be produced, by which the heat transfer from the hot canisters to the rock could be insufficient.

Laboratory tests with uniaxial water uptake in confined, cylindrical air-dry bentonite samples have shown that this process can be described as one of diffusion and applying the derived diffusion coefficient ($5 \cdot 10^{-10} \text{ m}^2/\text{s}$), the rate and distribution of the water uptake was calculated using FEM technique and the assumption that water is available only from clearly identified, water-bearing rock joints or fractures (Börgesson, 1982). Fig. 3 illustrates the predicted water content distribution for a particular case.

Thermally induced water migration from the vicinity of the hot canisters towards the confining rock was assumed to be relatively unimportant in the first, preliminary estimation of the uptake and redistribution of pore water in the bentonite. The field test shows, however, that this process is of major importance.

Temperature distribution

Available experimental and literature thermal data yield a maximum canister surface temperature of about 80°C and a temperature gradient of slightly more than 1°C per cm in the radial direction at mid-height of the canisters. Approximately the same temperature distributions are arrived at if the thermal conductivity $0.75 \text{ W/m}^\circ\text{C}$ of the freshly compacted non-saturated bentonite is applied as if the actual moisture-dependence of the conductivity is considered. This is explained by the fact that the drop in thermal conductivity in the hot zone close to the canisters is compensated by an increased conductivity in the wetted, peripheral part of the bentonite annulus.

THE STRIPA FIELD TEST

Test arrangement

A large underground test station was estab-

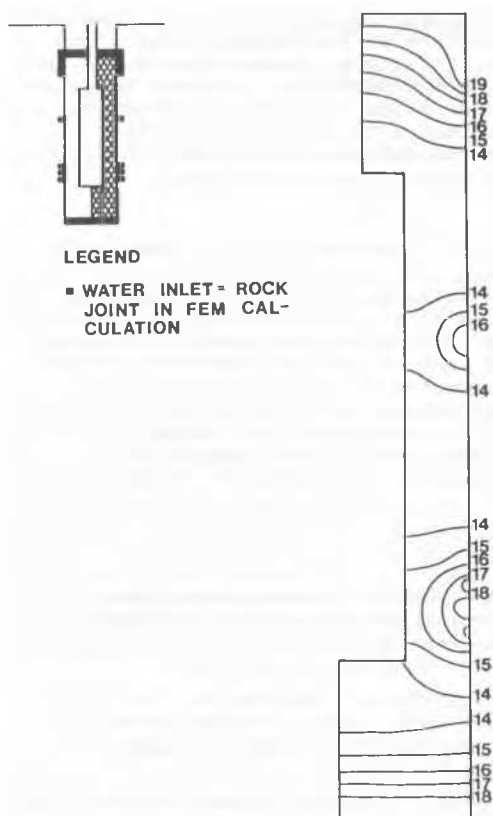


Fig. 3. Predicted distribution of the water content for a simple case. Time after onset of water uptake is 64 weeks.

lished in 1976 in an old abandoned iron ore mine at Stripa, about 250 km west of Stockholm. One of the drifts at 360 m depth has been used since 1981 for the international so-called Buffer Mass Test, which is an almost half scale application of the KBS concept (Pusch et al. 1983). The heat production of the canisters is simulated by electrical heating of six 600 to 1800 W-powered metal cylinders with a diameter of 38 cm and a length of 150 cm. The 76 cm diameter "deposition" holes, which have a depth of about 300 cm and 6 m spacing, were made by core drilling in rock with a largely varying degree of fracturing. The six holes therefore represent quite different hydrological conditions that yield a spectrum of water uptake rates in the bentonite, which₃ had an original bulk density of about 2.1 t/m³ and a water content of 10-13 %. The wettest hole (termed no 2) thus illustrates a saturation state after 3 years that will not be arrived at until 10-20 years in an actual repository where much less fractured rock will be required. The driest hole (no 6), on the other hand, probably yielded a slightly slower saturation than will be met with in practice. Saturation, swelling, and temperature conditions in these two holes will be discussed here with reference to the predictions. The power of the two heaters was 600 W throughout the test period.

Water uptake

The bentonite annuli were equipped with a large number of moisture sensors for continuous determination of the water uptake. This recording and a very comprehensive sampling in connection with the excavation of four holes (no 3-6) after more than 2 years test time, show that two mechanisms dominate:

1. The temperature gradient drives pore water from the vicinity of the "canisters" towards the rock. Thereby, the 1-3 cm peripheral zone becomes fully saturated, swells and forms a tight contact with the rock.
2. Water initially migrates from discrete water-bearing rock joints and fractures but they soon become sealed by penetrating clay if their apertures are wider than about 0.1 mm. Water under pressure in the rock is consequently forced through less wide fractures, which, in turn, tend to be sealed. Ultimately, water seems to migrate through the crystal matrix along fissures and incomplete grain boundaries and this yields a water migration from the rock into the clay which is remarkably uniform over the entire interface.

After more than 2 years test time the degree of water saturation in the "wet", fracture-

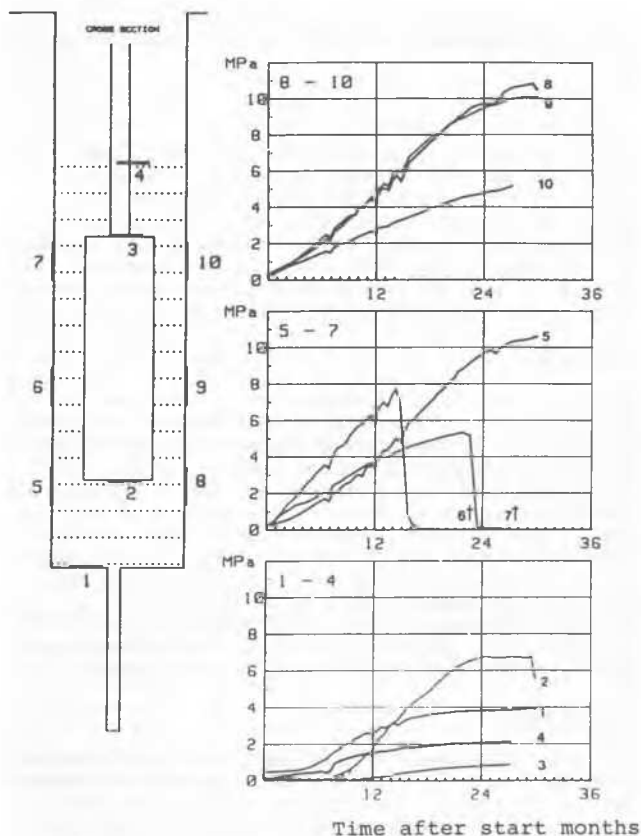


Fig. 4. Computer-plotted swelling pressures in the "wet" heater hole no 2. Cross means failed sensor.

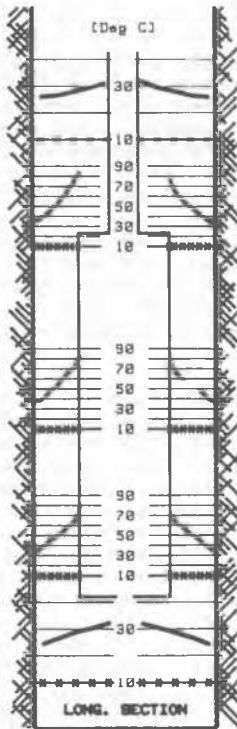


Fig. 5. Computer-plotted temperature distribution in the "dry" heater hole no 6. x denotes the location of thermocouples.

rich hole no 2 is about 90 %, as concluded from the moisture sensor signals, the outer 10 cm of the 19 cm thick bentonite annulus being completely saturated. At the excavation and sampling of the "dry", fracture-poor hole no 6 it was found that only the peripheral 3 cm zone was water-saturated, the water content being slightly higher than 20 %.

Swelling

The high degree of saturation in hole no 2 suggests that the swelling and therefore also the swelling pressures should be almost fully developed. With the actual density, the predicted swelling pressure at the clay/rock interface is about 10 MPa. As indicated by Fig. 4 the Gloetzl pressure cells located at this interface yield almost exactly this pressure. In the dry hole no 6 the saturation of the thin peripheral zone also produces a swelling pressure (about 0.5 MPa) but the deformability of the inner, non-saturated core prevents the build-up of higher pressures.

Temperature

All the holes were richly equipped with thermocouples through which the development of temperature fields in the clay and the surrounding rock has been continuously recorded. The heat distribution in the clay reached an almost steady state a few months after the test start. The temperature has then dropped slowly and slightly due to the water uptake, which increa-

ses the heat conductivity of the clay. The predicted temperature at the heater surface and the clay/rock interface, respectively, are in very good agreement with the recorded temperatures. The conditions in hole no 6 shortly before the termination of this test after slightly more than 2 years, is shown in Fig. 5.

CONCLUSIONS

The theoretically derived physical models for the development of swelling pressures and temperature fields were nicely confirmed by the field test. As to the distribution of the water uptake in the bentonite, the field test unanimously shows that it is much more uniform than the early conservative model predicts. The rate of water uptake in the practically important case of fracture-poor rock seems to be slower than predicted by the simple model based on diffusion from discrete, wider fractures.

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