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## **Migration experiments in Studsvik**

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## MIGRATION EXPERIMENTS IN STUDSVIK

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## ABSTRACT

Field experiments (two-well pulse method) on the migration of Sr and Cs in a gneiss at Studsvik on the Swedish east coast as well as supporting laboratory measurements on sorption/migration have been performed.

Interconnected fractures at about 100 m depth between boreholes were located and characterized. In the boreholes measurements of resistivity, spontaneous potential, pH, Eh, curvature, temperature, gamma radiation, and radon have been performed. The fracture zones were localized, using TV-logging, and the mineralogy of the water bearing fractures in a drill core from the area determined. The main fissure filling minerals were chlorite, calcite and smectite.

The groundwater composition was analyzed and it was found that the water in some cases is oversaturated with respect to calcium carbonate. Temperatures up to 12 °C of the pumped water have been observed, corresponding to water originating from 200-400 m depth.

In laboratory batch measurements it was found that the distribution coefficient for Studsvik gneiss and possible fissure filling minerals was 0.01-0.4 m<sup>3</sup>/kg. The sorption isotherm for the gneiss was almost linear, a Freundlich isotherm with the exponent 0.89 fits well to the data.

In two field experiments, performed between different boreholes, retention factors for Sr of 17 and 30, respectively, were obtained. In laboratory column experiments, performed on crushed borehole material, the retention factor was 30-35, corresponding to a distribution coefficient of 0.006-0.008 m<sup>3</sup>/kg.

For Cs, injected simultaneously with the Sr, the retention-factor is > 30. (No breakthrough after 5 000 h.)

In both the field experiments a minor amount of the Sr was migrating almost without retention (retention factor < 2). It does not seem probable that there should be an extremely short flow path, transporting minor amounts of the water in both the experiments, explaining the fast migration, as different boreholes were used. A more probable explanation is that some of the Sr is sorbed on particles or complexed by organics in the water and transported in one of these forms. A precipitation of Fe is for instance observed when aereating the groundwater, indicating the possibility of Fe-colloid formation. This effect was not observed for Cs, indicating that the process causing the fast transport of Sr is selective to Sr.

## 1 INTRODUCTION

The main objectives with the present work have been studies in fractured crystalline rocks ("in situ") of radionuclide migration and to compare the results with those from laboratory sorption experiments on rocks and minerals from the field test area. Different KBS-studies have been involved and coordinated, e.g. sorption studies at CTH, fracture mineral investigations at SGAB-SGU, Gothenburg, hydrological investigations at SGAB, Uppsala, and field migration experiments at STUDSVIK.

Very few in situ migration experiments in fractured crystalline rocks have so far been reported. One study was performed in another part of the Studsvik area in 1977-78 (Landström et al, 1978). In France a series of migration experiments have been performed in a granite (Calmels et al, 1979, Goblet et al, 1979, Goblet et al 1981) and the results have been interpreted in terms of a surface distribution coefficient and sorption/desorption kinetics. The distribution coefficient was found to be one order of magnitude larger for Sr than for  $\text{Na}^+$  (only  $\text{NO}_3^-$ ,  $\text{Cl}^-$ , and  $\text{I}^-$  were observed to be "good tracers" i.e. non-sorbing). A very low retention (retention factor about 1.2) was observed for Sr. Migration studies in the Climax Stock granite in Nevada are planned and site characterization is going on (Isherwood et al, 1982). Migration experiments with stable non-sorbing and sorbing (Sr and Cs) tracers have been carried out in the Finnsjö area as part of KBS hydrogeological project (Gustafsson and Klockars, in progress).

The present field experiments have been performed in a new test site at Studsvik, situated about 90 km S of Stockholm (Figure 1). The work was initiated by Prav (Swedish National Council for Radioactive Waste) but is now carried out on commission from KBS.

## 2 PREPARATION AND CHARACTERIZATION OF THE FIELD TEST SITE

Geologic and tectonic mapping of the bedrock and geophysical measurements from the ground surface (refraction-seismic, resistivity and electromagnetic) were carried out. Based on the results from these investigations 8 percussion-drilled holes (B1N-B8N, Figure 1) and one core-drilled hole (K1N, Figure 1) were drilled. The core-drilled hole K1N (diameter 56 mm and length 200 m) was drilled at an angle of  $60^{\circ}$  in towards the selected test area. The hammer-drilled holes B1N-B8N (diameter 115 mm and lengths around 100 m) were drilled at an angle of  $80^{\circ}$  (except B2N and B3N for which the angle was  $70^{\circ}$ ).

The following borehole measurements were carried out:

- Resistivity
- Spontaneous potential
- pH of the borehole liquid
- Eh of the borehole liquid
- Curvature measurements
- Temperature
- Natural gamma radiation
- Radon measurements
- TV-logging for identification of fractures and their orientation

In order to determine hydraulic conductivities around and between the boreholes and to find suitable flow paths for the migration experiments, water injection tests in single holes and interference tests were carried out. Based on the results of the hydrogeological tests in the boreholes, B6N was chosen as pump hole and B1N, B5N and B8N as injection holes for the migration studies (Figure 2). The hydraulic properties of the selected flow paths and water transit times were determined using a two-well pulse method with I-131 as non-sorbing tracer.

Details and results of the preparatory works are reported elsewhere (Klockars et al, 1980, Landström et al, 1980, Klockars et al 1982). The results are summarized in the following with addition of new results from later investigations.

## 2.1 Geology

The bedrock is well exposed, mainly in long strip outcrops running in the NNW direction (Figures 1 and 7). Intervening low-lying areas are covered with soil, mostly banks of gravel containing some sands. The thickness of the soil cover at the drill hole positions varied between 0.3 to 2.5 m. The bedrock in the region belongs to the Svecokarelian belt of Sweden (1800-2000 Ma). The rock type present at the test site is a gneiss of sedimentary origin, which in parts is strongly migmatized and veined. Also thin sheets and layers of metabasite occur. Just to the west of the test site is a synorogenic, migmatized granodiorite outcropping (Lundström, 1976).

The core from borehole K1N has been mapped considering rock types, fractures and fracture



fillings (Klockars et al, 1980, Tullborg and Larsson, 1982). Three types of rocks have been distinguished (Figure 3):

- Veined gneiss (the most common type) which by Lundström (1976) is characterized as a very heterogeneous rock with biotite-foliated paleosomes. Typical minerals in the paleosomes are quartz (40-70 vol %), feldspars (10-40 vol %) and biotite as well as chlorite. Andalusite, sillimanite, garnet and cordierite are minerals which usually constitute up to 20 vol % of the paleosomes. Garnet as well as cordierite is also present in the neosomes.
- Granite-pegmatite which mostly represents the leucocratic material of migmatites with a granitic composition and with aplitic to pegmatitic textures.
- Metabasite-metadiorite, dominated by plagioclase, hornblende and biotite.

For the laboratory sorption tests drill cuttings were sampled from 3 m sections down to the 76 m level of borehole B8N. Seven of these sections were analyzed for main, minor and trace elements (Lundström, 1980). Average values of the seven samples are shown in Table 1 and are considered as representative for the bulk composition of the bedrock in the test area.

## 2.2 Fissure filling minerals

Sampling and characterization of representative fissure fillings is important for the supporting laboratory sorption experiments and for

interpretation of in situ experimental data. Unfortunately the in situ test boreholes were not core-drilled. Thus the investigated fissure fillings are from the core drilled nearby hole K1N. Crushed zones of the K1N core are marked in Figure 3.

Fissure fillings were sampled at levels distributed along the whole core length (marked in Table 2). The filling material was studied by microscope and/or by XRD. Instrumental neutron activation techniques was applied to some samples for elemental analysis and fissure materials from interesting zones were selected for laboratory sorption studies.

Open as well as sealed fissures were considered. Most of the fissures observed in the core are coated and open. Many of these have, however, probably been opened during the drilling operation. This suspicion is strengthened by the fact that rough and fresh uncoated fissures are frequent in the drill core.

Originally open fissures (i.e. water-transmitting) can in favourable cases be localized by geophysical and geochemical borehole logs, as exemplified by radon, temperature and resistivity logs in the percussion-drilled holes (Landström, 1980). Different logs in K1N thus indicated water-transmitting fissures around 104 m. Materials from these fissures (which probably are representative for the sorption media in the tracer tests) have been subject to more detailed investigations.

Most coated fissures are thin (usually less than 1 mm) and mostly with a coating of chlorite and calcite. By careful investigations of the coatings it was found that also clay minerals

appear. XRD studies (samples treated with ethylenglycol) made clear that most of the clay minerals were of a swelling type (smectite). The structure formula calculated from analysis (carried out by Joëlle Duplay, Strassbourg) of three mineral grains were

1.  $(\text{Si}_{3.42} \text{Al}_{0.58}) (\text{Al}_{0.135} \text{Fe}_{1.24} \text{Mg}_{1.88}) \text{K}_{0.055} \text{O}_{10} (\text{OH})_2$
2.  $(\text{Si}_{3.55} \text{Al}_{0.34} \text{Fe}_{0.11}) (\text{Fe}_{1.34} \text{Mg}_{1.21}) \text{O}_{10} (\text{OH})_2$
3.  $(\text{Si}_{3.515} \text{Al}_{0.485}) (\text{Al}_{0.045} \text{Fe}_{1.27} \text{Mg}_{1.27}) \text{O}_{10} (\text{OH})_2$

This gives a composition intermediate between saponite and nontronite. The look of this smectite is nearly black and velvet like. The black color could be due to a mixing with organic material. Preliminary results from neutron activation analysis of a smectite-rich fissure filling at 104.2 m level are shown in Table 3 (mean values of two samples, weighing about 20 mg each and which were scraped from the fissure surface with a plastic knife).

The frequency diagram in Figure 4 is based on core mapping of fissure minerals in the interval 50 to 125 metres. As seen from the figure, smectite is a main fissure filling mineral together with calcite and chlorite. Other filling minerals observed are feldspar, pyrite, palygorskite and some mica.

A comparison between open and sealed fissures has been omitted due to the difficulties in defining whether a fracture originally was open or sealed in situ. The fact that clay mineral

coating is common makes the fracturing very easy during the drilling operations. It is, however, quite evident that feldspar is most frequent in the sealed fissures. In contrast to this palygorskite has only been observed within open fractures.

It has not been possible to distinguish different mineral generations of the filling material. At the 104.2 metre level, which possibly is a fissure with fillings of similar kind as in the test flow paths, the filling material is smectite, calcite and quartz as registered on the XRD (Figure 5). However, thin section studies did not show the presence of quartz and the XRD sample may have been contaminated with quartz from the underlying rock during the sampling. The filling is dominated by smectite (Figure 5). When treated with ethylenglycol a small content of chlorite was identified together with smectite and calcite. Only one generation smectite and calcite is verified.

As smectite as well as calcite can be deposited at low temperatures and pressures a recent age for these minerals can not be excluded. Some of the analyzed groundwater samples from the area show saturation in respect of calcite. To find out if smectite is a stable mineral phase within the present water environment, data of sampled groundwater from borehole B6N have been plotted in a Helgesson (1971) stability diagram. This shows that most of the water samples allow kaolinite as a stable mineral phase. However, the montmorillonite (smectite) field is very close to the plots representing the water analyses (Figure 6). The water also plots within the quartz saturation field.

### 2.3 Fracture tectonics

Fractures have been mapped in outcrops (Figure 7) and in boreholes by TV-logging (Klockars et al, 1980). The directions of fractures observed in outcrops correspond to the regional picture and give a clear maximum in the north-westerly direction (Figure 8).

A total of 310 fractures were oriented by TV-logging. Their strike and dip are shown by the stereographic point diagram in Figure 9 and by the histogram in Figure 10. A large majority of the fractures, 63 % dip towards the north and most of these have a strike of between N and N40W. The fractures dipping towards the south are concentrated between N20E and N50E.

In Figures 11-13 are shown stereographic projections (the intersection of the planes with the lower hemisphere) of the following sections with fractures which are utilized in the migration experiments:

B6N: 41-45 m, 47-50 m and 62-68 m  
B1N: 88-94 m  
B5N: 78-80 m

### 2.4 Hydraulic properties of the flow paths

The hydraulic properties of the selected flow paths, (B1N-B6N, B5N-B6N and B8N-B6N, respectively, cf Figure 2) and water transit times were determined using a two-well pulse method with I-131 as non-sorbing tracer. The pumping rate in B6N was  $1.2 \text{ l, min}^{-1}$ . About 370 MBq of I-131 in 0.5 l of groundwater was injected during 1 hour. Prior to and after the tracer injection, groundwater was pumped down into the injection

zone with a rate of  $0.01 \text{ l min}^{-1}$  in order to develop a steady state flow into the fractures. In both the pumped groundwater and in the tracer solution NaI was added (0.08 M) as a carrier.

In Table 4 the location and length of the flow paths B1N-B6N and B5N-B6N are shown (the flow path B8N-B6N was discarded because of unsatisfactory hydrodynamic properties). The number of fractures and their strike and dip are found in Table 5.

Water samples for monitoring tracer concentration were taken from pumphole B6N and in Figures 14-15 are shown the experimental breakthrough curves for I-131 in the flow paths B1N - B6N and B5N - B6N, respectively. By fitting breakthrough curves according to the method described by Zuber (1974) and Lenda and Zuber (1970) the mean transit time  $t_0$  and the dispersion parameter  $D/v$  is obtained. Values of these parameters are marked in Figures 14-15. The TV-log showed 4 fractures in the injection section 91.0 - 92.3 of borehole B1N and the three partial curves in Figure 14 are interpreted as representing different flow paths.

In Table 6 are shown obtained values of the following parameters

- Hydraulic conductivity of rock mass,  $K_p$  (from injection test of 2 metre sections).
- Hydraulic conductivity of the flow paths,  $K_e$  (calculated from data of the I-131 tests).

- Kinematic porosity,  $\theta_k$ , defined as the ratio between the measured hydraulic conductivity  $K_p$  and the fracture conductivity  $K_e$ .

## 2.5 Ground water chemistry and temperature

Chemical analysis of pumped water from borehole B6N has been made at regular intervals throughout the experiments in order to check possible changes in the chemical environment. In Table 7 are shown typical values of the water composition during the non-sorbing tracer tests. The analyses "80-12-10" show the effect of a packer accident which caused an inflow of surface water (with a lower mineral content) to the flow paths. As is clear from the analyses 80-12-29 and 81-02-24 the previous equilibrium values are fairly soon recovered. The ground water composition has remained largely the same during the Sr-85 and Cs-134 migration tests.

In Table 8 are shown the concentration of certain trace elements in pumped water from B6N. They were determined with a neutron activation technique which was developed for the analysis of stable tracers, e.g. rare earth elements (Landström, in progress).

Measurements of  $E_h$  in pumped water in connection to the first Sr experiment gave  $E_h = -104$  mV, i.e. reducing conditions. The groundwater varies with time in both composition and temperature. Some of the varying parameters may affect the sorption/migration of Sr (Cf Andersson, 1983).

- pH - the distribution coefficient is somewhat pH-dependent, probably due to

a pH-dependency in the surface charge of the minerals.

- Ca-concentration - Ca is chemically similar to Sr and may be expected to compete for sorption sites.
- HCO<sub>3</sub><sup>-</sup> - the water is on some occasions oversaturated with respect to carbonate (at atmospheric pressure) and a precipitation of calcium carbonate may occur (that this is the case is obvious from the presence of calcite precipitates in water bearing fractures in a calcite free rock). An increase in temperature may cause a precipitation of calcite or aragonite. Co-precipitation of Sr may occur in connection with this.
- Fe (tot) - at high Fe-concentration a precipitation of Fe-hydroxides may occur when aerating the water. This leads to a formation of colloidal particles, which may act as carriers for Sr.

The content of organic carbon is probably of minor importance as no change in Sr-sorption has been observed in batch experiments when adding humic acid.

The temperature of the pumped water has been fairly high, about 12 °C, during the migration experiments. Lower temperatures were, however, observed in connection to packer accidents and other occasions with an inflow of surface water of lower temperature. As the temperature gradient in the area is 12-13 °C km<sup>-1</sup> and a temperature



of about 7.4 °C was measured at 100 m level the pumped water (or part of it) originates from levels of the order of 200-400 m. Figure 16 which shows temperature-depth profiles in B5N before and during pumping clearly shows the inflow of warmer water from deeper levels. The observed temperature differences are treated elsewhere (Landström et al, in progress).

### 3 Sr AND Cs IN GROUNDWATER/ROCK SYSTEMS - CHEMISTRY AND SORPTION

The only valence state of Sr in aqueous solutions is Sr (II) (Bailar et al, 1973) and among the anions usually found in plutonic rock groundwaters, only carbonate and hydroxide are forming Sr-complexes of any importance (Sillén et al 1964). At trace concentrations of Sr ( $10^{-8}$  M) the solubilities of  $\text{SrCO}_3$  and  $\text{Sr(OH)}_2$  are high enough to ensure that no precipitation of Sr will occur ( $\log K_s (\text{SrCO}_3) = -9.03$ ,  $\log K_s (\text{Sr(OH)}_2) = -3.2$ ). A formation of  $\text{SrCO}_3$ -complexes is however occurring. At pH above 7.8 and 200 ppm carbonate in the water more than 1 % of the Sr will exist as carbonate complexes and at pH 9 about 15 % will be complexed ( $\log K = 3$  estimated value, cf Torstenfelt et al, 1981).

Sorption of Sr on rocks and minerals has been studied by many investigators (Andersson, 1983). Some of the important parameters for Sr-sorption are:

- pH -  $K_d$  is usually increasing with increasing pH.
- Ionic strength -  $K_d$  decreases rapidly with increasing ionic strength.

- Cation exchange capacity (CEC) of solid
  - a good correlation between  $K_d$  and CEC is observed (cf also. Andersson et al, 1982).

Some other parameters that often are studied in sorption measurements are:

- Nuclide concentration - for Sr the sorption isotherms usually are close to linear at low concentrations (up to  $10^{-4}$  -  $10^{-3}$  M).
- Temperature has a small influence - a change from 20 °C to 70 °C gives an increase in  $K_d$  of about a factor of 2. For systems where calcium carbonate is precipitated as a fracture filling mineral, temperature may be important as coprecipitation (inclusion) of Sr with the  $\text{CaCO}_3(\text{s})$  may occur.
- The liquid/solid ratio is of little importance.
- Contact time is of minor importance at least for times over one day. Minor changes in  $K_d$  that are observed in laboratory measurements may often be attributed to changes in the mineral.
- Redox potential has no importance for the Sr-chemistry as Sr only has one valence state, but also here changes in the mineral may be caused by Eh-changes.

Cesium in water solutions has only one valence state (+I) and Cs-migration is thus not sensitive to the redox potential. Among the

complex formers found in natural waters no one will form any strong Cs-complexes. A migration/sorption of  $\text{Cs}^+$  will thus take place in a groundwater rock system. The parameters that may be of importance for migration are pH, ionic strength, competing ions etc (cf Andersson, 1983).

#### 4 FIELD MIGRATION EXPERIMENTS

Migration experiments with Sr have been carried out in the flow paths B1N - B6N and B5N - B6N, respectively. The latter experiment which is still going also includes Cs.

The injections were carried out in mainly the same way as in the flow mapping experiments with non-sorbing tracers. The tracer injection were preceded by about one week's injection of groundwater (10 ml/min) in order to develop a steady state flow into the fissures. 0.5 lit of tracer solution was then injected with the same flow rate. After the tracer pulse, groundwater was continuously injected (in B1N during 3 months and in B5N during 6 months) to ensure the entrance of the tracers into the transport channels of the fracture zone. Aereated groundwater was used in the B1N - B6N experiment. In the B5N - B6N experiment, however, pumped water from B6N was recirculated in a closed system (cf Figure 17). Tracer injection data for the two experiments are shown in Table 9. H-3 and I-131 were used as reference tracers. Water samples were taken from the pumphole B6N with the aid of an automatic sampler and analyses for the tracers were performed in the laboratory. I-131, Sr-85 and Cs-134 were analyzed by gamma ray spectrometry and H-3 using a liquid scintillation counter.

The results are shown in Figures 18 a and b, where the H-3 and Sr-85 breakthrough curves are compared in the two experiments. The tracer concentration  $C$  in the sampled water from B6N is related to the concentration  $C_0$  of the injected 0.5 l tracer solution. The I-131 curve (not shown in the figure) was in both experiments almost identical with that of H-3. In the experiment B1N-B6N NaI carriers were added but in the experiment B5N-B6N I-131 was injected without the addition of carriers.

The retention factor for the major part of the Sr-85 in experiment B1N-B6N is about 17 but a minor amount of Sr seems to migrate with the same rate as H-3. Also in the experiment B5N-B6N two peaks occur, the first one slightly delayed relative to the H-3 peak (retention factor about 1.8) and the second one showing a retention factor of the order of 30.

Cs-134 has not been detected (up to 5 000 hours after injection). The detection limit for Cs-134 is  $C/C_0 < 10^{-10}$ . The retention factor for Cs is thus  $> 30$  and no migration of Cs with the velocity of the water has been observed. From laboratory batch sorption data, retention factors  $> 1\ 000$  may be expected for Cs.

## 5 LABORATORY MEASUREMENTS

### 5.1 Experimental

Sorption measurements have been performed using both static (batch) and dynamic (column) methods. In the batch experiments the dependence of pH, nuclide concentration and time have been studied, while in the column experiments only

the transport of a radionuclide pulse in a rock column has been investigated.

## 5.2 Batch method

In the batch experiments, 45 ml of the water phase was contacted with 0.9 g of the crushed solid in a glass vial. The crushed solid was first washed and then allowed to pre-equilibrate with the water for one week. After the water had been changed and pH adjusted, inactive carrier and 0.1 ml of the radionuclide stock solution was added. As reference, vials without solid but otherwise treated in the same way as the solid-containing ones were prepared. After 1 day, 1 week and in some cases longer contact times the vials were centrifuged (5000 g) for 1 h and 1 ml samples of the water were counted for radioactivity using a NaI well-type detector. The samples were afterwards poured back into the vials and pH was measured in each vials after every sampling. The distribution coefficient  $K_d$  was calculated from

$$K_d = \frac{A_o - A}{A} \cdot \frac{V}{m} \quad (\text{m}^3/\text{kg})$$

where  $A_o$  = activity of reference sample (cpm)  
 $A$  = " of sample (cpm)  
 $V$  = volume liquid ( $\text{m}^3$ )  
 $m$  = mass of solid (kg)

Some characteristic data for the batch experiments:

- Solid phases: Crushed Studsvik gneiss from borehole B8N (unweathered and weathered in autoclave, 150 °C, 1 week), fissure filling minerals (from fissure surfaces or from other mineral samples) and rock-forming minerals.
- Liquid phases: Natural as well as artificial Studsvik groundwater and synthetic groundwater of "typical" Swedish composition. cf Table 10.
- Particle size: 0.090 - 0.120 mm.
- Contact time: 1 day, 1 week, 3 months, 6 months.
- pH: varied between pH 6 and 9.5.
- Radionuclide: Sr-85, carrier free.
- Concentration: Trace ( $10^{-8}$  M) and varied by adding inactive  $\text{SrCl}_2$  as a carrier to  $10^{-7}$ ,  $10^{-6}$ ,  $10^{-5}$  and  $10^{-4}$  M initial concentration.
- Additives: 10 ppm humic acid (in one experiment for a granite).

### 5.3 Column method

For the column retention studies standard equipment for HPLC was used: Stainless steel column, pump and injection valve. The sampling was performed using an automatic fraction collector. Groundwater was pumped through the

column and a pulse of the tracer injected. Non-sorbing tracers were used to determine the dead volume of the system.

Some characteristic data for the column experiments:

Solid phase: Crushed Studsvik gneiss mixture, sieved (0.090 0.125 mm).

Liquid phase: Artificial Studsvik groundwater, cf Table 10.

Column diameter: 4 mm

Column height: 130 mm

Porosity of column filling: 0.35

Flow rate: 0.01 ml/min.

Injected tracers: 50 l containing 1.5 kBq carrier free I-131, Na-22 or Sr-85.

Temperature: Ambient.

The column experiment showed that under laboratory conditions a considerable retention of Sr is obtained, the retention factor (water velocity/nuclide velocity) being about 30-35, cf Figure 19. The retention factor is related to the distribution coefficient ( $K_d$ ) by

$$R = \left( 1 + K_d \cdot \rho_p \cdot \frac{1 - \epsilon}{\epsilon} \right)$$

where

$\epsilon$  = porosity of the bed. Inserting relevant data gives a distribution coefficient of 0.006 - 0.008 m<sup>3</sup>/kg.

The "non-sorbing" tracers I-131 and Na-22 have different retention, indicating that I-131 acts as a "better non-sorbing" tracer than Na-22. This effect has earlier been observed in in situ experiments (Goblet et al, 1979).

Distribution coefficients vs pH for Studsvik gneiss, measured in batch and column experiments are given in Figure 20 where a comparison with values for some fissure filling minerals is done. The increase in distribution coefficient with pH is typical for ion exchange reactions, as discussed above, but as the carbonate content of the water is high, there is also a possibility of contribution from carbonate complexation of the Sr. The pronounced difference in sorption between natural and synthetic water disappeared at longer contact times.

As is shown in Figure 21, the sorption isotherm is almost linear up to initial concentrations of 10<sup>-5</sup> M. At higher concentration there is a deviation, indicating a limited sorption capacity. The data at 10<sup>-8</sup> to 10<sup>-5</sup> M Sr may with good accuracy be fitted into a Freundlich isotherm on the following form:

$$q = 6 \cdot 10^{-3} C^{0.89}$$

where  $q$  = mol/kg solid  
 $C$  = mol/m<sup>3</sup> liquid, at equilibrium.

The time dependence of Sr-sorption on rocks and minerals usually is, as mentioned above, almost negligible, and for Studsvik gneiss this also



seems to be the case. As is shown in Figure 22, the main part of the sorption occurs within one day. For fissure filling and rock forming minerals there are usually no significant changes in  $K_d$  after 1 day's contact time. An exception is, as is shown in the figure, the zeolite stilbite, where a significant increase in  $K_d$  occurs during the studied 27 days. This may be explained by a slow diffusion of Sr into the pores of the zeolite, although Sr-transport into zeolites usually is extremely fast. All these measurements are performed for finely crushed solids.

The uptake of Sr on fissure filling surfaces has up to now not been studied to any large extent. One preliminary study of the uptake on a fissure surface, mainly containing calcite and taken from a water-bearing fissure in a drill core sample from Finnsjön (Torstenfelt et al, 1982) has been performed, cf Figure 23. Here a very high uptake was observed, compared to granite surfaces. As calcite has a very low distribution coefficient (although it may be higher on "fissure filling calcite") this is probably not the only explanation.

A recent study of Cs uptake on fissure filling surfaces (Torstenfelt et al, 1982, b) shows that diffusion of adsorbed radionuclides through a surface coating and further into the rock matrix is possible. A diffusion into the rock matrix via minerals with high sorption capacity occurs, and an apparent diffusivity into the rock of about  $10^{-13} \text{ m}^2/\text{s}$  was observed. After a contact time of three months, Cs had penetrated from a water solution, containing about  $10^{-8} \text{ M}$  ( $\text{Cs}^+$ ), through the fissure filling and into the rock. A penetration of 3-4 mm for Studsvik

gneiss with surface coating was observed. A similar behaviour of Sr may be expected, and Sr-diffusion experiments on Studsvik gneiss and other rocks with fracture fillings have been started.

Addition of 10 ppm humic acid to the water phase did not affect the Sr sorption on a Stripa granite.

Laboratory sorption measurements for Cs gives higher distribution coefficients than for Sr, cf Table 11. A column experiment under the same conditions as for Sr gave a retention factor > 33. Unfortunately, the experiment had to be interrupted due to pump failure but up to this point no trace of Cs had been observed in the water. From batch sorption data, a retention factor of 1 000 - 3 000 would be expected.

## 6 DISCUSSION

There is a considerable retention/sorption of Sr in both the field and laboratory experiments. In the two field experiments carried out in separate flow paths, retention factors of about 17 and 30, respectively, were obtained. The corresponding value for laboratory column experiments was 30-35, and the agreement is thus good.

Cs which was injected simultaneously with Sr in the second field experiment has not yet been detected (up to 5000 hours after the injection). This is also in agreement with the results of laboratory sorption studies on material from the field site. From these a retention factor of 1000 - 3000 would be expected.

The comparison between sorption data from laboratory and field experiments requires laboratory samples which as close as possible resemble the solid phase of the in situ flow paths. In this work it was decided to use two kinds of material, namely a "whole rock" sample and selected fissure fillings from the core of K1N.

The whole rock sample consist of drill cuttings from bore-hole B8N. Its mineral composition is roughly an average of a 76 metre rock profile and is thought to fairly well represent the bulk rock in the test area. As the drill cuttings were obtained in a dry form any fissure filling material in the whole rock sample is probably from sealed fissures. Of other alternative rock sources, samples from outcrops were discarded (weathered and partly contaminated with organic material) and the K1N drill core was reserved for other investigations.

Different fissure types (with specific composition of filling minerals and coatings) may build up the test flow paths, e.g.

- fissures in which water was flowing before the drilling
- fissures which were closed before drilling (i.e. contained stagnant water) but became water-transmitting by connection to other fissures through the boreholes.

It is thus difficult to predict the composition of the filling material in the flow paths. Some clues are, however, given by the frequency diagram of fissure filling minerals in the K1N

core and by the identification (with borehole logs in K1N) of fissures which at present are water-transmitting. It was shown that such fissures usually are dominated by a clay mineral, identified as smectite. Special attention has been given to fissure fillings of this kind as they probably occur in the test flow paths.

The investigated fissure filling samples were taken from the core of the K1N borehole which was drilled for purposes like this and which is very close to the flow paths. Diamond drilled test holes had, of course, been advantageous as filling material from the flow path positions then could be used (although of limited size and not necessarily representing the whole sorbing surface of the fissures involved). Moreover, borehole walls and fissures are better preserved in diamond drilled holes making the hydraulic parameter measurements more precise.

The distribution coefficients for Sr do not differ by more than one order of magnitude for most rocks and minerals studied. Expected differences between field and laboratory retention values due to different mineral composition are thus of the same magnitude as that measured. For nuclides with larger differences in distribution coefficient between minerals, the knowledge of the mineral composition of the reacting solids (fissure fillings and rocks) is more important.

In both the field experiments a minor amount of Sr migrated almost without retention, the retention factor being  $<2$ . The reason for these fast Sr pulses is not known. One explanation may be that some Sr is sorbed on particles or complexed by organics in the water and transported in one of these forms. Precipitation

of Fe is observed when aerating the groundwater, indicating the possibility of Fe-colloid formation. Filtering of the water samples shows that the Sr-85 activity passes 0.02  $\mu\text{m}$  pores. If there are Sr-transporting particles in the groundwater these are  $< 0.02 \mu\text{m}$ .

No migration of this kind has been observed for the Cs injected simultaneously with Sr. A selectivity of this transport mechanism thus seems to occur. A complex formation may be the explanation, although a study of the influence of humic acid on Sr-sorption did not give any sign of influence of an addition of 10 ppm humic acid. Other humic and fulvic acids than the one used at the laboratory experiment may however be present in the Studsvik water, and an analysis of these and a study of their complex formation with Cs and Sr is required to determine whether this affects the migration.

The temperature of the pumped water indicates that the water (or part of it) originates from depths of the order of 200-400 m. Differences in water temperatures of 4 - 5 $^{\circ}\text{C}$  are thus possible in the flow paths region. As the pumped water is nearly saturated with respect to carbonate small temperature variations could cause precipitation of Ca with the risk of coprecipitation of Sr. Recirculation of pumped water in the second experiment (B5N-B6N) eliminates this risk.

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SiO <sub>2</sub>	65.5 %	Sc	19.6 ppm	Ta	1.0 ppm
TiO <sub>2</sub>	0.6 "	Cr	87 "	Th	8.3 "
Al <sub>2</sub> O <sub>3</sub>	15.1 "	Rb	126 "	U	2.1 "
Fe <sub>2</sub> O <sub>3</sub>	0.6 "	Cs	1.9 "		
FeO	5.3 "	Ba	692 "		
MgO	2.9 "	La	29 "		
CaO	2.7 "	Ce	55.3 "		
Na <sub>2</sub> O	2.9 "	Nd	27 "		
K <sub>2</sub> O	3.0 "	Sm	5.1 "		
MnO	0.1 "	Eu	1.3 "		
P <sub>2</sub> O <sub>5</sub>	0.1 "	Tb	0.86 "		
CO <sub>2</sub>	0.2 "	Yb	3.6 "		
F	0.1 "	Lu	0.57 "		
S	0.2 "	Hf	5.8 "		

Table 1 Average chemical composition of drill cuttings from borehole B8N

Borehole	Level *)	Open/sealed	Dip **)	Identified minerals	Microscopy	XRD	Sorption test
K1N 60 <sup>o</sup>	5.4	open	20	Ca, Q, Pa		x	x
"	11.9	sealed	30	Ca, Q, Cl, Pa		x	x
"	12.6	open	0	Sm, Ca, Q			
"	17.7	sealed		Ca, Q, Cl, Il		x	
"	17.9	open		Cl			
"	29.3	open	70, 30	Ca, Q, Cl	x	x	
"	42.5	sealed	50	Ca, Q, Cl	x		
"	74.9	open	40	Ca, Q, Py		x	x
"	79.9	open	30	Sm, Ca, Q		x	x
"	94.9	open	40	Ca, Q		x	
"	102.3	open	CZ ***)	Ca, Q, Cl		x	
"	104.2	open	0, 30 ***)	Sm, Ca, Cl	x	x	x
"	116.7	open	CZ ***)	Cl, Ca, Q		x	x
"	123.3	open	10	Ca, Q, Cm		x	
"	139.8		20	Ca, Cl, Cm, Py		x	x
"	146.2	sealed	60	Ca, Cl, Q		x	x
"	148.5	open	30	Ca, Q, Sm		x	
"	160.2	open	15	Sm, Q, Cl	x	x	
"	165.5	open	10, 30	Ca, Q, Cl		x	
"	169.25	open	30	Q, Sm, Ca, Cl		x	
"	169.4	open	40-50	Ca, Q, Cl, Sm	x		
"	172.8	open	40	Ca, Q, Cl, Sm		x	
"	179.3	open		Cl, Ca, Q		x	
"	183.1	sealed	10	Ca, Q	x	x	x
"	185.3	sealed	70	Cl, Q, Py			
"	189.45		20	Ca, Q		x	
"	193.6	open	70	Ca, Cl	x	x	
"	198.0	sealed	30	Ca, Q, Cl		x	
"	198.25	open	?	Bi, Q	x	x	

\*) core length in metres

\*\*) deviation from core axis in degrees

\*\*\*) crushed zone

\*\*\*\*) sample for sorption studies (Allard, in progress)

Ca = Calcite, Q = Quartz, Sm = Smectite, Cl = Chlorite, Py = Pyrite, Pa = Palygorskite, Bi = Biotite, Il = Illite, Cm = Unidentified clay minerals

Table 2 Investigated fissure fillings from the K1N drill core

<u>Element</u>		<u>Element</u>	
Na	0.31 %	Th	< 0.5 ppm
Fe	13.2 "	La	3.4 "
Mn	1074 ppm	Ce	8.4 "
Sc	10.3 "	Nd	5.4 "
Cr	4.1 "	Sm	1.2 "
Co	4.3 "	Eu	0.14 "
Rb	< 75 "	Tb	0.2 "
Ba	< 200 "	Yb	0.82 "
Cs	< 0.5 "	Lu	0.12 "
Hf	< 0.5 "		
Ta	< 0.5 "		
Zn	65 "		

Table 3 Element content for a smectite-rich  
fissure filling at 104.2 m

Injection hole	Section	Pumphole B6N, section	Distance
B1N	91.0 - 92.3 m	94 - 102 m	11.8 m
B5N	78.8 - 80.1 m	64 - 66 m	14.6 m

Table 4 Distance between contact zones in different boreholes

Borehole	Section	Number of fractures	Strike	Dip
B1N	91.0 - 92.3 m	4	N 35 - 45°W	55 - 75°N
B5N	78.8 - 80.1 m	1	N 5°W	45°N
		6	N 15°W	35 - 55°N
		1	N 25°W	35°N
B6N	64 - 66 m	3	N 30 - 35°W	60 - 70°N
		4	N 45 - 60°W	55 - 70°N
		1	N 80°W	75°N

Table 5 Number of fractures in borehole sections and their strike and dip

Fracture zone	$K_p$ (m/s)	$K_e$ (m/s)	$\theta_k$
B1N - B6N	$1.4 \cdot 10^{-7}$	$6.6 \cdot 10^{-5}$	$2.1 - 2.2 \cdot 10^{-3}$
B5N - B6N	$7.6 \cdot 10^{-7}$	$1.7 \cdot 10^{-4}$	$4.5 - 4.7 \cdot 10^{-3}$

Table 6 Hydraulic conductivity of rock mass  $K_p$ , weighted average hydraulic conductivity of fractures  $K_e$  and kinematic porosity  $\theta_k$

	80-06-16	80-07-09	80-09-17	80-10-28	80-12-10	80-12-29	81-02-24
Conductance,							
μs/cm	300	300	330	350	245	360	380
pH	7.2	6.6	6.5	6.7	6.4	6.4	7.0
KMnO <sub>4</sub> , mg/l	20	23	26	27	32	33	28
Ca, "	31	43	30	34	26	37	44
Mg, "	9	7	12	12	8	12	13
Na, "	39	40	43	43	26	45	39
K, "	2.8	2.9	2.9	3.7	2.4	2.8	2.9
Cl, "	12	11	12	12	10	12	12
SO <sub>4</sub> , "	29	29	29	30	34	29	26
HCO <sub>3</sub> , "	195	200	218	230	133	227	256
NH <sub>4</sub> , "	0.04	0.04	0.04	0.05	0.01	0.01	0.02
NO <sub>2</sub> , "	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01
NO <sub>3</sub> , "	0.03	0.04	0.03	0.03	0.39	0.01	< 0.01
PO <sub>4</sub> , "	0.02	0.01	0.01	0.01	0.01	0.01	0.01
Fe (II) "	0.07	0.04	0.04	0.04	0.05	0.09	0.05
Fe (tot) "	0.07	0.06	0.07	0.07	0.18	0.11	0.15
Mn, "	0.28	0.29	0.30	0.33	0.10	0.36	0.41
F, "	0.30	0.26	0.22	0.23	0.12	0.24	0.27
SiO <sub>2</sub> , "	11	11	12	11	10	11	11
C, "	4.6	4.7	4.8	5.4	14.9	14.3	6.6

Table 7 Analyses of water pumped from borehole B6N

<u>Element</u>		<u>Element</u>	
Sc	0.039 µg/l	Th	< 0.02 µg/l
Cr	2.7 "	U	3.7 "
Co	0.37 "	La	0.36 "
Rb	2.2 "	Ce	0.50 "
Sr	270 "	Sm	0.088 "
Ba	66 "	Eu	0.022 "
Cs	0.021 "		
Zn	102 "		
W	1.8 "		
As	8.7 "		
Br	15 "		

Table 8 Trace elements in water pumped from borehole B6N

<u>Flow path</u>	<u>Tracer injected</u>	<u>Chemical form</u>	<u>Total radio-activity</u>	<u>Concentration (carriers included)</u>
B1N-B6N	I-131	NaI	352 MBq	0.08 M
	H-3	Tritiated water	357 "	
	Sr-85	SrCl <sub>2</sub>	707 "	10 <sup>-4</sup> M
B5N-B6N	I-131	NaI	309 MBq	
	H-3	Tritiated water	345 "	
	Sr-85	SrCl <sub>2</sub>	750 "	1.1·10 <sup>-4</sup> M
	Cs-134	CsCl	1280 "	0.4·10 <sup>-4</sup> M

Table 9 Data on the Sr-85 and Cs-134 migration tests

Specie (mg/l)	Natural groundwater Borehole B6N (800609)	Synthetic groundwater S	Synthetic groundwater A
Na <sup>+</sup>	37	93	65
K <sup>+</sup>	3.6	4.0	3.9
Ca <sup>2+</sup>	36	14	18
Mg <sup>2+</sup>	8.7	9.0	4.3
Cl <sup>-</sup>	7	28	70
SO <sub>4</sub> <sup>2-</sup>	35	34	9.6
HCO <sub>3</sub> <sup>-</sup>	204	200	123
Si <sub>tot</sub>	5.3	5.5	5.3
pH	8.1	7.8 - 8.0	8.0 - 8.2

Table 10 Water compositions

Mineral/Rock	$K_d$ (Sr) (m <sup>3</sup> /kg)	$K_d$ (Cs) (m <sup>3</sup> /kg)
Quartz	0 - 0.002	0.001
Laumontite	0.029	0.117
Stilbite	0.188	0.950
Hornblende	0.005 - 0.013	0.213
Biotite	0.016 - 0.024	0.624
Muscovite	0.071 - 0.114	0.765
Smectite (MX-80)	2.9	1.4
Kaolinite	0.029	0.059
Illite	0.335	> 5
Chlorite	0.050	0.129
Orthoclase	0.002 - 0.005	0.011
Plagioclase	0.004	0.011
Microcline	0.012	0.177
Epidote	0.010	0.019
Prehnite	0.010, 0.016 <sup>a)</sup>	0.013, 0.048 <sup>a)</sup>
Calcite	0.001 - 0.003, 0.004 <sup>b)</sup>	0.001, 0.006 <sup>b)</sup>
Apatite	0.005 - 0.008	0.003
Gypsum	0.003	0.006
Pyrite	0.003	0.003
Fluorite	0.002 - 0.003	0 <sup>a)</sup>
Magnetite	0.002 - 0.003	0.007
Hematite	0.004 - 0.007	0.006
Dolomite	0.003	0.005
Granite from Stripa	0.006	0.152
Granite " Finnsjö	0.020	0.490
Diabase " Blekinge	0.010	0.368
Gneiss " "	0.009	0.107
Gneiss " Studsvik	0.015	0.943
Gneiss " " c)	0.006	1.214
Gneiss " " d)	0.040	0.560

a) From fracture surface Finnsjö, contains 10 % calcite.

b) From fracture surface Finnsjö. c) In natural water.

d) Artificially weathered.

Table 11 Distribution coefficients for Sr and Cs for rocks and rockforming, Accessory and fracture filling mineral at pH 8-8.5 trace conc. ( $\sim 10^{-8}$  M), ambient temp., synth. groundwater. (Torstenfelt et al 1981, Allard et al, 1981)



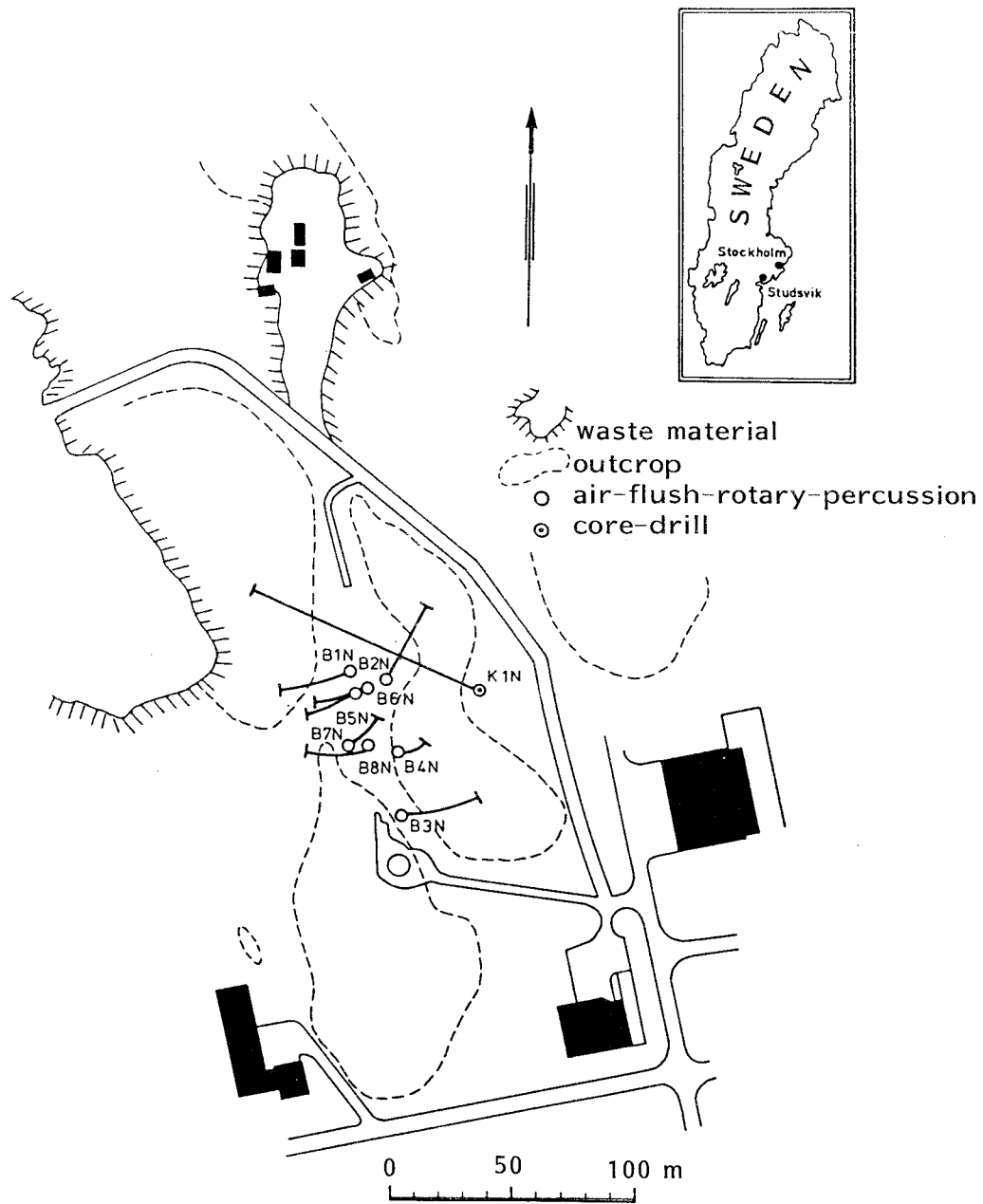


Fig 1. Location of boreholes and their deviation projected on the horizontal plane

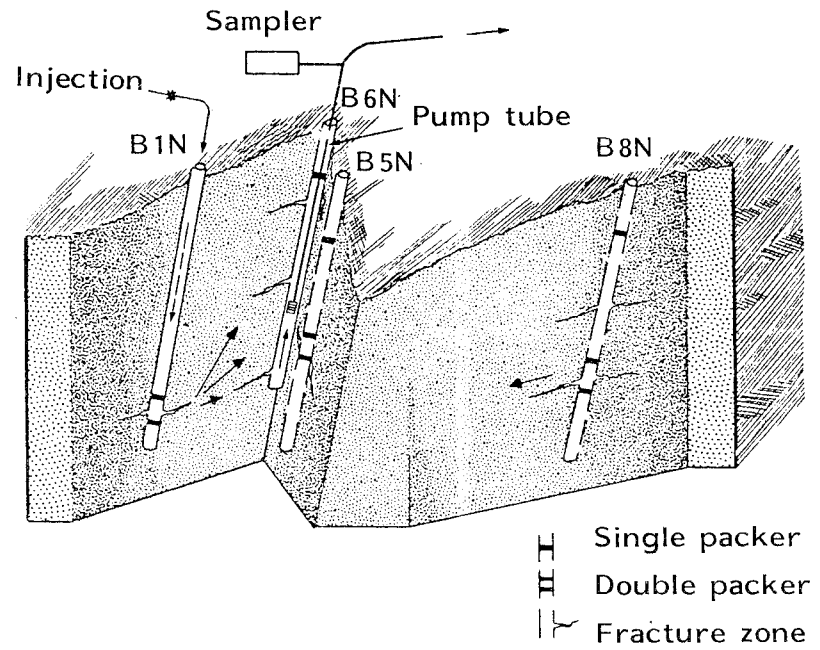


Fig 2. Cutaway view of the in situ test site

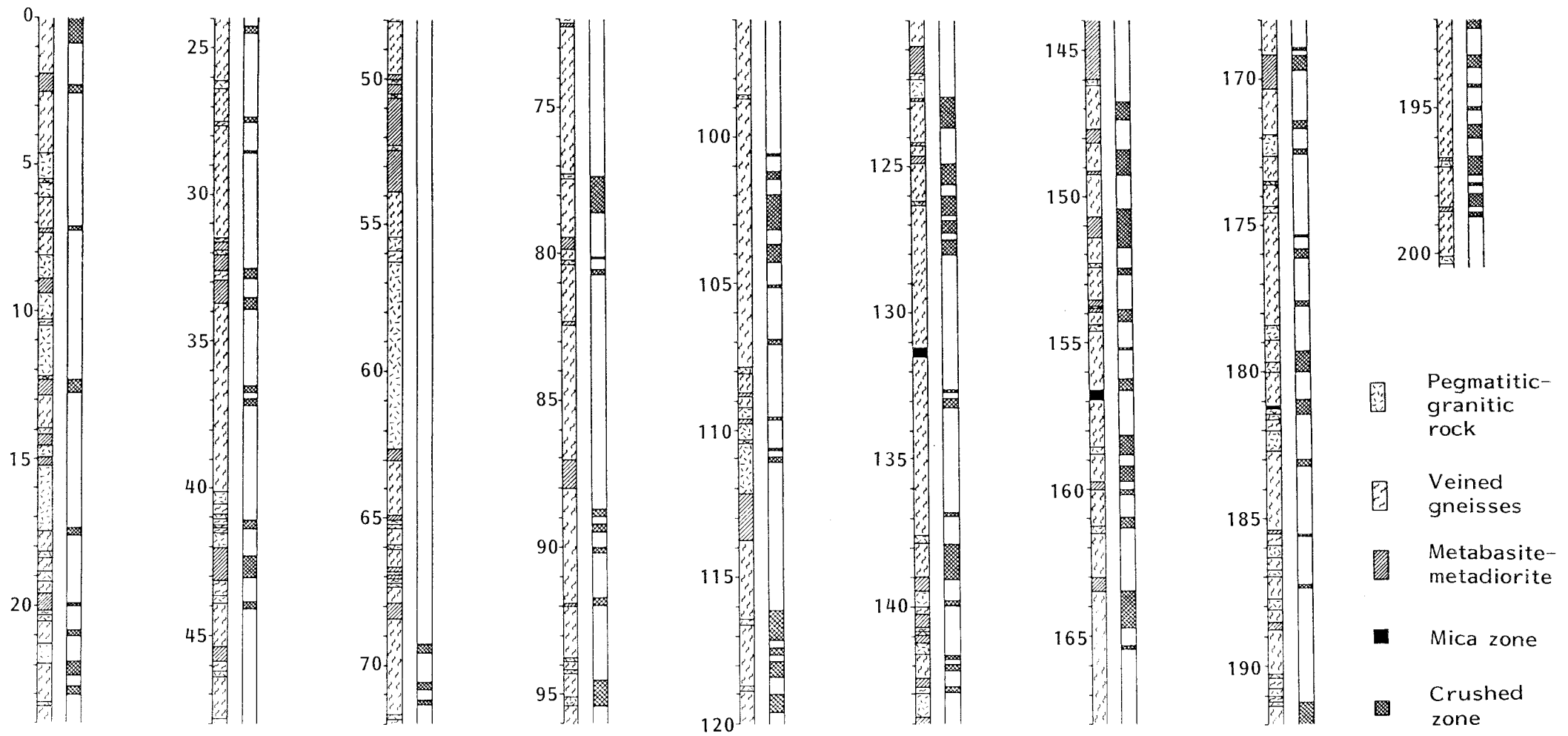


Fig 3. Core mapping of borehole K1N. Rock types and crushed zones are distinguished

Fissure filling materials

Borehole K1 50 - 125 m

N = 261

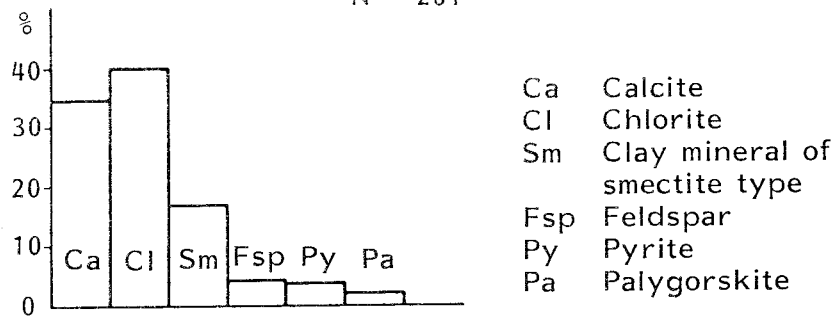


Fig 4. Frequency of fissure filling minerals in the K1N drill core

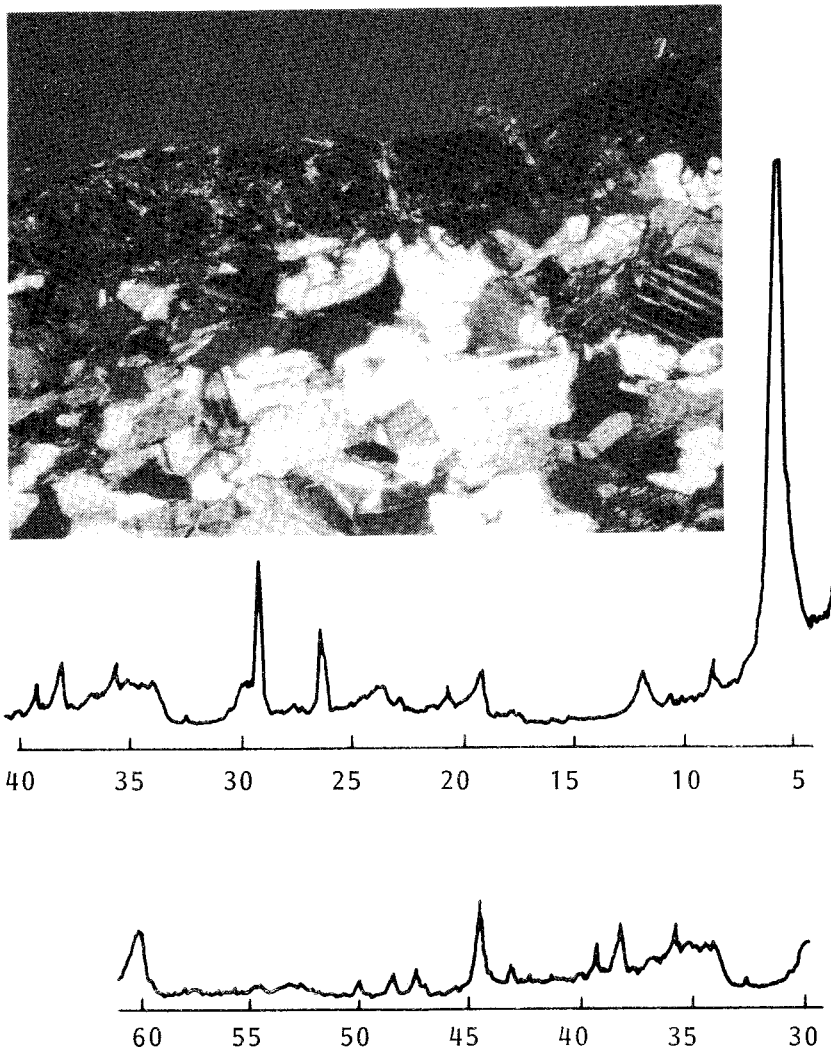


Fig 5. Microphotograph and X-ray diffractogram showing smectite as fissure filling in sample 104.2 m (the fibrous, dark grey material seen in the upper part of the photo)

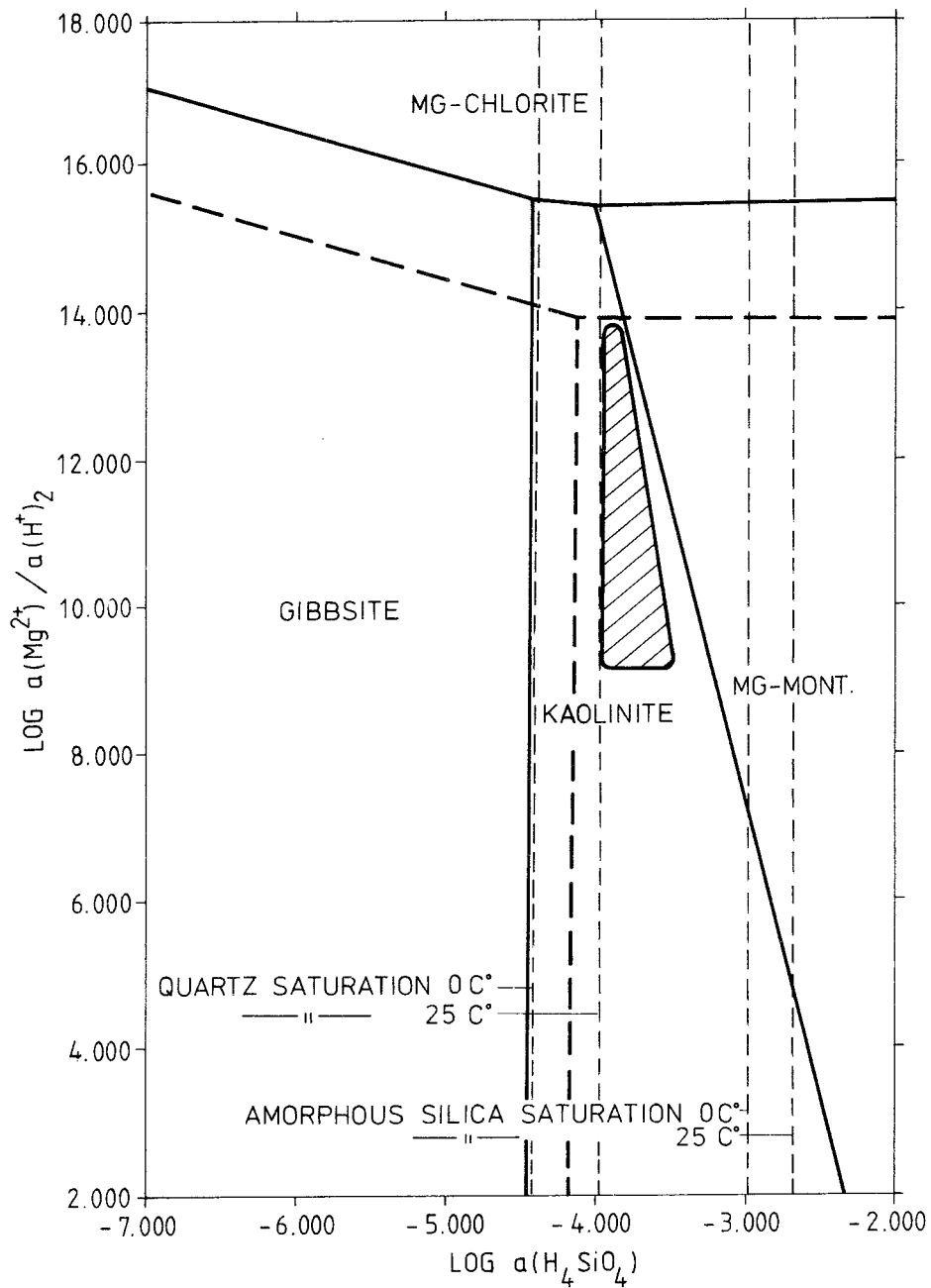


Fig 6. Stability diagram of the system HCL-H<sub>2</sub>O-AL<sub>2</sub>O<sub>3</sub>-CO<sub>2</sub>-MGO-SIO<sub>2</sub> at 0 degr. C. (full core) and 25 degr. (broken line). According to Helgesson (1971). Ruled area includes analysed water from B6N (Landström, 1980)

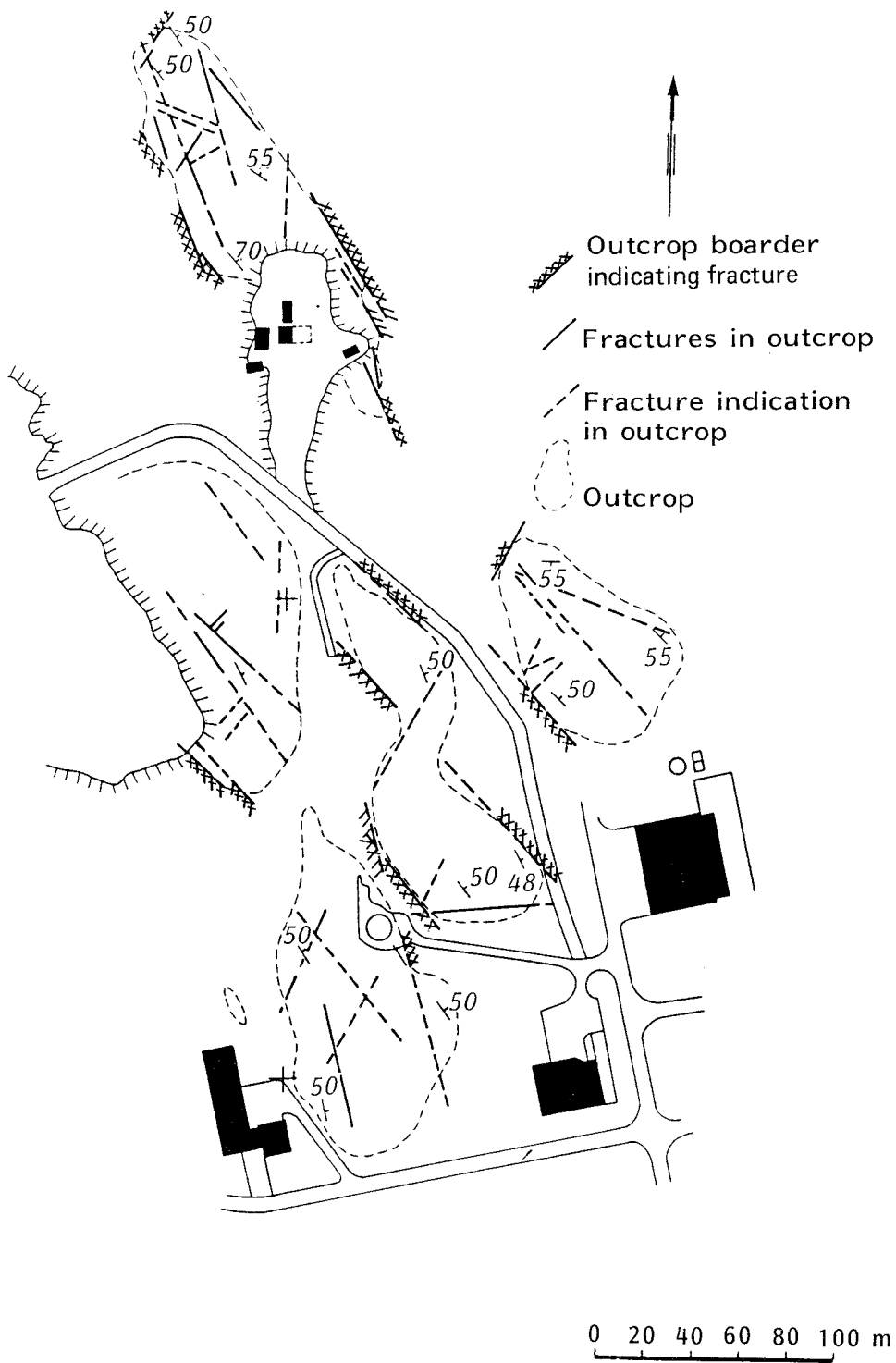


Fig 7. Fracture map of the test area

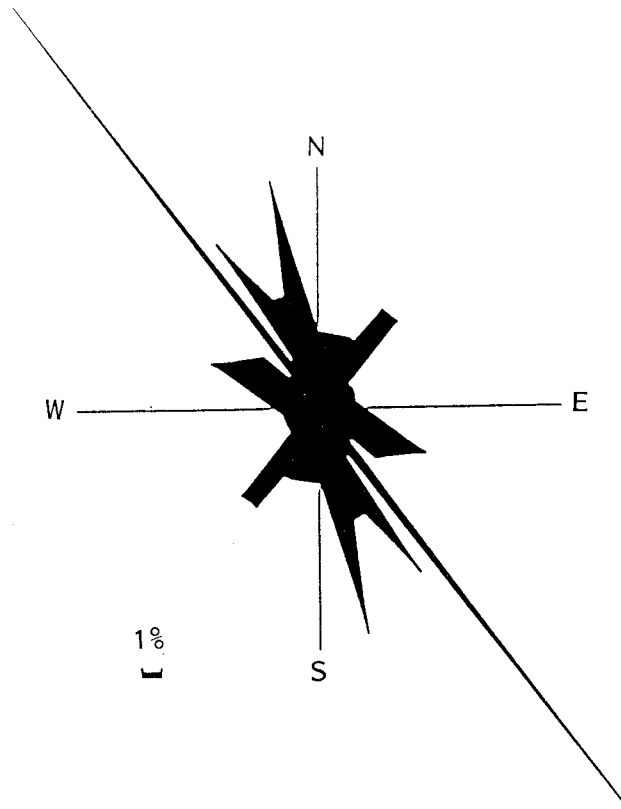


Fig 8. Rose diagram showing 52 mainly vertical fractures

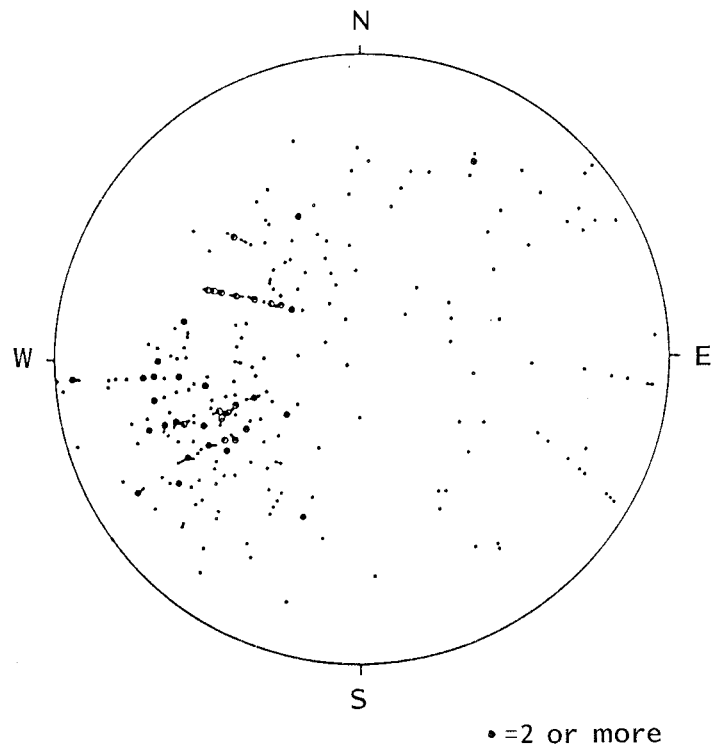


Fig 9. Stereographic pole-diagram including all fractures which are orientated by TV-inspection. The normal of the fracture planes projected on the lower hemi-sphere

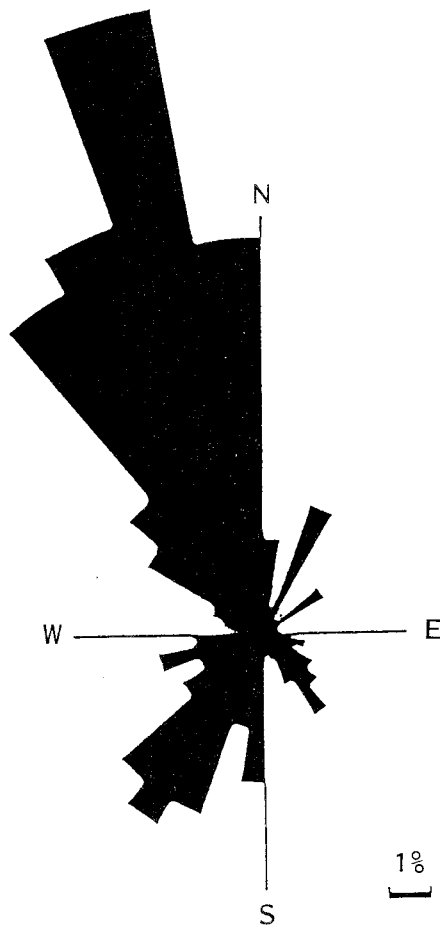


Fig 10. Histogram over the strike of fractures orientated by TV-inspection. Fractures dipping towards the north are plotted north of the W-E line while the others are plotted south of the W-E line

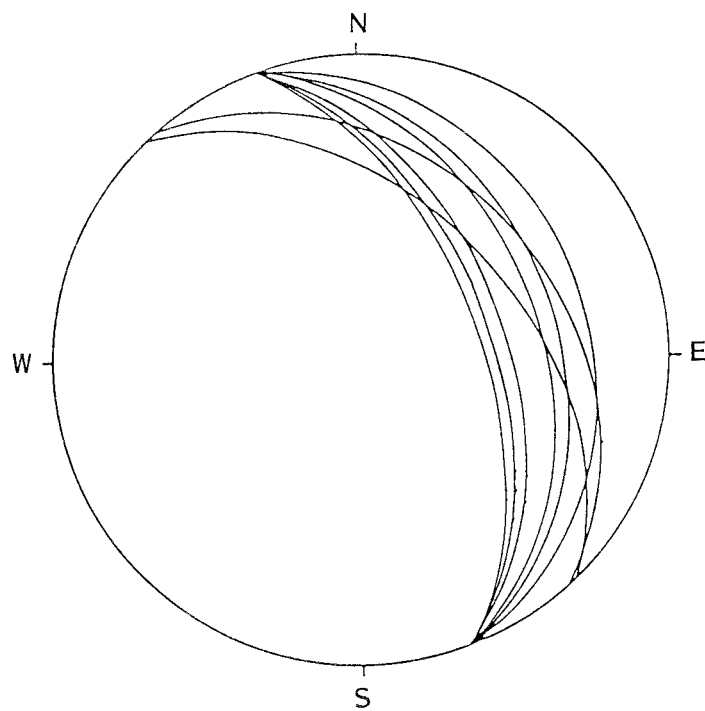


Fig 11. Stereographic projection of the fracture planes intersection with the lower hemisphere, borehole B5N, section 78 - 80 m

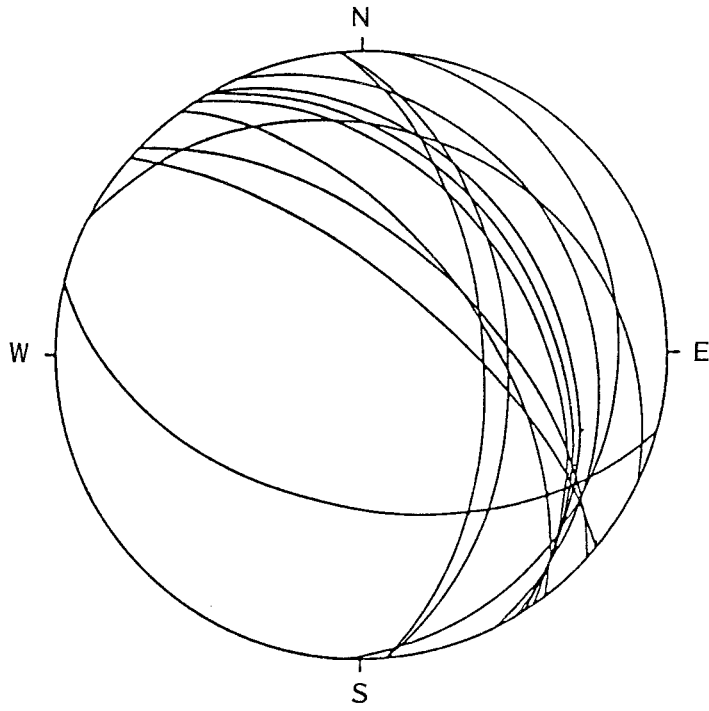


Fig 12. Stereographic projection of the fracture planes intersection with the lower hemi-sphere, borehole B1N, section 84 - 94 m

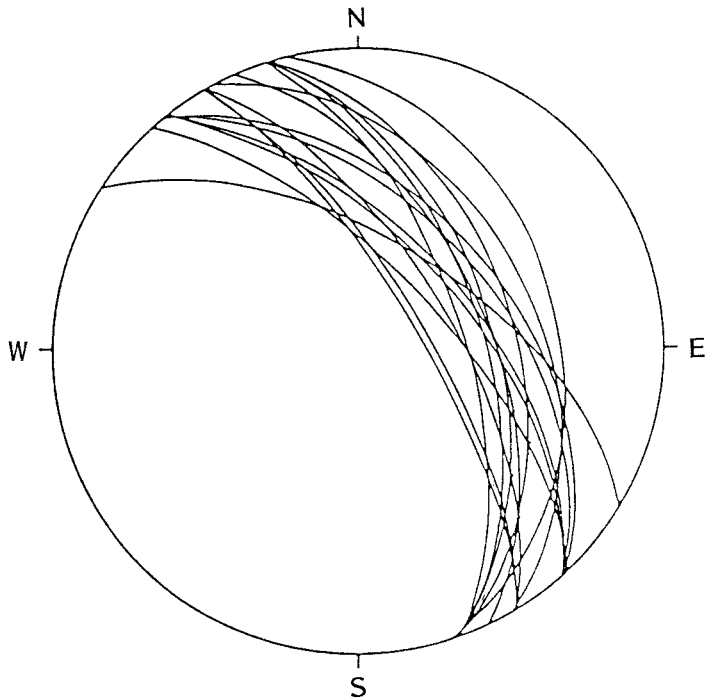


Fig 13. Stereographic projection of the fracture planes intersection with the lower hemi-sphere, borehole B6N, section 41 - 45 m, 47 - 50 m, and 62 - 68 m



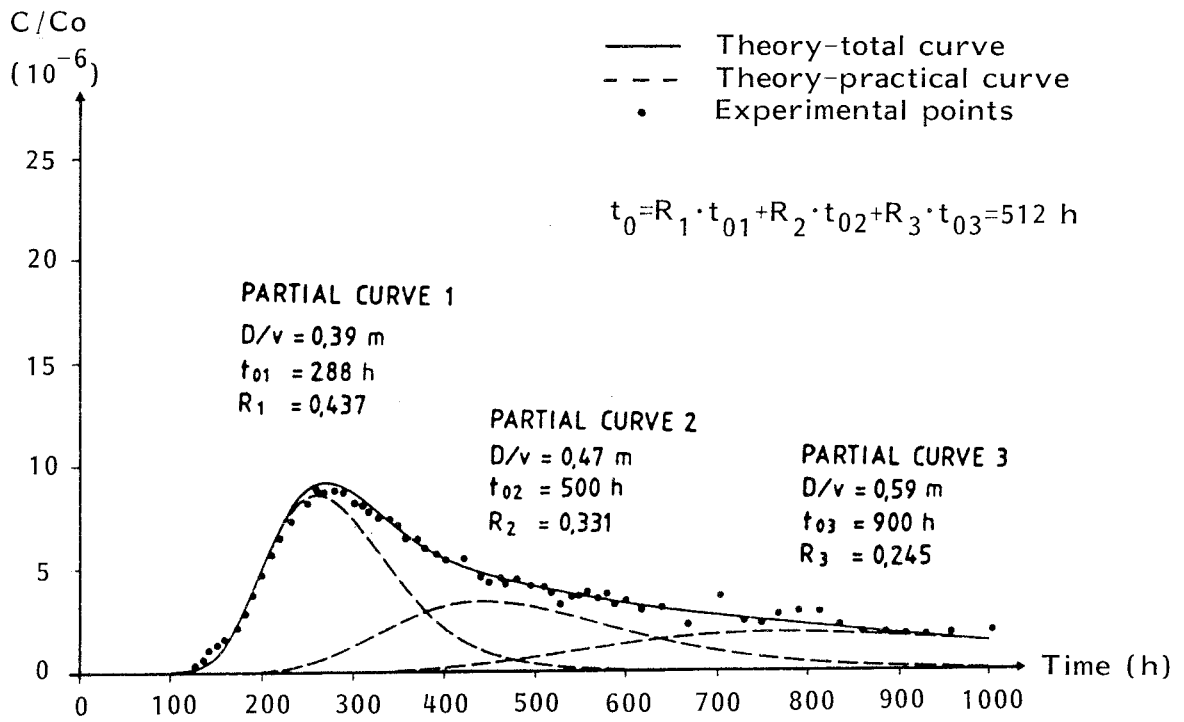


Fig 14. Experimental break-through curve for I-131 in B1N-B6N and fit with theoretical curves

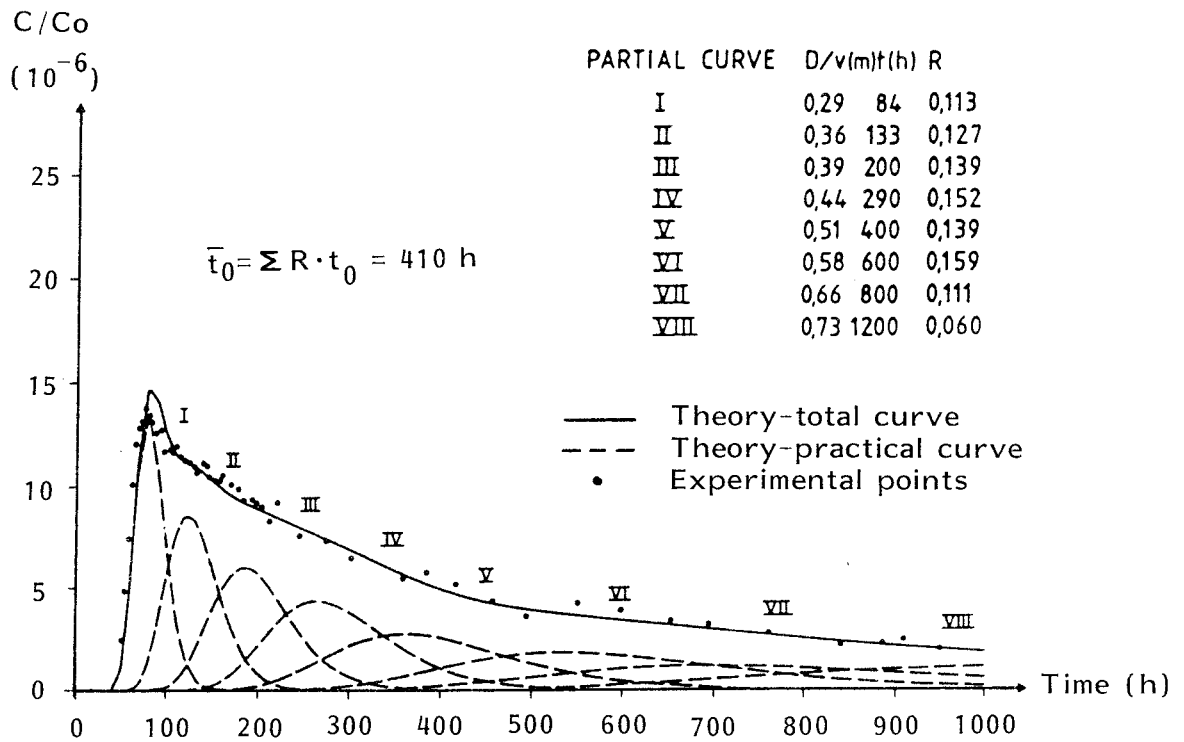


Fig 15. Experimental break-through curve for I-131 in B5N-B6N and fit with theoretical curves

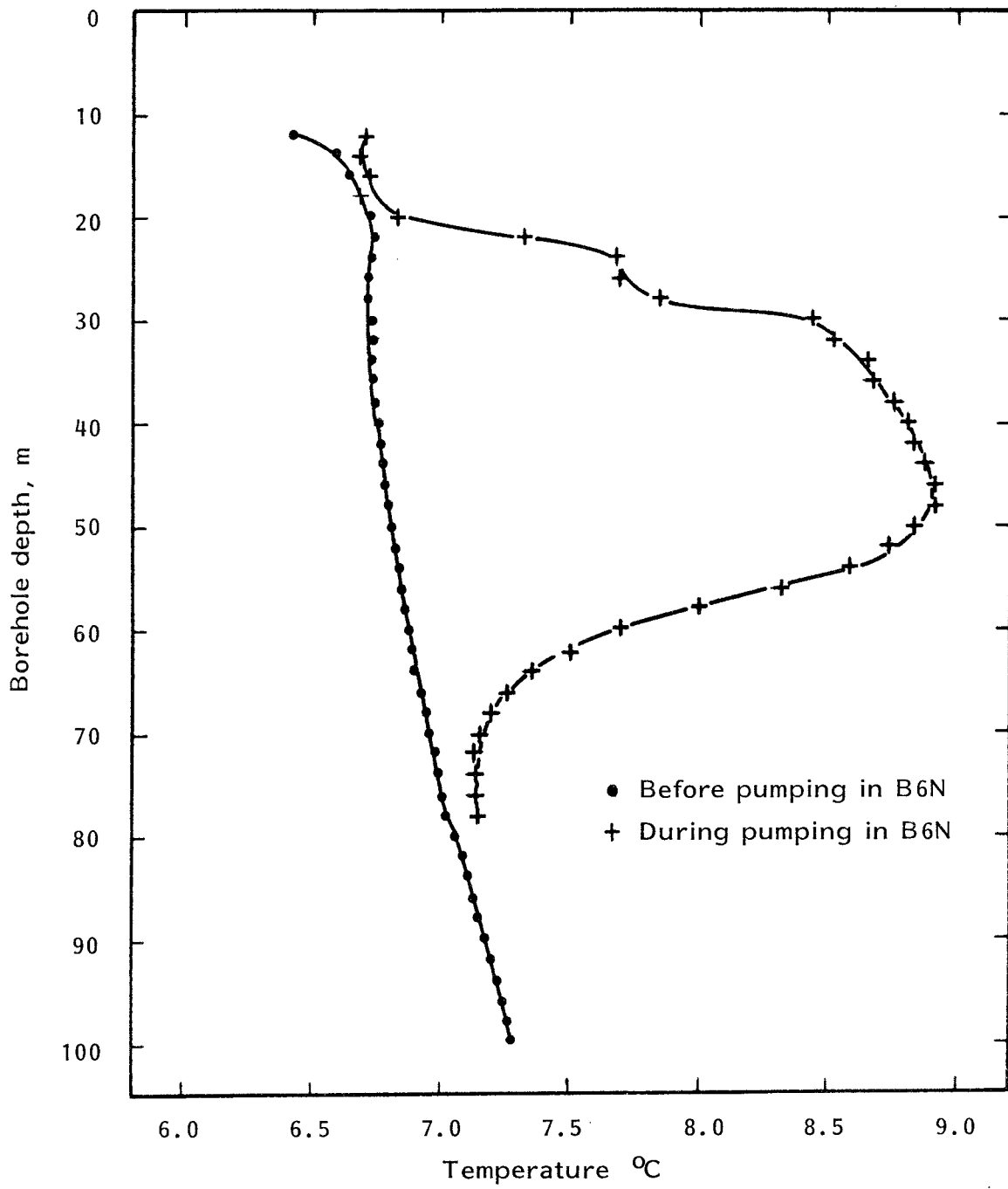


Fig 16. Temperature - depth profiles in borehole B5N

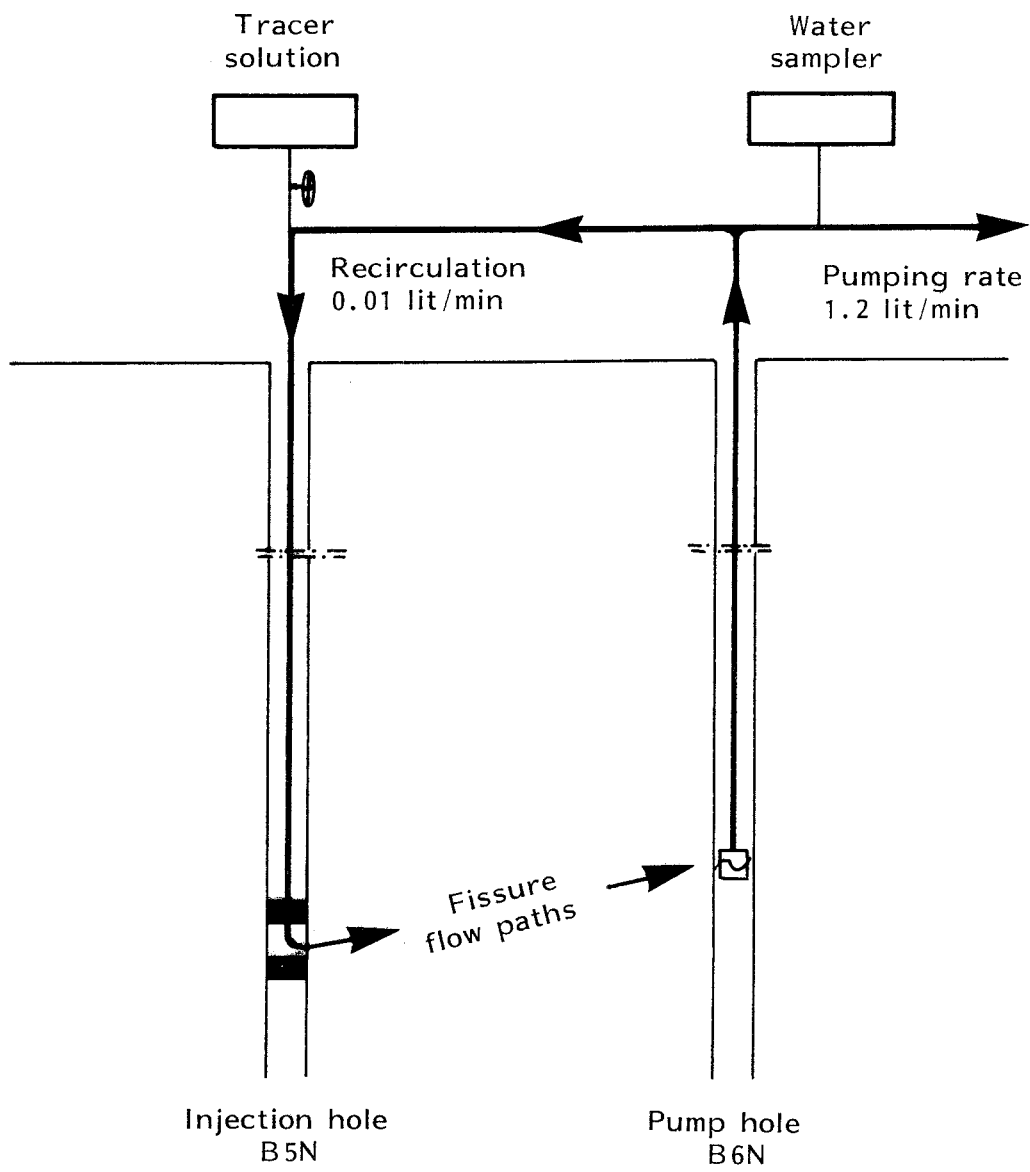


Fig 17. Outline of the recirculation system

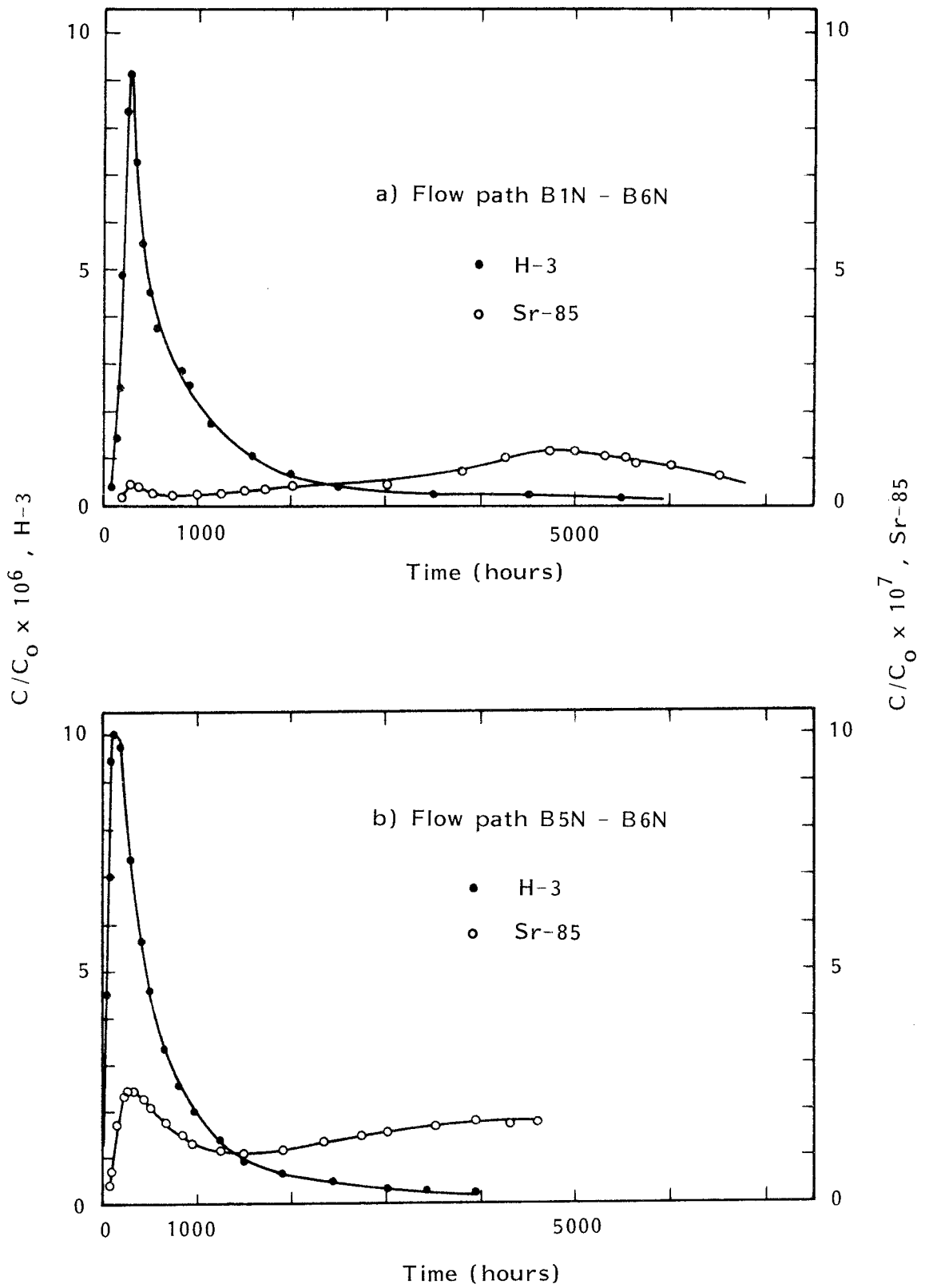


Fig 18. Field experiments H-3 and Sr-85 break-through curves

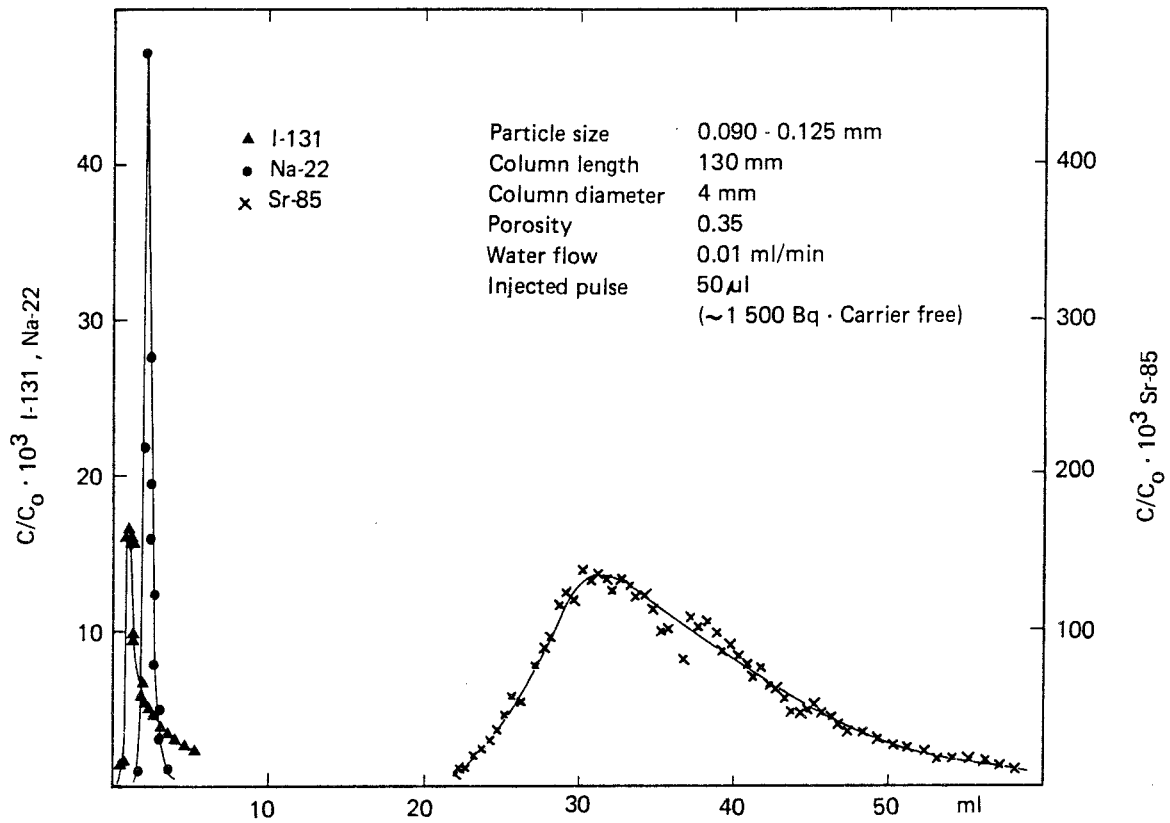


Fig 19. Laboratory experiment I-131, Na-22 break-through curves

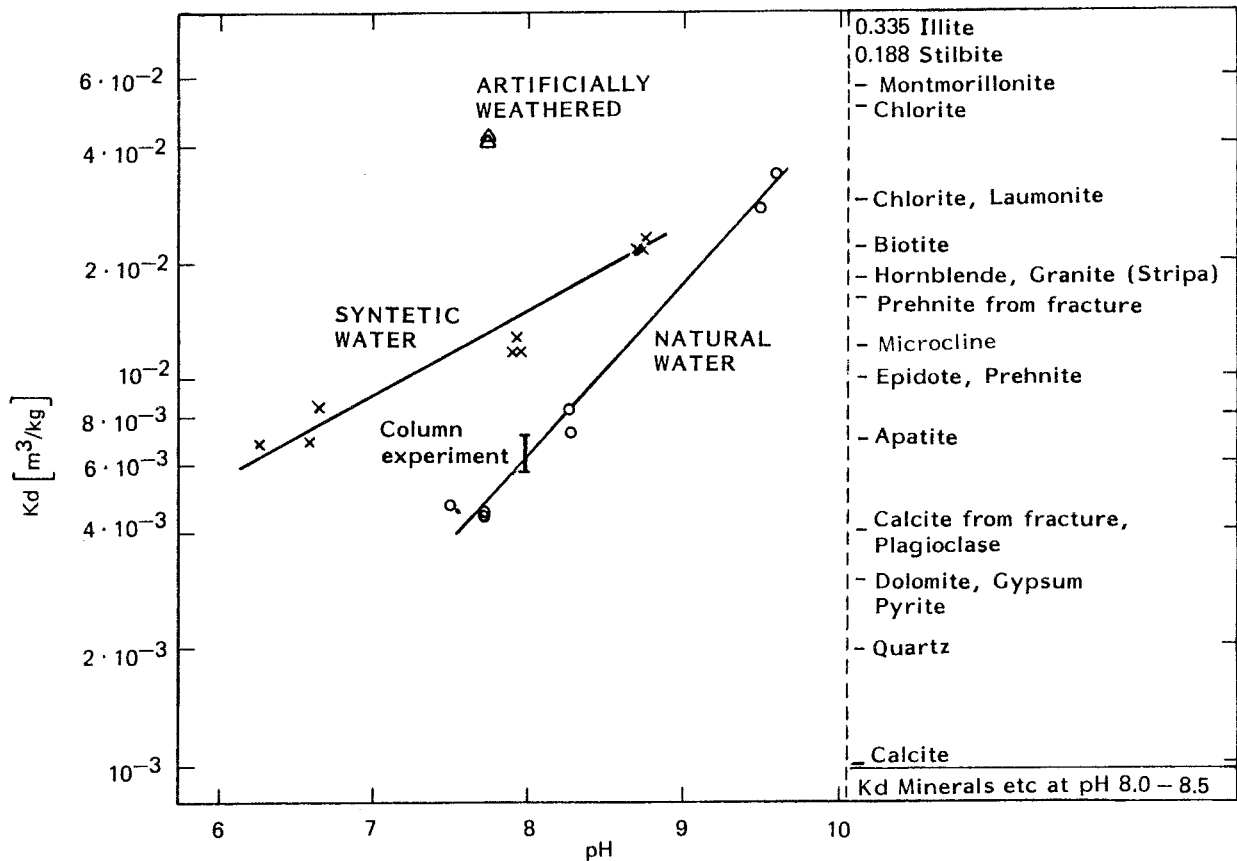


Fig 20. Distribution coefficients for Sr-85. 1 week contact time,  $\sim 10^{-8}$  M (Sr)

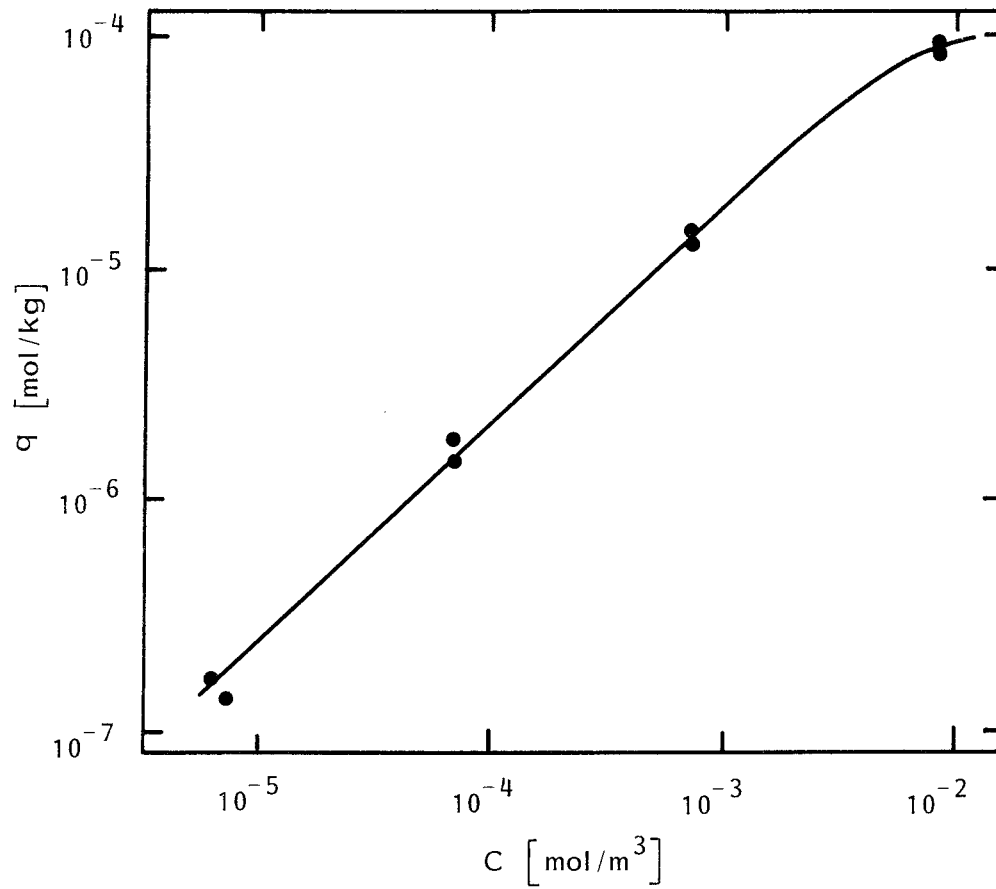


Fig 21. Sorption isotherm for Sr on Studsvik gneiss  
Initial concentrations:  $10^{-8}$ ,  $10^{-7}$ ,  $10^{-6}$ ,  $10^{-5}$  M

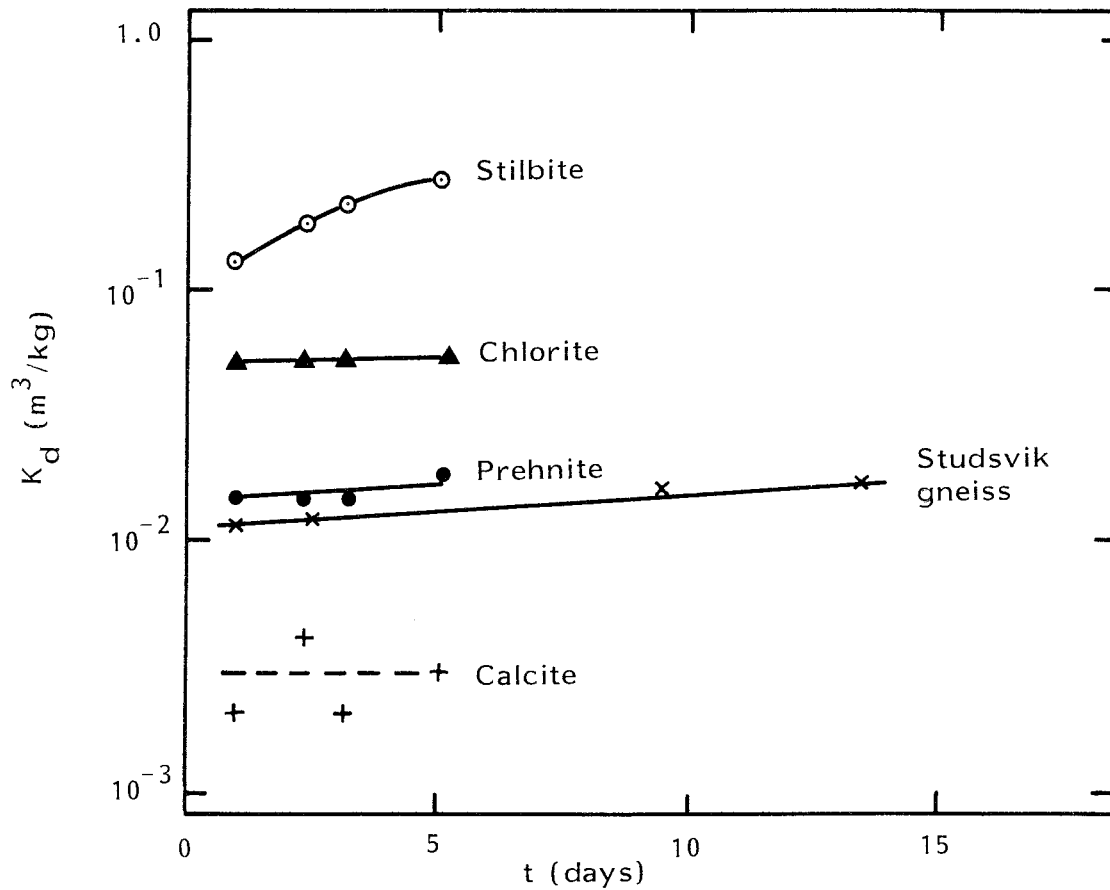


Fig 22. Time dependence for Sr-sorption 1.0

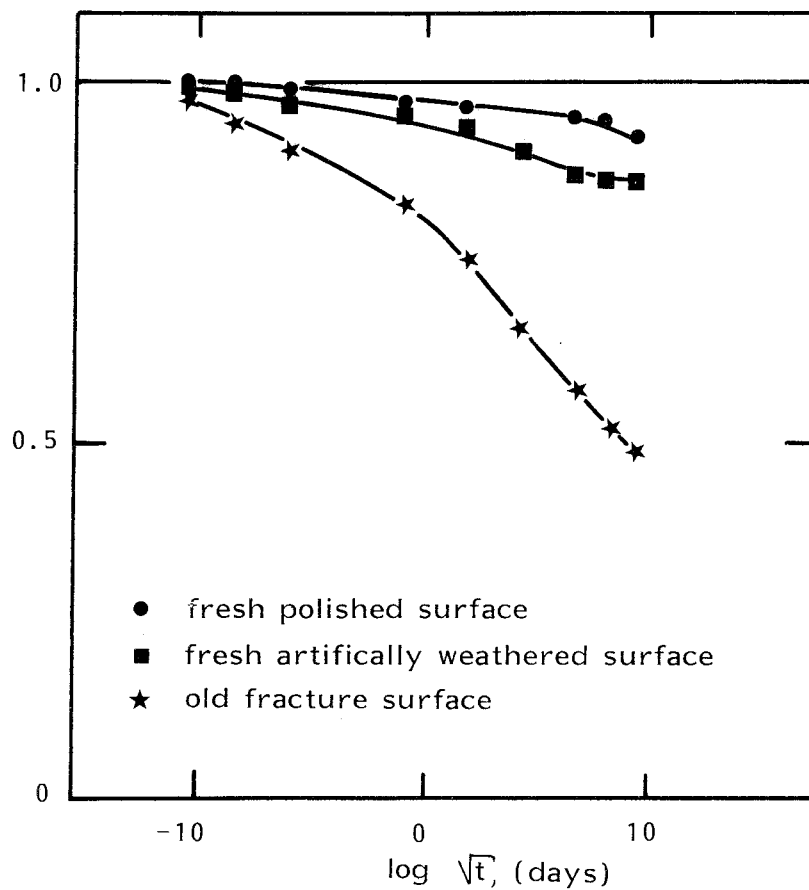


Fig 23. Sorption on macro-surfaces: Sr uptake vs. time for various granite surfaces (Finnsjö granite)

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