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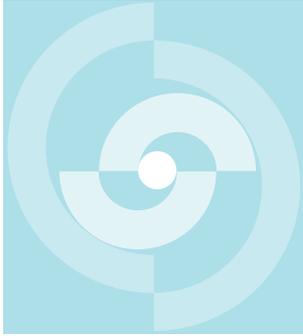
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# TECHNICAL REPORT 84-24

PERMEABILITY, POROSITY, AND RETARDATION  
CHARACTERISTICS OF GRANITE SAMPLES  
FROM THE BOETTSTEIN BOREHOLE;  
A LABORATORY STUDY

Lars Carlsen  
Dorte Platz

March 1985

Risø National Laboratory, Chemistry Department  
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Le présent rapport a été préparé sur demande de la Cédra. Les opinions et conclusions présentées sont celles des auteurs et ne correspondent pas nécessairement à celles de la Cédra.

This report was prepared as an account of work sponsored by Nagra. The viewpoints presented and conclusions reached are those of the author(s) and do not necessarily represent those of Nagra.

PREFACE

Safety analyses for nuclear waste repositories require extensive data material on the host rock which cannot all be obtained from field tests during the ongoing Nagra deep drilling program in Northern Switzerland.

Among the initial goals of this study at the Risø National Laboratory was to find out about the composition of porewater isolated in "old" fissures; these determinations failed, largely due to the low porosities and inadequate sample conservation after core recovery. The application of the "liquid chromatography" technique, however, yielded interesting data on the permeability of Böttstein granite, as well as some valuable insights on how future fissure flow experiments might be improved.

The reader should keep in mind that the preparation of the laboratory samples unavoidably disrupts the original stress field, and therefore the geometry of the micropores by decompression. Furthermore the experiments were carried out with water which was not in chemical equilibrium with the rock, but was chosen because many other studies were based upon this rather saline deep ground water from the crystalline of Bad Säckingen, before Nagra defined a different reference water for its studies in the crystalline bedrock of central Northern Switzerland.

SUMMARY

A liquid chromatography technique was utilized to explore permeability, porosity, and retardation laboratory characteristics of a series of granite samples from 489-670 m depth of the Böttstein borehole, as part of the Nagra deep drilling programme in Northern Switzerland. The samples comprised cores with old healed, mineral/clay-filled fractures and cores of "fresh" intact rock without filled fractures. These samples revealed no detectable water flow at pressure drops up to 90 bar, i.e., their hydraulic conductivities were smaller than  $4 \cdot 10^{-14}$  m sec<sup>-1</sup>.

After artificially fissuring and compressing the samples hydrostatically with pressures approximating the initial rock overburden, the "fresh" and "reopened" fissures exhibit values of  $10^{-9}$ - $10^{-10}$  and  $10^{-11}$  m sec<sup>-1</sup>, respectively, with flow porosities usually smaller than 0.5 %. These fissures displayed a strong tendency to rapid "self-healing" by contact with the relatively HCO<sub>3</sub><sup>-</sup>-rich Säckinggen water which was used in the experiments. These results suggest that groundwater transport through unfractured solid Böttstein granite is limited to aqueous phase diffusion in micropores.

Retardation of the sorbing species TcO<sub>4</sub><sup>-</sup>, Sr<sup>2+</sup>, and Co<sup>2+</sup> was established in an artificially induced fissure in a "fresh" granite sample. The values of 0.16, 0.24, and 0.04, respectively, must be interpreted with great caution, since the applied hydraulic gradient of 2800, and hence the resulting flow rate were unrealistically high.

RESUME

Une méthode de chromatographie liquide est utilisée pour déterminer la perméabilité, la porosité et quelques facteurs de rétention d'une série de carottes de granite, extraites entre 489 et 670 m de profondeur, dans le forage de Böttstein qui fait partie du programme de forages profonds de la Cédra dans le nord de la Suisse. Les échantillons comprennent aussi bien des carottes avec d'"anciennes" failles fermées que du matériel rocheux "frais" et non-diaclasé. Ces échantillons n'ont pas révélé la moindre circulation d'eau à des pressions montant jusqu'à 90 bar, ce qui signifie en d'autres termes que les conductivités hydrauliques sont inférieures à  $4 \cdot 10^{-14}$  m sec<sup>-1</sup>.

Quelques échantillons cylindriques sont ensuite artificiellement fissurés longitudinalement puis comprimés dans la presse jusqu'à ce qu'ils retrouvent environ leur surcharge rocheuse initiale. Après ce traitement le matériel frais et non-diaclasé indique alors des perméabilités de  $10^{-9}$  à  $10^{-10}$  m sec<sup>-1</sup>, tandis que les échantillons dont les diaclases se sont réouvertes révèlent une perméabilité d'environ  $10^{-11}$  m sec<sup>-1</sup>. Difficiles à déterminer, les porosités sont en tout cas inférieures à 0.5 %.

Ces fissures artificielles montrent une forte tendance à se refermer rapidement au contact de l'eau de Säkingen relativement riche en  $\text{HCO}_3^-$ , qui a été utilisée pour ces expériences.

Les résultats indiquent que le transport de l'eau à travers le granite de Böttstein est essentiellement dû à la diffusion du fluide dans des micro-pores.

La rétention de  $\text{TcO}_4^-$ ,  $\text{Sr}^{2+}$  et  $\text{Co}^{2+}$  a été définie dans une fissure artificielle d'un échantillon de granite frais. Les valeurs de 0.16, 0.24 et 0.04 qui en résultent doivent toutefois être interprétées avec une extrême prudence, vu qu'elles sont obtenues sous des gradients hydrauliques (2'800) et donc également des vitesses d'écoulement, beaucoup trop élevés.

### ZUSAMMENFASSUNG

Eine Flüssig-Chromatographie-Methode wurde angewandt, um die Permeabilität, Porosität und einige Retentionsfaktoren einer Reihe von Granitkernen aus 489-670 m Tiefe aus der Sondierbohrung Böttstein im Rahmen des Nagra-Tiefbohrprogramms im nord-schweizerischen Grundgebirge abgeteuft wurde, zu bestimmen. Die Proben wurden sowohl aus Kernen mit "alten", verheilten Klüften als auch aus "frischem", unzerklüftetem Material geschnitten. Keine dieser Proben liess irgendwelchen Durchfluss bei Druckdifferenzen bis 90 bar erkennen; das heisst, die hydraulischen Konduktivitäten lagen durchwegs unter  $4 \cdot 10^{-14} \text{ m sec}^{-1}$ .

Einige Probenzylinder wurden künstlich längs aufgespalten und in der Druckabpresseinrichtung hydrostatisch annähernd auf die ursprüngliche Gesteinsüberlastung komprimiert. Frisches, unzerklüftetes Material wies nun Permeabilitäten von  $10^{-9}$  bis  $10^{-10} \text{ m sec}^{-1}$ , Proben mit wieder aufgespaltenen, verheilten Klüften solche von  $10^{-11} \text{ m sec}^{-1}$  auf. Porositäten waren nur schwierig zu bestimmen, lagen aber durchwegs unter 0.5 %.

Diese künstlichen Spalten zeigten eine starke Tendenz, rasch wieder "zuzuheilen" im Kontakt mit relativ  $\text{HCO}_3^-$ -reichem Säckinger Wasser, welches für diese Experimente verwendet wurde.

Die vorliegenden Resultate deuten darauf hin, dass der Wassertransport durch den Böttstein-Granit vorwiegend durch Flüssigkeitsdiffusion innerhalb von Mikroporen stattfindet.

Die Retention von  $\text{TcO}_4^-$ ,  $\text{Sr}^{2+}$ , und  $\text{Co}^{2+}$  Ionen innerhalb einer künstlichen Spalte im "frischen" Gestein wurde bestimmt. Es resultierten Werte von 0.16, 0.24, bzw. 0.04, die aber mit äusserster Vorsicht zu interpretieren sind, da diese bei unrealistisch grossen hydraulischen Gradienten (2'800) und daher zu hohe Durchflussgeschwindigkeiten gemessen wurden.

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## 1. INTRODUCTION

It is generally accepted that migration of radionuclides released from high-level waste repositories placed in granite formations mainly takes place through the fissures in the crystalline rock /1/. In this connection it appears important to characterize fissured rock samples by their physico-chemical properties such as porosity and permeability, as well as their ability to sorb radionuclides, possibly present in the groundwater, onto the fissure surface. Both "old" fissures, filled with weathering products, i.e. minerals and clays, and "fresh" fissures, e.g. originating from drilling operations have to be considered.

In connection with the Nagra program on management and disposal of high-level radioactive waste in granite formations, we have studied a series of core samples originating from the test-site at Böttstein in the northern part of Switzerland /2/.

In principle four different types of samples have been considered: 1) unfissured intact rock samples, 2) intact rock samples containing "old" mineral-filled fissures, 3) samples as under 2), however, artificially cracked along the filled fissures, and 4) intact samples, originally unfissured as under 1), however, artificially cracked. It is emphasized that in all cases the term fissure refers to "macro fissures", whereas "micro fissures", which may significantly participate in the retardation mechanism of diffusing species /3/, have not been included in the present study.

The investigations to elucidate the physico-chemical properties of these types of samples have been conducted by application of a liquid chromatographic technique, which is a slightly modified version of the previously described procedure /4/ (cf. experimental section).

## 2. GROUNDWATER COMPOSITION

In all cases groundwater originating from the formation /5/ was applied in the LC studies. The content of selected ions in the groundwater is given in Table 1. The pH was found to be 7.93.

**Table 1: Concentrations (ppm) of selected ions in groundwater from Bad Säckingen (B.S.) compared to Risø "standard" groundwater (stand) /6/**

	Na <sup>+</sup>	K <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	Fe <sup>2+</sup> / Fe <sup>3+</sup>	Cl <sup>-</sup>	HCO <sub>3</sub> <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>
B.S.	925	128	15	150	1	1'910	275	110
stand	23	0	12	20	0	36	61	48

Helium purge of the water samples prior to use in order to remove oxygen (vide infra) did not change the HCO<sub>3</sub> content, nor the pH.

Attempts to study the composition of water present in the granite samples were unsuccessful. The apparent lack of water in the samples was ascribed to the unfavourable storage of the core samples (unwrapped) leading to draining.

### 3. SAMPLES INVESTIGATED

It must be strongly emphasized that only a very limited number of core samples was available for the investigations described. Four core samples were received by our laboratory, one of these, however, was overly fractured for the sample coring procedures and yielded desintegrated columns unsuited for further use in the compression module (cf. experimental section). In Table 2 data on the single core samples and the resulting "columns" for the liquid chromatographic studies are summarized.

**Table 2: Core samples received by Risø National Laboratory for column preparation, and the resulting column characterization**

Core no.	depth (m)	resulting columns*
I	489.63 - 489.97 (cf. Fig. 1)	A: 5.75 cm artificially fissured B: 10.0 cm with "old" clay/mineral filled fissure
II	609.30 - 609.60 (cf. Fig. 2)	A: 10.0 cm unfissured B: 10.0 cm artificially fissured C: as B
III	667.27 - 667.64 (cf. Fig. 3)	A: 10.0 cm with "old" clay/mineral filled fissure B: 7.8 cm as A, however, artificially fissured along the natural fissure
IV	696.50 - 696.72 (cf. Fig. 4)	none

\* all sample cores exhibit a diameter  $\phi = 2.5$  cm

The preparation of the cylindric sample columns ("plugs") was carried out by the Core Laboratory at the Danish Geological Survey as described in the experimental section. The photographic documentation of the core samples (before sample-coring) (cf. Figs. 1 - 4) was also carried out by DGU.

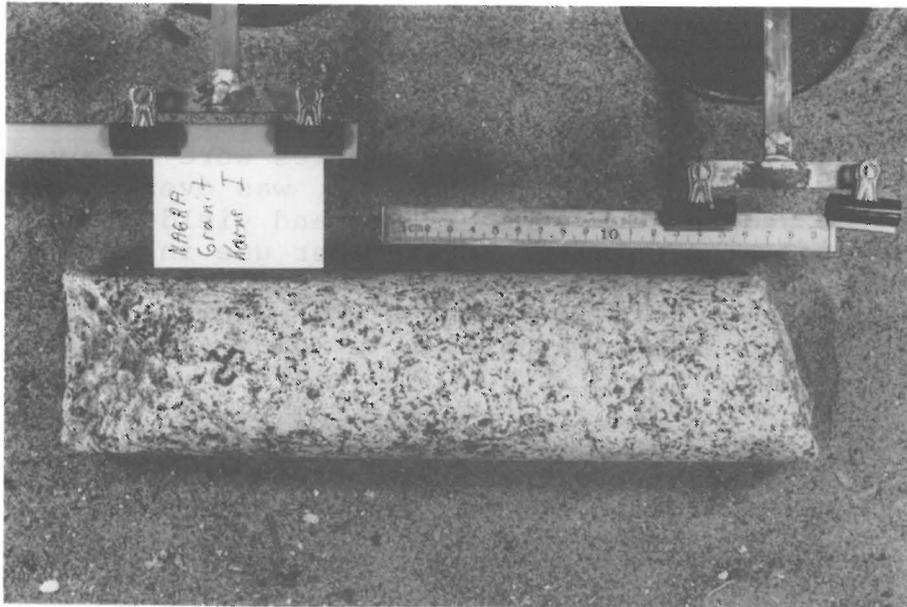


Fig. 1: Core sample no. I (Böttstein granite, depth 489.63 - 489.97 m): Solid core with clay mineral filled fracture

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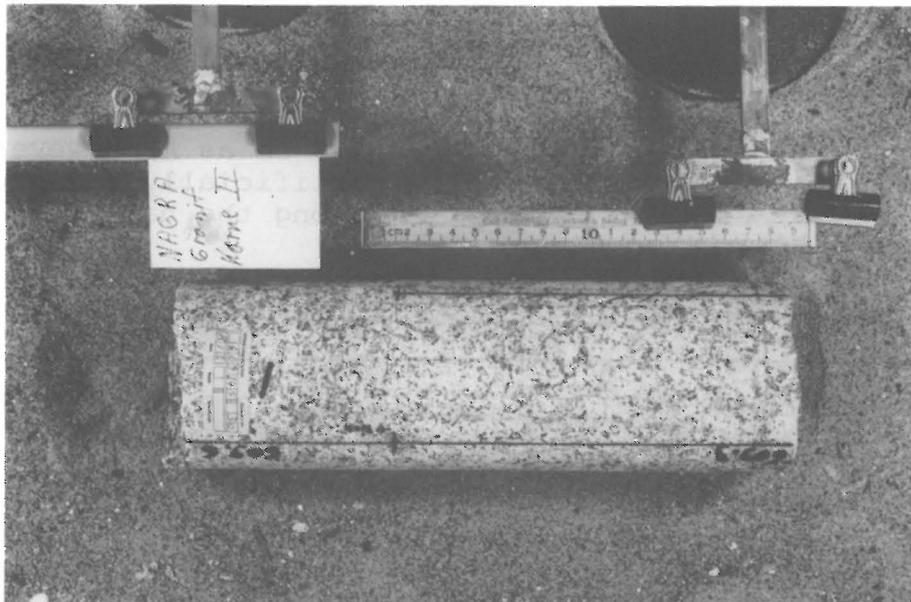


Fig. 2: Core sample no. II (Böttstein granite, depth 609.30 - 609.60 m): Solid core without visible cracks and fissures

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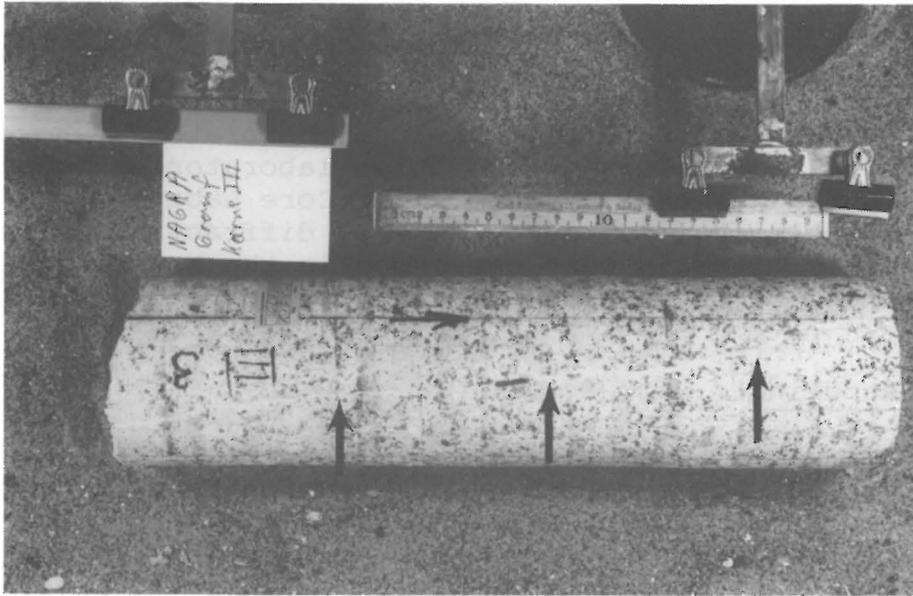


Fig. 3: Core sample no. III (Böttstein granite, depth 667.27 - 667.64 m): Solid core with large mineral/clay filled fissure over its entire length as indicated by the arrows

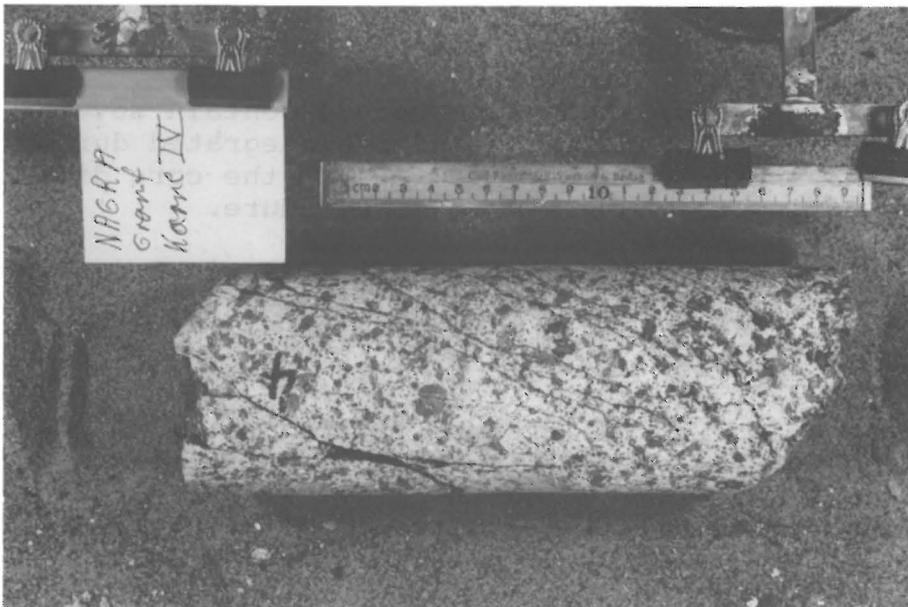


Fig. 4: Core sample no. IV (Böttstein granite, depth 696.50 - 696.72 m): Core with several cracks and fissures. This core exhibited a reddish colour, possibly owing to some hematite content

#### 4. EXPERIMENTAL

The major part of the experiments was carried out at the Chemistry Department (Risø National Laboratory), however, assistance was received from other laboratories; sample preparation was carried out by the Core Laboratory of the Danish Geological Survey, and X-ray diffraction analyses of fissure minerals were performed in the Metallurgy Department of the Risø National Laboratory.

##### 4.1 Core Handling

The drill core samples were cut in pieces of about 10 cm length, and each core piece was held together by hoop-clamps. At the Technological Institute selected core pieces were fractured. Throughgoing fractures in the long axis of the core were created with a hydraulic press. The core was mounted between two steel cones and a pressure of 1.5 t was exerted to generate the fracture.

At the Core Laboratory (DGU) the fractured and clamped cores were mounted in a pillar drill. Cylindrical sample cores were cut with a water cooled diamond-drill. The diameter of these cores were 1" (approx. 2.54 cm) and the length 2 - 4" (approx. 5 - 10 cm). The cutting direction was along the fracture in order to create sample cores (columns) exhibiting fissures through the total length of the core. Some cores were cut in solid non-fractured core material (cf. Table 2 and Figs. 1-4). The sample cores were removed from the drill cores, clipped with hoops and the ends were cut perpendicular to the long axis.

Some sample cores, which appeared to contain several natural cracks and fissures and disintegrated during handling. In the case of core no. IV, the core disintegrated during the sample-coring procedure.

An illustration of the core handling is given in Fig. 5.

##### 4.2 X-Ray Diffraction

A sample of pulverized fissure filling material was analyzed for its composition by X-ray diffraction applying the Cu-K $\alpha_{1,2}$  radiation ( $\lambda = 1.542 \text{ \AA}$ ). The obtained diffraction pattern was analyzed based on standard D-values for selected minerals.



Fig. 5: Clamped core with material from sample-coring

#### 4.3 Liquid Chromatography

The basic principles of liquid chromatography for migration studies have been reported previously, as well as the evaluation of the chromatograms /4/. However, for the present studies some modifications of the system have been introduced. Figures 6 and 7 illustrate the experimental set-up and a close-up of the compression module with the fissured column, respectively.

In the original approach /4/, where the pressure drop over the column which is determined by the permeability of the column and the flow rate through the column, could be controlled only by the flow rates available by the pump. The present system, however, allows adjustment (by the needle valve A, cf. Figs. 6 and 7) of the pressure drop over the column (measured by the manometer  $M_1$ , cf. Fig. 6) to the desired value. The eluent (delivered by the pump  $P_1$ , cf. Fig. 6) passes, at a relatively high flow rate (0.5 - 1.0 ml/min), over the column end surface in a small chamber of a volume of about 0.3 ml (cf. Fig. 7, B).

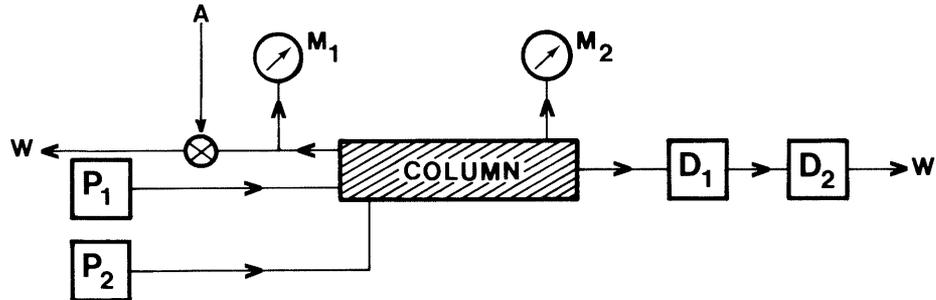


Fig. 6: Experimental set-up for liquid chromatography investigations. P<sub>1</sub>: pump to deliver the flow through the column, the pressure drop over the column being controlled by the needle valve A, and measured by the manometer M<sub>1</sub>. P<sub>2</sub>: pump to deliver hydrostatic pressure on the column, which can be measured by the manometer M<sub>2</sub>. D<sub>1</sub> and D<sub>2</sub> denote the radioactivity - and ultra-violet detection systems, respectively, w denotes waste

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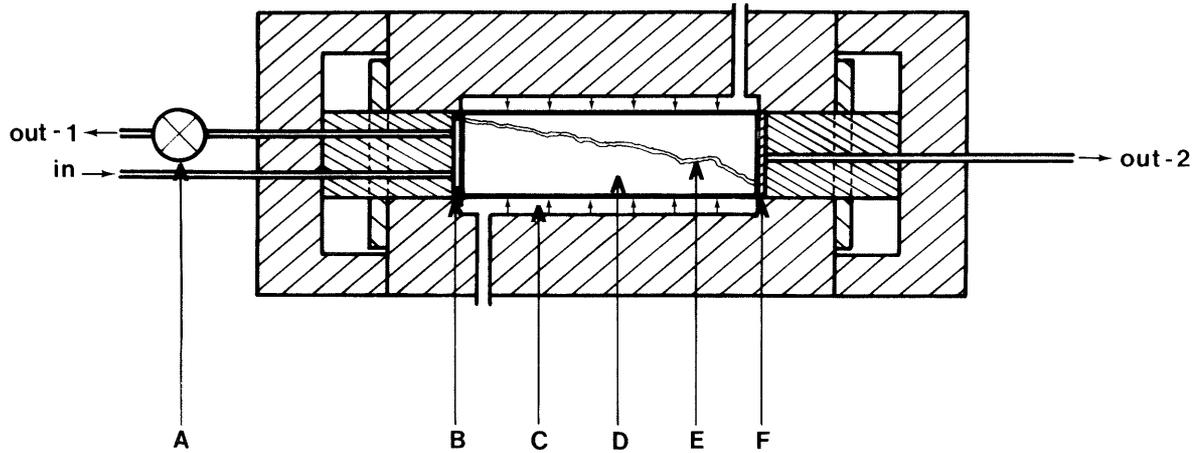


Fig. 7: Compression module with granite column. The column and the inlet/outlet joints are encapsulated in shrinkable polyethylene tubing. A: needle valve to control the pressure drop over the column ( $P_i$ ), B: 1 mm spacer, C: liquid (e.g. water) to maintain the hydrostatic pressure on the column ( $P_o$ ), D: column, E: fissure, F: filter paper

The amount of eluent passing through the column (to "out-2", cf. Fig. 7) relative to the amount passing through the valve to waste (denoted by "out-1", cf. Fig. 7) was determined by appropriate adjustment of the needle valve A. Apart from the possibility of arbitrary choice of pressure drops, the present set-up ensures that the full surface at the inlet side constantly is in contact with the eluent. Furthermore, the dead-volume of the system can be kept rather low (ca. 0.65 ml).

The eluent passing through the columns could be analyzed by one of two (or both) detector systems (cf. Fig. 6): D<sub>1</sub>: Radioactivity Monitor, D<sub>2</sub>: Ultra-violet detector. The hydrostatic pressure on the columns, which mimic the pressure conditions in the geological formation, is controlled by the pump P<sub>2</sub> and measured a the manometer M<sub>2</sub>.

From the chromatograms the following informations can be obtained using Q (volume flow rate through the column (cm<sup>3</sup>/s)), L (length of the column (m)), A (cross sectional area of the column (cm<sup>2</sup>)), h (pressure drop over the column (metre of water)), P<sub>i</sub> (pressure drop over the column (bar)), V<sub>0</sub> (elution volume of a non-retarded solute (ml)), and V<sub>r</sub> (elution volume of a retarded solute (ml)):

$$\text{Volume Porosity} \quad \epsilon = 100(V_0/A \cdot L) \quad \% \quad (1)$$

$$\text{Permeability} \quad K = Q \cdot L / A \cdot h \quad \text{cm/s} \quad (2)$$

which, in the present case (A = 5.07 cm<sup>2</sup> and L = 0.1 m), can be rewritten to

$$K = 2.00 \cdot 10^{-5} \cdot Q / P_i \quad \text{m/s} \quad (3)$$

$$\text{Sorption (expressed as retention factors)} \quad R_f = V_0 / V_r \quad (4)$$

The eluent applied for the present study (from Bad Säckingen Badquelle, vide supra) was throughout the experiment purged with helium in order to reduce the gas content (primarily oxygen) in the eluent /7/. It is important to note that the HCO<sub>3</sub><sup>-</sup> content in the eluent is not reduced by the He-purge.

All experiments were carried out by applying the appropriate hydrostatic pressure (P<sub>0</sub>) to the columns, i.e. P<sub>0</sub> being adjusted corresponding to the depth from which the core samples have been collected. An average value of 0.2 bar/m has been used. Hence, the hydrostatic pressures applied to the columns were 98-100 bar, 120-125 bar, and 135-140 bar for the columns of type I, II, and III, respectively (cf. Table 2).

In all cases, to ensure eluent flow through the columns and not along the outer surface, the pressure drop over the columns was kept below the value of the hydrostatic pressure, i.e.  $P_i < P_o$  (cf. ref. /4/).

The applicability of the modified LC-system was tested in a series of test experiments utilizing a column consisting of two half-cylindrical pieces of stainless steel, the resulting cylinder having the same dimensions as the granite columns (i.e.  $L = 0.1$  m,  $\phi = 2.5$  cm). It appeared that the system allows experiments to run for long periods of time regardless of the adjustment of the pressures  $P_i$  and  $P_o$ . Only very minor drift in the pressure drops  $P_i$  was observed.

## 5. RESULTS AND DISCUSSION

The liquid chromatography experiments were divided into four groups according to the nature of the columns:

- i) uncracked plugs without fissures (IIA)
- ii) intact, i.e. uncracked, plugs with "old" mineral/clay filled fissures (IB, IIIA)
- iii) plugs with "old" mineral/clay filled fissures artificially cracked along these fissures (IIIB)
- iv) artificially cracked plugs, i.e. plugs with "new" (fresh) fissures (IA, IIB, IIC).

### 5.1 Porosity and Permeability

Taking into account that water transport mainly takes place through macro fissures /1/, it does not seem surprising that we were unable to observe any groundwater flow (i.e.  $Q < 0.01 \mu\text{l}/\text{min}$  through the column IIA) even when applying pressure drops ( $P_i$ ) up to approx. 110 bar, corresponding to a hydraulic gradient of about 11'000, for 12 hours. On the other hand, it appeared at first sight somewhat surprising that also the columns containing "old" mineral/clay filled fissures (IB and IIIA) turned out as impermeable under the applied conditions, i.e.  $P_i$  up to approx. 90 bar for 18 and 24 hours for the two columns, respectively.

In contrast to this it appeared that the columns containing an artificially cracked "old" filled fissure (IIIB) exhibited a measurable permeability, although rather low. Within the first 4 - 5 hours after initiating the experiment ( $P_o/P_i \approx 140/80$ ), flow rates in the order of 1.8 to  $2.7 \mu\text{l}/\text{min}$  were determined. The corresponding permeability was calculated to be  $(7.6 \pm 2.5) \cdot 10^{-12} \text{ m/s}$  ( $2.4 \cdot 10^{-4} \text{ m/y}$ ). After approximately 18 hours in standing water without flow the flow rates were found to be significantly lower, i.e.  $0.5 - 0.9 \mu\text{l}/\text{min}$ , corresponding to a permeability equal to  $(2.2 \pm 0.7) \cdot 10^{-12} \text{ m/s}$  ( $6.8 \cdot 10^{-5} \text{ m/y}$ ). After further standing for approximately 18 hours, we were unable to detect any measurable flow through the fissure. Hence, we conclude that the observed decrease in permeability and the eventual apparent impermeability of the fissure most probably is a result of an effective "self-healing" of the fissure due to the contact with  $\text{HCO}_3^-$ -rich ground water (cf. Table 1).

Owing to the very low flow rates, which were determined by weighing the eluate coming through the column, we were unable to determine the porosity of these fissures, although it must be expected that  $\epsilon$  would turn out very low, i.e.  $\ll 0.5 \%$ .

In contrast to the above, the artificially cracked samples exhibited a reasonable high porosity, at least immediately after initiating the experiments. In the case of column IIB, the flow rates immediately after initiation was found to be 0.39, 0.23, 0.07, 0.05, 0.04, and 0.03 ml/min for  $P_o/P_i$  equal to 50/28, 75/28, 100/28, 125/28, 150/28, and 200/28, respectively. The first two settings yielded porosities of 0.77 and 0.39 %, respectively. The permeabilities were estimated to be  $4.64 \cdot 10^{-9}$ ,  $2.38 \cdot 10^{-9}$ ,  $8.33 \cdot 10^{-10}$ ,  $5.95 \cdot 10^{-10}$ ,  $4.76 \cdot 10^{-10}$ , and  $3.57 \cdot 10^{-10}$  m/s, respectively, clearly demonstrating the effect of the hydrostatic pressure  $P_o$ . The permeability further decreased gradually as a function of time to a value around  $2.2 \cdot 10^{-10}$  m/s ( $P_o/P_i \approx 130/75$ ,  $Q \approx 0.05$  ml/min), which could be maintained for several hours, corresponding to a porosity  $\epsilon \approx 0.12$  %. Prolongation of the experiment led to an increased instability of the  $P_i$  pressure, eventually leading to interruption of the experiment, at which time only very minor amounts of eluate were observed to pass through the column.

Visual inspection of the inlet side of the column after disconnecting the LC-system revealed an apparent formation of weathering products in the fissure, probably accounting for the instability in  $P_i$  due to clogging of the fissure.

An identical picture, as the above described, developed not surprisingly in the study of the column IIC. From an initial flow rate around 0.1 ml/min ( $P_o/P_i \approx 125/50$ ), corresponding to a permeability equal to  $6.7 \cdot 10^{-10}$  m/s, the flow rate was observed within approximately 8 hours to drop to ca 0.02 ml/min ( $P_o/P_i \approx 125/50$ ), corresponding to a permeability  $K = 1.3 \cdot 10^{-10}$  m/s. After disconnecting the flow through the fissure, the wet column was allowed to stand for one week ( $P_o/P_i \approx 125/0$ ), at which time the flow was reestablished, i.e.  $P_o/P_i \approx 125/50$ . It appeared that the flow rate was now approximately one order of magnitude lower, i.e. 2.6  $\mu$ l/min, corresponding to a permeability  $K = 1.7 \cdot 10^{-11}$  m/s. Further standing for one week without flow ( $P_o/P_i \approx 125/0$ ) caused apparently complete clogging of the fissure, as the column at this time appeared impermeable ( $P_o/P_i \approx 125/50$ ).

In order to explain the above described behaviour of the columns exhibiting "fresh" artificially introduced fissures, as well as the apparent impermeability of the columns with "old" filled fissures, we studied the composition of the fissure-filling material by application of X-ray diffraction. For this purpose a small sample of the fissure-filling material found in core no. III was used.

The analysis unambiguously revealed that the fissure-filling material predominantly consisted of two minerals: calcite ( $\text{CaCO}_3$ ) and quartz ( $\text{SiO}_2$ ). In addition some minor constituents were observed, the identification, however, being rather difficult, owing to low abundances of these minerals and consequently low intensities of the diffraction signals. However, the diffraction pattern suggests that muscovite and possibly clay minerals such as illite may be present. The findings are in good agreement with recent theoretical considerations, based on the geochemical modelling WHATIF /8/, which suggest that granites containing calcium, i.e. granites containing anorthite (Ca-feldspar) and/or plagioclase (Na/Ca-feldspar), especially in the presence of  $\text{CO}_3^{2-}/\text{HCO}_3^-$ -rich groundwaters, rapidly are converted into calcite and muscovite, the latter eventually being transformed into illite. On the other hand, the quartz fraction will be transformed only very slowly.

On this background we suggest that the observed clogging of the artificially introduced fissures may be a result of beginning weathering, a process which most probably will be accelerated owing to the relatively high  $\text{HCO}_3^-$  content in the groundwater (cf. Table 1). However, it can not completely be ruled out that precipitation of salts from the groundwater in the fissures may play a role in the clogging process. Furthermore, it appears reasonable that the "old" calcite/quartz filled fissures are of extreme low permeability. For comparison it can be mentioned that previously published data on cretaceous formations reveal rather low permeabilities (approx.  $10^{-9}$  m/s) despite of high porosities (approx. 40 %) /9/.

## 5.2 Sorption Characteristics

In the original approach, it was foreseen that the sorption of a series of radionuclides in different types of fissures should be investigated. However, due to the apparent impermeability of the unfissured plugs, as well as of the plugs exhibiting "old" calcite/quartz filled fissures, we unfortunately were limited to study sorption characteristics in plugs containing artificially introduced fissures. It must be strongly stressed that the sample material for these investigations has been very limited, indeed only sorption in the column IIB could be studied, i.e. the results are to be considered as highly preliminary and should, as such, be taken with extreme caution.

Before clogging of the fissure we were able to estimate the sorption of  $^{99}\text{Tc}$  (as pertechnetate),  $^{89}\text{Sr}^{2+}$ , and  $^{60}\text{Co}^{2+}$ . In all three cases retention was observed, the retention factors being estimated to be  $R_f(\text{TcO}_4^-) \approx 0.16$ ,  $R_f(\text{Sr}^{2+}) \approx 0.24$ , and  $R_f(\text{Co}^{2+}) \approx 0.04$ . It is not clear whether the sorption is a result of interaction with the "pure" granite surface, or it involves the calcite filling, although the latter most probably is the more important factor. Previous results /9/ have demonstrated the sorption ability of calcium carbonate for strontium and cobalt. It is also unclear whether technetium is sorbed as pertechnetate or in a reduced form. However, a Tc(VII) to Tc(IV) reduction is not directly intelligible taking the total absence of ferrous iron in the groundwater into account (cf. Table 1). In order to elucidate these questions, as well as to demonstrate the reproducibility of the data, more core material must be available.

## 6. CONCLUSION

The aim of the present study has been to elucidate the movement of possibly contaminated groundwater in fractured/fissured granite. For this purpose a series of granite samples from the Böttstein borehole has been characterized by determination of permeabilities, porosities, and sorption ability for selected radionuclides.

Based on the rather limited core material available for the present investigations, the following conclusion may tentatively be drawn:

- "Old" fissures are highly impermeable ( $K \lesssim 10^{-14}$  m/s) owing to an extremely dense filling by calcite and quartz, and possibly minor amounts of muscovite and illite. Hence, any possible transport of solutes in groundwater through these fissures will take place as aqueous phase diffusion solely. The effective diffusion coefficient is, however, most probably rather low (compared to that in pure water) due to size effects (cf. /9/). Furthermore, the filling material will cause an effective retardation of metal cations due to sorption, ion exchange, and precipitation reactions. In addition the retardation due to the possible involvement of micro fissures in the granite matrix should be considered (cf. /3/).
- "Old" calcite-filled fissures, artificially cracked along the natural fissures, may exhibit a pronounced tendency to rapid "self-healing" by contact with the  $\text{HCO}_3^-$ -rich groundwater.
- Artificially introduced fissures, i.e. "fresh" fissures, may within a reasonably short time-frame develop "self-healing" probably due to clogging by weathering products of the fresh granite surfaces, i.e. anorthite and plagioclas, enhanced by contact with the  $\text{HCO}_3^-$ -rich groundwater, eventually leading to impermeability of the fissures.
- Even artificially introduced fissures appear to exhibit sorption ability towards radionuclides, possibly influenced by the immediate formation of weathering or precipitation products.

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8. REFERENCES AND NOTES

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