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Mobilities of radionuclides in fresh and fractured crystalline rock

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This report concerns a study which was conducted for SKBF/KBS. The conclusions and viewpoints presented in the report are those of the author(s) and do not necessarily coincide with those of the client.

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SUMMARY

Sorption and migration of technetium, cesium and americium on fracture surfaces and fresh surfaces of granites taken from drilling cores from the Finnsjön and Studsvik areas and the Stripa mine are reported. The three elements were used as reference elements with different chemistry and behaviour in water; under the conditions used in the experiments technetium exists as the heptavalent TcO_4^- -ion, cesium as the non-complexed monovalent cation Cs^+ and americium as the strongly hydrolysed $\text{Am}(\text{OH})_x^{3-x}$ ($x=1-4$). The waters used were synthetic groundwaters representative of waters from the drilling holes. After the exposure of the fracture samples to spiked groundwater solutions for a period of three up to six months the penetration depths and concentration profiles were analysed and autoradiographs of cesium and americium distribution vs depth were taken. The sorption of technetium was found to be negligible. The transport of TcO_4^- depends on accessibility to fractures and micro-fissures in the rock. Cesium is sorbed through an ion-exchange process. Migration of cesium depends not only on the transport in water into fractures and micro-fissures, but also on migration through mineral veins with a high CEC. Americium is strongly sorbed on most solid surfaces and did not migrate significantly during the contact time of three months. The diffusivity in granite was found to be around $10^{-13} \text{ m}^2/\text{s}$ for cesium; preliminary values for technetium and americium were $10^{-12} \text{ m}^2/\text{s}$ and less than $10^{-16} \text{ m}^2/\text{s}$, respectively.

1. INTRODUCTION

The present Swedish concept for disposal of radioactive waste proposes storage in deep underground repositories in crystalline rock as a feasible method to avoid releases of hazardous radioactive material to the biosphere. A number of engineered and natural barriers will prevent the migration of radionuclides from the repository (1).

Radionuclides released from the waste would be transported by groundwater mainly in fractures, either in solution or as particulates, but diffusion into microfissures in the bedrock would also be expected (2). Crystalline rocks, e.g. granite, are composed of some major rock-forming minerals such as quartz, feldspars (orthoclase, plagioclase, etc.), micas (biotite, muscovite), amphiboles (hornblende) and pyroxenes. In old fractures, which have been exposed to groundwater, three main categories of minerals could be found, besides these major mineral components (3):

1. Weathering and alteration products originating from the host rock.
2. Precipitates and crystallization products from the aqueous solution.
3. Metamorphic products.

In order to allow a good description of radionuclide transport processes in the rock/groundwater environment the chemical properties of the pathways, as well as the radionuclide sorption properties and diffusion into the fracture surface coatings must be well characterized.

The sorption of cesium on natural fracture surfaces in granitic bedrock and the subsequent diffusion into the rock matrix as well as some preliminary results for technetium and americium diffusion in fracture surfaces are discussed in this report.

2. NATURAL FRACTURES IN GRANITE

The fracture mineralogy is not necessarily the same as the mineralogy of the bulk rock. Not only the rock-forming minerals and weathering and alteration products of these, but also precipitates and crystallization products of species transported by water from outside the rock massif could be present. Thus, the physical and chemical environment in the fractures depend both on the mineralogy of the rock and the groundwater composition in the rock.

2.1. Fracture mineralogy

The fractures studied were taken from drilling cores in granites from three different locations in Sweden (the Finnsjön area, the Studsvik area and from the Stripa mine) at depths down to 500 m. The mineralogy in the fractures, both in open and sealed fractures, were identified by chemical analysis and X-ray diffractometry (3). In sealed fractures the dominating minerals were quartz and calcite. Some of the calcites were young, possibly even of post-glacial origin. In open fractures the dominating minerals were calcite and prehnite. Fracture mineralogy at these locations is discussed in detail in ref. 3.

2.2. Groundwater chemistry

The groundwater compositions in the areas where the fracture samples were collected were analysed (4-7), and synthetic groundwaters representative of the waters in contact with the various core samples were prepared. The compositions of these synthetic waters are given in Table 1.

Table 1 Composition of synthetic groundwaters (mg/l) representative of the fracture samples studied.

Species	Finnsjön			Studsvik	Stripa
	6 ^a	7 ^b	8 ^c		
Na	959	274	320	90	49
K	16	16	12	2.8	0.59
Ca	554	134	37	31	14
Mg	69	16	12	9	0.23
SO ₄ ²⁻	205	45	42	36	4.9
HCO ₃ ⁻	123	280	263	195	86
Cl ⁻	2407	477	387	57	35
SiO ₂ tot	17	13	12	11	5.5
NO ₃ ⁻	6	-	2.0	-	-
F ⁻	0.97	1.75	2.3	-	-

pH	7.7	8.1	8.3	7.3	8.9

^a Level below surface: 184 m

^b Level below surface: 322 m and 511 m

^c Level below surface: 115 m

3. STUDIED RADIONUCLIDES AND SYSTEM CHARACTERIZATION

For the studies of uptake on fracture surfaces and subsequent diffusion into the rock matrix the elements technetium, cesium and americium were selected. These elements, added as ⁹⁹Tc, ¹³⁷Cs and ²⁴¹Am, could serve as reference elements probably having different sorption and mobility in rock because of their different chemistry.

Technetium and cesium are both fission products. Technetium exists as the negatively charged TcO₄⁻-ion and cesium as the positively charged Cs⁺-ion in aerated natural groundwaters. Neither technetium nor cesium would form complexes or be hydrolyzed in environmental aerated aqueous systems.

Americium, which is trivalent, is highly hydrolyzed, but also forms strong complexes with carbonate (8).

For the sorption and diffusion studies of technetium, cesium and americium about 80-90 samples with natural fracture surfaces were selected from the three areas. In Table 2 some data on location and mineralogy are summarized.

4. EXPERIMENTAL

The drilling cores containing the fractures were sawn into rock tablets of an approximate size of 2x2x2 cm in such a way that one of the square surfaces of the rock specimen was a natural fracture surface. The tablets were coated with a plastic resin except on the natural fracture surface and submerged in synthetic groundwater containing one of the nuclides ^{99}Tc , ^{137}Cs or ^{241}Am at trace concentration ($<10^{-6}$ M).

The total uptake of the nuclides was measured as a function of time (up to 6 months) by counting the decrease in radioactivity in the water (with correction for the sorption on the vessel walls).

4.1. Penetration depths into the rock matrix

After the exposure of the rock tablets to the spiked groundwater the tablets were removed and washed once with distilled water and dried. The dry tablets were fixed to a metal rod, which was fitted into a special grinding equipment (C.f. Figure 1, (9)). With this device ca. 0.1 mm thick successive layers of the fracture surface were removed. The activity on the fresh surface as well as in the removed powdered layer was measured. Concentration profiles (concentration vs penetration depth) in the three granites were thus obtained.

Table 2 Characterization of the studied systems.

Radio-nuclide ^a	Fracture surface	Level (m)	Mineralogic composition	Synth. water	Contact time(days)
⁹⁹ Tc	Finnsjön	526.5	b	Finnsjön 7	85
	Finnsjön	72.0	calcite, quartz prehnite, laumontite	Finnsjön 8	167
	Studsvik	29.3	calcite, quartz chlorite, (biotite)	Studsvik	167
	Stripa(1:5)	-	quartz ^c , feldspars ^c , epidote, chlorite, muscovite	Stripa	167
¹³⁷ Cs:	Finnsjön	309.0	calcite, (prehnite)	Finnsjön 6	167
	Finnsjön	318.8	quartz, plagioclase, biotite, chlorite, calcite ^d	Finnsjön 7	132
	Finnsjön	72.0	calcite, quartz, prehnite, laumontite	Finnsjön 8	167
	Studsvik	5.4	calcite, quartz (biotite), palygorskite	Studsvik	132
	Studsvik	104.2	smectite, (calcite), (quartz) ^e	Studsvik	167
	Stripa ^e	-		Stripa	85
	Stripa(A:IN)	-	calcite, epidote, chlorite, quartz, plagioclase, (biotite)	Stripa	167
	Stripa(MS:2)	-	quartz ^c , feldspars ^c , calcite, epidote, muscovite, chlorite	Stripa	167
	Stripa(1:1)	-	quartz ^c , feldspars ^c , epidote, chlorite, muscovite		
²⁴¹ Am:	Studsvik ^f	183.1	f	Studsvik	85

^a Initial nuclide concentration in the aqueous phase $<10^{-6}$ M.

^b Reference sample of fresh granite; quartz, orthoclase, plagioclase and biotite.

^c Microbreccia of mainly quartz and feldspars.

^d Calcite in the latest precipitation.

^e Reference sample of fresh polished granite; quartz, plagioclase, microcline, muscovite and hornblende.

^f Reference sample of fresh granite; quartz, plagioclase, hornblende and biotite.

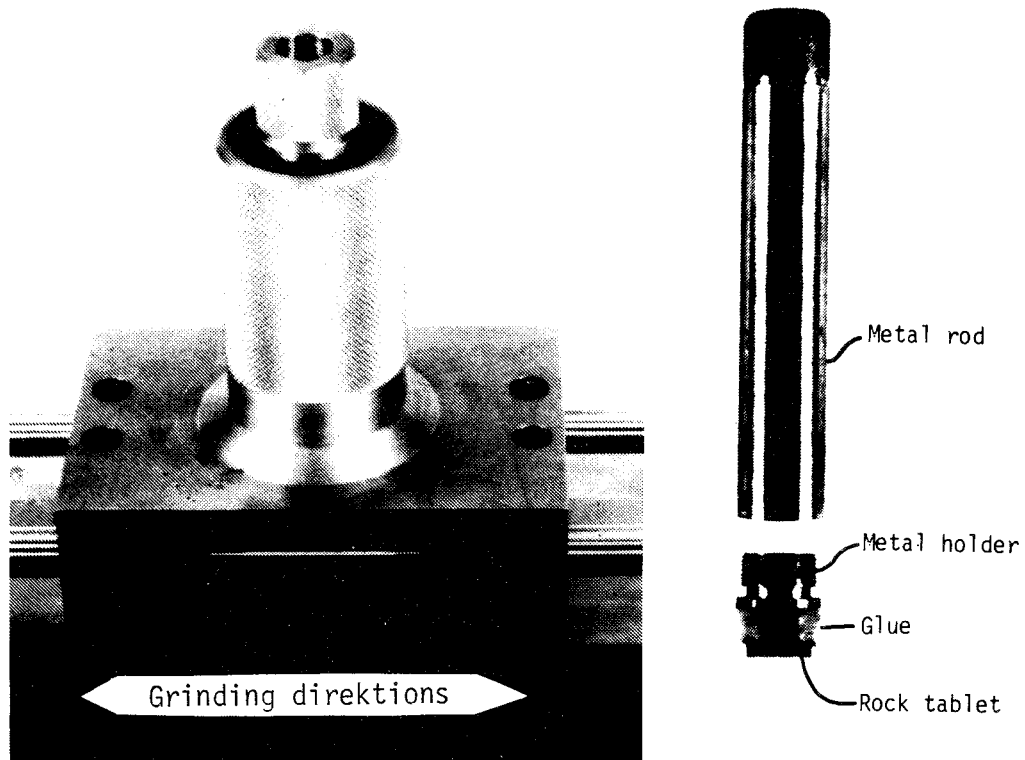


Figure 1. The rock surface grinding equipment.

4.2. Surface distribution studies by autoradiography

In order to distinguish the sorption on the various minerals in the rock, autoradiographs on the fracture surfaces were taken after each grinding cycle. The film used was Ilford Technical Film, sensitive for all three types of radiation, alfa, beta and gamma. Kodak D-72 was used as a developer and Kodak F-24 as fix. The exposure times were from 5 h up to several days depending on the amount of radioactivity on the surface. Before each exposure the active surface was coated with a very thin plastic film, and then pressed against the photographic film as illustrated in Fig. 2.

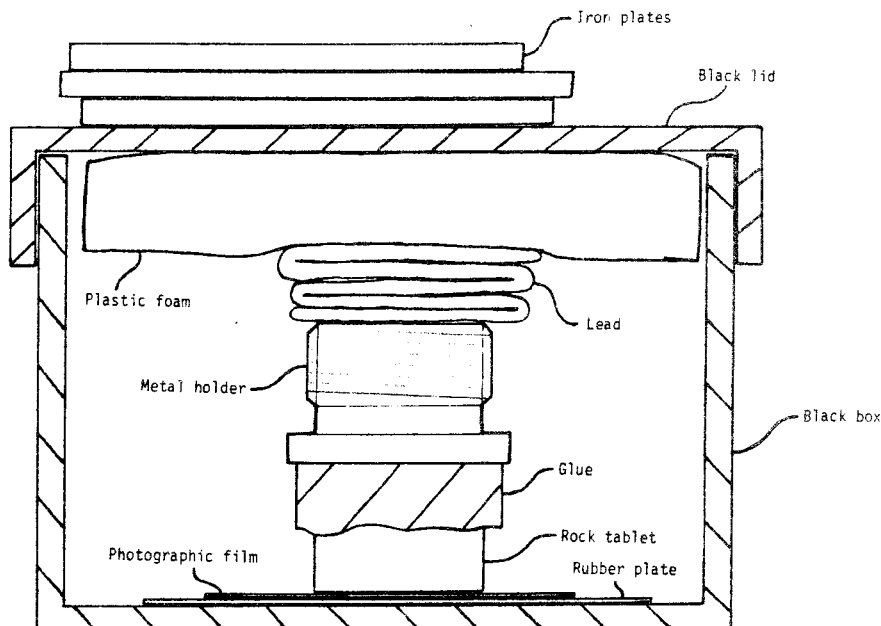


Figure 2. Experimental method used for autoradiography studies.

5. RESULTS AND DISCUSSIONS

5.1. Distribution coefficients

The distribution coefficients, K_d (moles/kg solid per moles/kg liquid), for technetium, cesium and americium, as obtained in conventional batch measurements, are given in Table 3. For the negatively charged TcO_4^- -ion almost no sorption would be expected on granitic minerals. The mobility in a natural fissure would thus be related to the water transport rate in pores and microfissures.

The sorption of cesium is mainly a cation exchange process (10). Minerals with a high cation exchange capacity (CEC) also exhibit a high sorption of cesium. Many fracture minerals have a high CEC as well as the rock-forming minerals hornblende and the micas (11). Thus, cesium would sorb strongly on most minerals found in fractures, as indicated in Table 3 (10,11).

Americium sorbs strongly on all minerals fairly independent of CEC, which is significant of a non-specific physical adsorption process.

Table 3 Distribution coefficients (K_d) for cesium and americium on major rock-forming minerals and common fracture minerals of granite. Contact time 1 day, pH 8-8.5 (12-14). For Tc $K_d = 0$ is assumed.

Mineral	Distribution coefficient (kg/m^3) $\times 10^3$	
	Cs	Am
Quartz	1	1260
Plagioclase	11	
Microcline	177	3980
Hornblende	214	15900
Biotite	650	12600
Muscovite	600	12600
Calcite	7	8910
Epidote	19	3160
Chlorite	129	12600
Granite:		11200
-Finnsjö	490	
-Studsvik	1110	
-Stripa	150	

5.2. Migration into fracture surface

The depth penetration was evaluated by counting the activity of the layers removed in the grinding, and also by measuring the remaining total activity in the rock. Diffusion into the rock was calculated according to the equation:

$$c = c_0 \operatorname{erfc} \frac{x}{2\sqrt{D_a t}} \quad (1)$$

where c = concentration (mol/m^3) at distance x (m), c_0 (mol/m^3) = the concentration at $x = 0$, t = time (s) and D_a the apparent diffusivity (m^2/s) (15). The diffusivities for technetium, cesium and americium are given in Table 4 and some diffusion curves in Fig. 3 and 4.

5.2.1. Technetium migration

No retardation of technetium caused by sorption on the rock would be expected (c.f. Table 3). Thus the transport depends largely on the porosity of the rock. Preliminary value for the diffusivity of technetium in Finnsjön granite is $1.5(+0.8) \times 10^{-12} \text{ m}^2/\text{s}$.

5.2.2. Cesium migration

A considerable migration of cesium into the rocks was found already after a couple of months diffusion time (Fig. 4). The apparent diffusivity D_a in granite was of the order $10^{-13} \text{ m}^2/\text{s}$. By taking the sorption into account according to the equation

$$D = D_a(1+k_d g) \quad (2)$$

(15), where g is density of the rock ($2.65 \times 10^3 \text{ kg}/\text{m}^3$ for granite (16)), a diffusivity D of the order of $10^{-10} \text{ m}^2/\text{s}$ was obtained (c.f. Table 4). This corresponds to a much higher transport rate than would be expected for a non-retarded diffusion process. Obviously there are different transport mechanisms for cations and anions. A possible explanation for the observed diffusivity is given in 5.3.

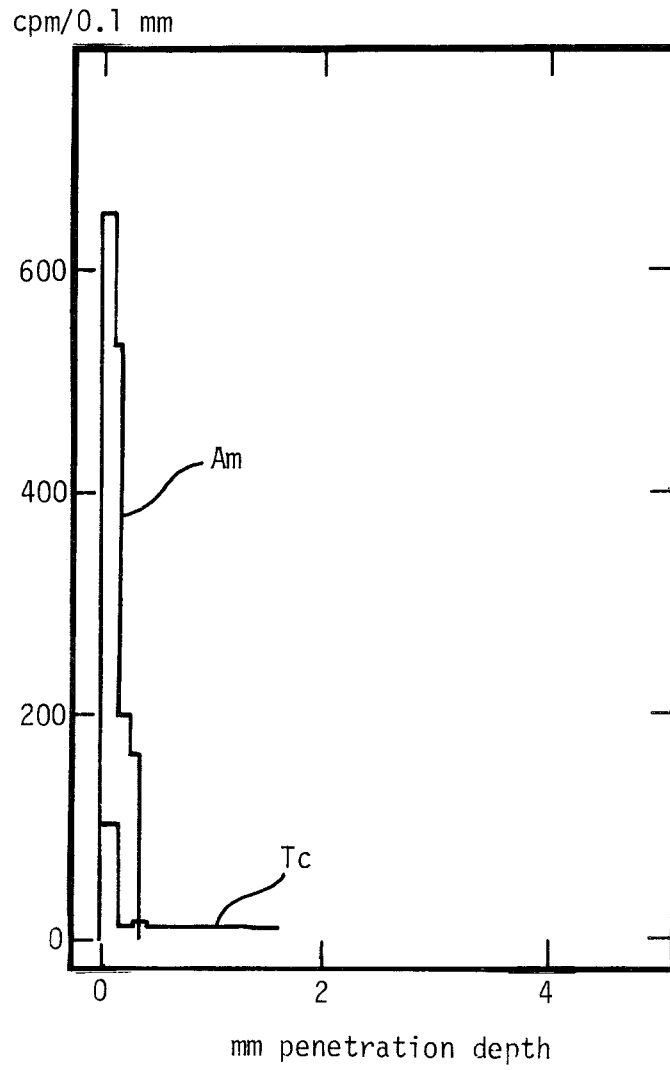


Figure 3. Penetration depth for Tc in Finnsjön granite and Am in Studsvik granite (85 days diffusion time).

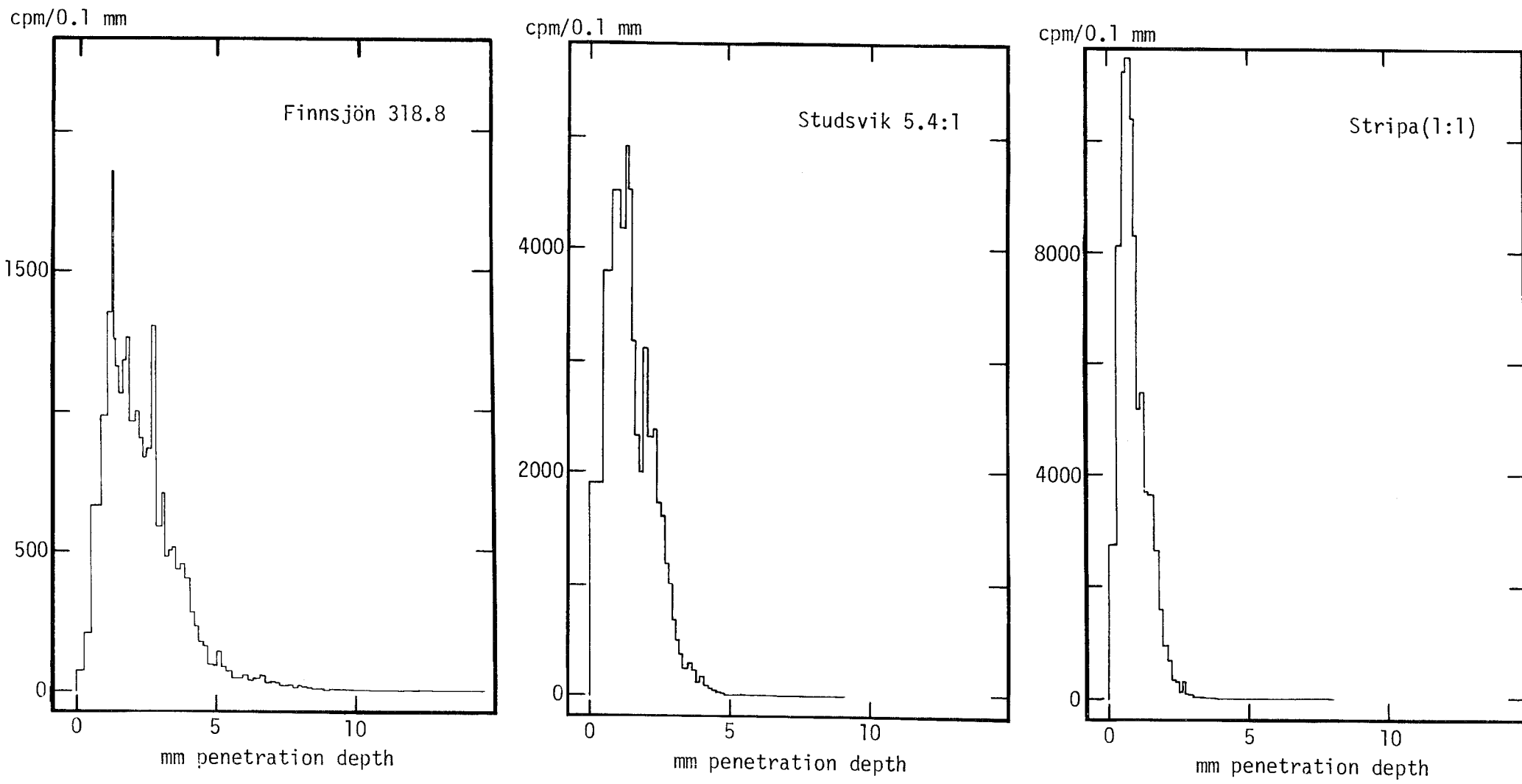


Figure 4. Penetration depth for cesium in granite (132 days diffusion time).

5.2.3. Americium migration

The migration of Am is very slow; within a diffusion time of almost three months the total penetration depth was less than 0.5 mm. Most of the americium was not significantly transported into the rock below the non-planar fracture surface. For a transport less than 0.1 mm into the surface in 85 days contact time the apparent diffusivity D_a would be of the order of $10^{-16} \text{ m}^2/\text{s}$. From eqn. (2) a diffusivity D of less than $10^{-11} \text{ m}^2/\text{s}$ was calculated. This is of the same order of magnitude as for technetium.

5.3. Autoradiographs

Autoradiographs of the fracture surfaces containing cesium and americium are shown in Fig. 5 and 6. The autoradiographs clearly show that the sorption of cesium is largely concentrated on the minerals with high CEC (Fig. 5). Black minerals on the photograph (the high-capacity minerals, i.e. biotite, hornblende and chlorite) are also black on the autoradiographs. Thus, cesium appears to migrate through fracture coating minerals with high CEC into the underlying rock. The migration into the rock follows the high-capacity mineral veins (e.g. biotite and hornblende). The sorption on fractures coated with calcite is much lower than on chlorite/epidote coated fracture surfaces (17).

Americium is strongly sorbed on most minerals, but has a higher sorption on high-capacity minerals (Fig. 5). However, the transport into the rock underlying the fracture surface coating is minor. According to the autoradiographs in Fig. 6 a removal of a 0.1 mm thick layer of the surface would also remove essentially all the americium activity.

Table 4 Measured apparent diffusivities (D_a) and calculated diffusivities (D) according to eqn. (2).

Element	Granite	Time d	D_a^a m^2/s	D m^2/s
^{99}Tc	Finnsjön	85	$1.5(+0.8) \times 10^{-12}$ c	$1.5(+0.8) \times 10^{-12}$
^{137}Cs	Finnsjön	132	$2.1(+1.0) \times 10^{-13}$	$2.7(+1.3) \times 10^{-10}$
	Studsvik	132	$7.9(+4.2) \times 10^{-14}$	$2.3(+1.2) \times 10^{-10}$
	Stripa	132	$9.5(+5.2) \times 10^{-14}$	$3.8(+2.1) \times 10^{-11}$
^{241}Am	Stripa ^b	85	$3.1(+1.3) \times 10^{-13}$	$1.2(+0.5) \times 10^{-10}$
	Studsvik	85	$<10^{-16}$ d	$<10^{-11}$

^a D_a calculated from the measurements of the activity removed by grinding.

^b Reference sample of a fresh polished surface.

^c Preliminary values due to not completed penetration depth analysis.

^d Preliminary values due to short diffusion time leading to a too short penetration depth.

6. RETENTION IN THE ROCK

The retention behaviour of the three elements discussed in this study indicates different transport mechanisms.

Technetium as TcO_4^- would not be chemically retarded in relation to the groundwater movement.

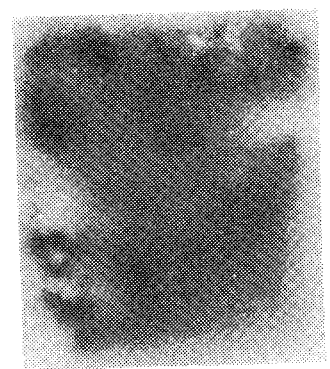
Americium would interact strongly with all exposed rock components through physical adsorption processes, fairly independent of the mineralogic composition.

The migration of cesium depends largely on the fracture mineralogy in water carrying fractures. The retention is minor on fracture surfaces with low-capacity minerals. However, in fractures coated with high-capacity fracture minerals or in veins of high-capacity

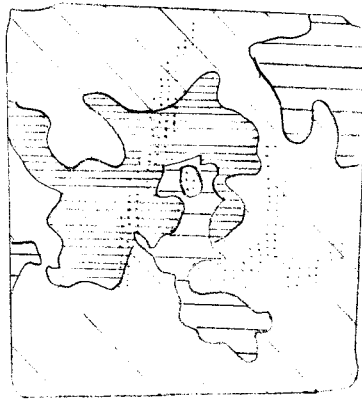
Photograph (Fi7 318,8:1) Mineralogic composition



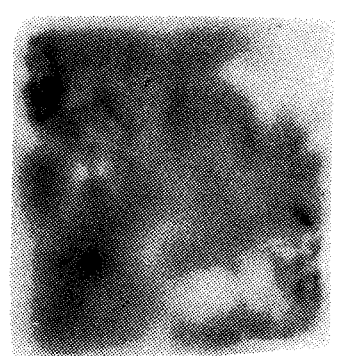
Autoradiograph

















Photograph (Fi8 72,0:1) Mineralogic composition



Autoradiograph

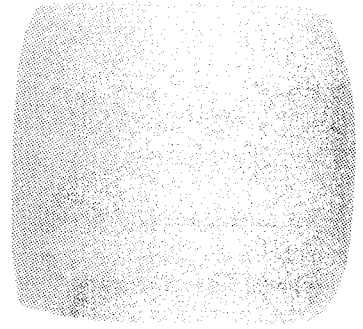
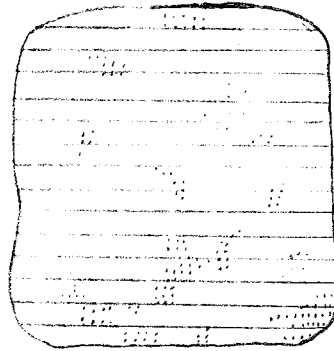
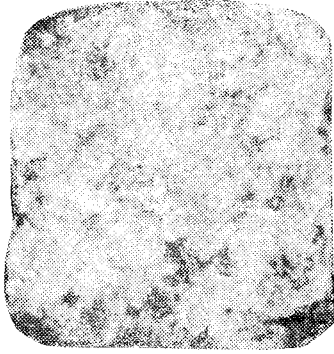


- | | |
|---|--|
|  = Biotite |  = Calcite |
|  = Pyrite |  = Biotite and chlorite |
|  = Feldspars and minor quartz grains |  = Clay mineral (smectite) |
|  = Major quartz grains |  = Calcite, quartz and feldspars |
|  = Fragments of quartz and feldspars ^a |  = Chlorite and epidote |
|  = Sealing of chlorite, epidote and muscovite ^a |  = Quartz and calcite |
| |  = Quartz and prehnite |
| |  = Laumontite |

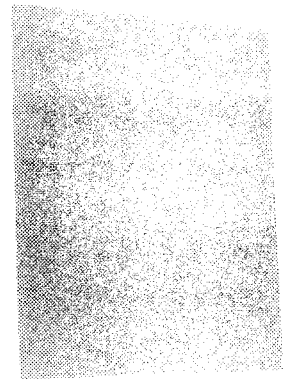
^aMicrobreccia

Figure 5. Photographs and mineralogic composition of the studied granite surfaces. The autoradiographs show the sorption of ^{137}Cs and in the last two autoradiographs the sorption of ^{241}Am .

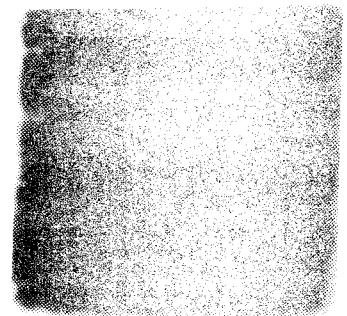
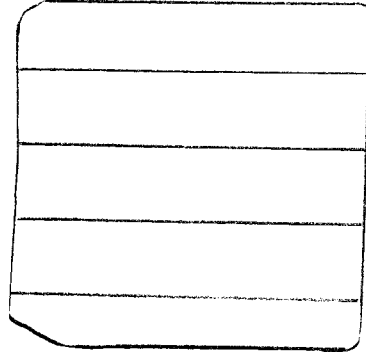
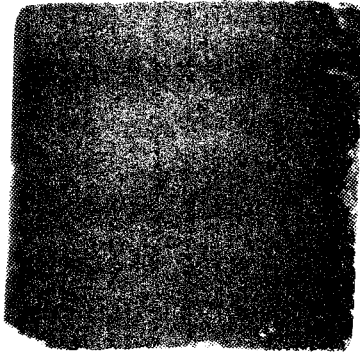
Photograph (Fi8 358,1:1) Mineralogic composition Autoradiograph



Photograph (Stu 5,4:1) Mineralogic composition Autoradiograph



Photograph (Stu 104,2) Mineralogic composition Autoradiograph



Photograph (Stu 183,1:1) Mineralogic composition Autoradiograph

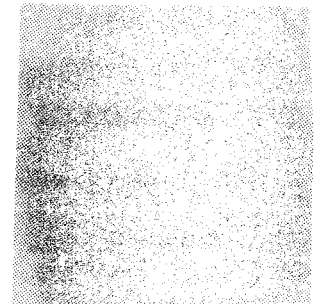
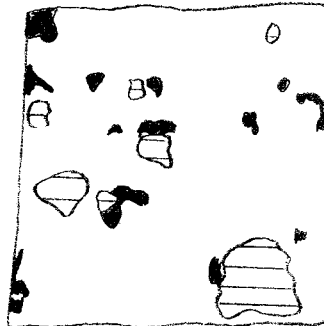
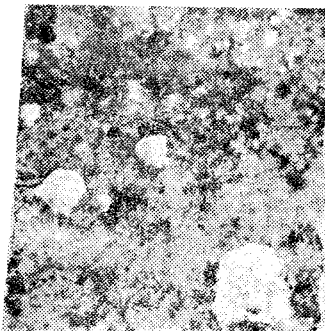


Figure 5. Cont.

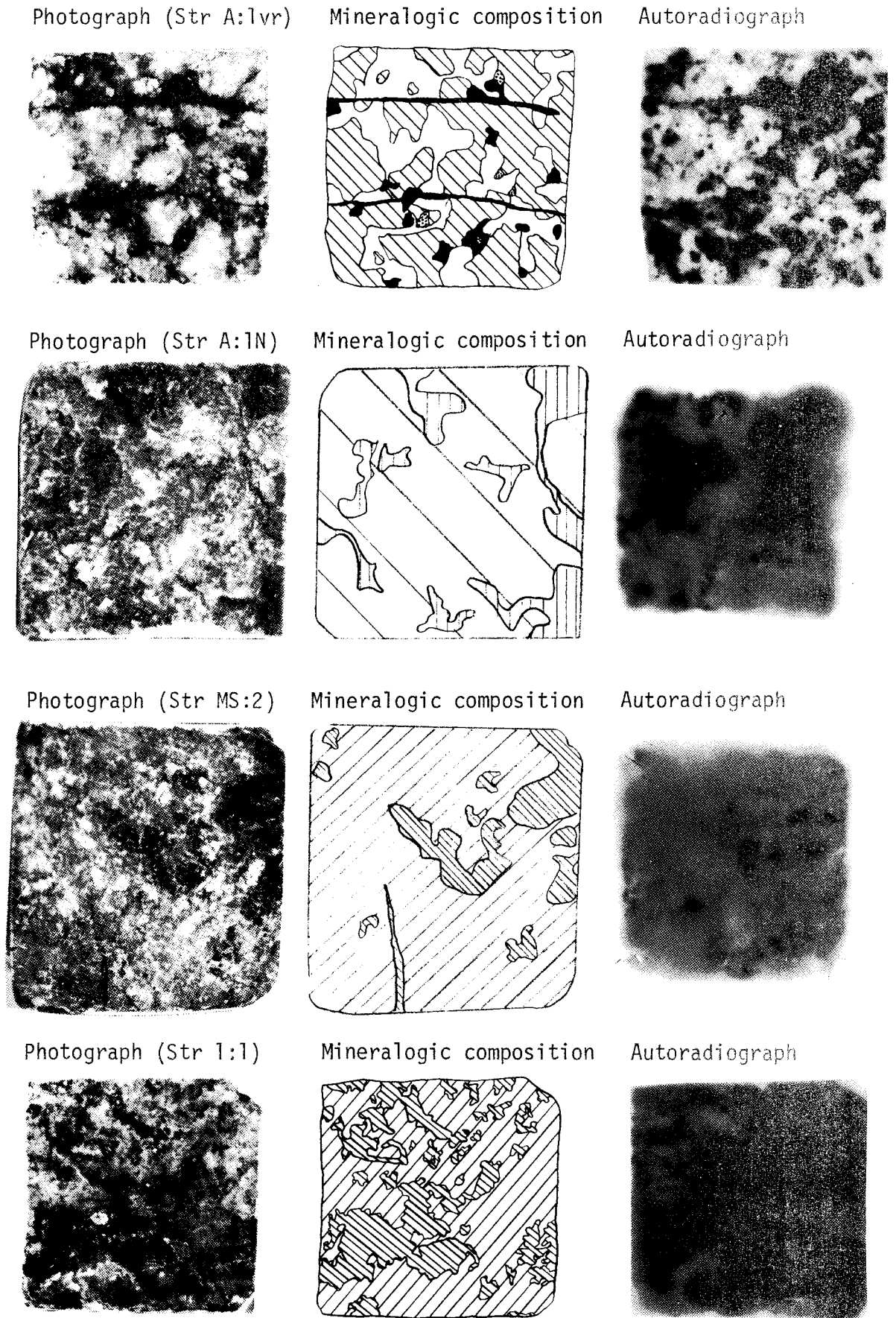
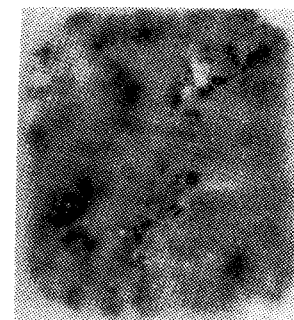
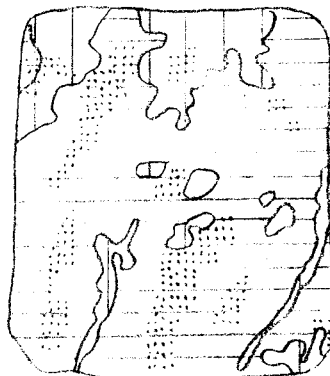


Figure 5. Cont.

Photograph (Fi7 526,5:3) Mineralogic composition Autoradiograph



(Stu 183,1:1r) Mineralogic composition Autoradiograph

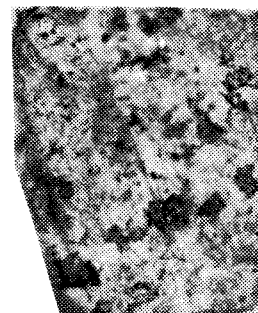
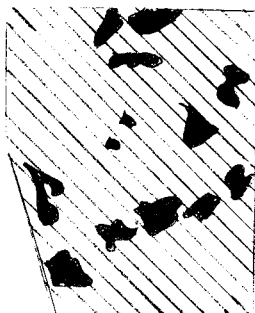
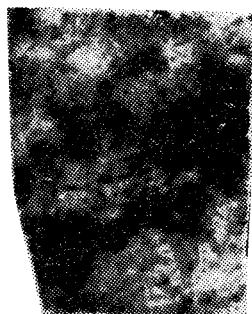
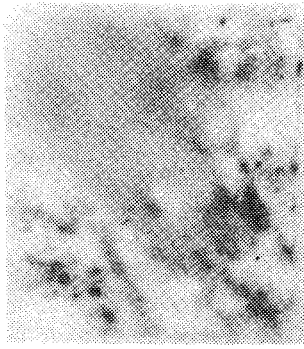


Figure 5. Cont.

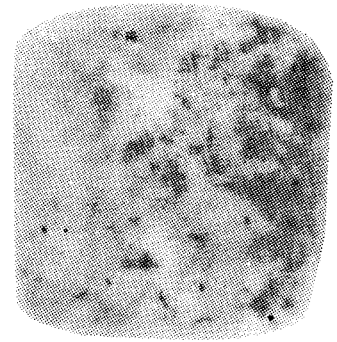
Autoradiographs



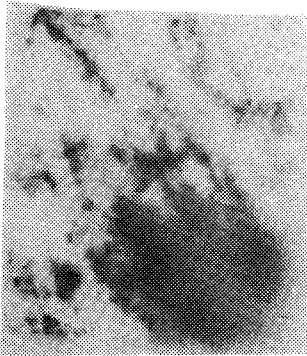
(1.14)



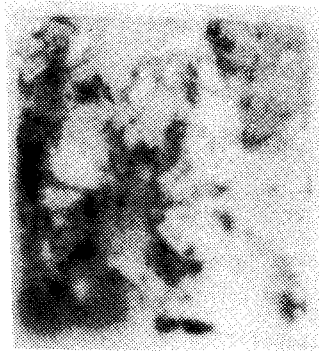
(1.25)



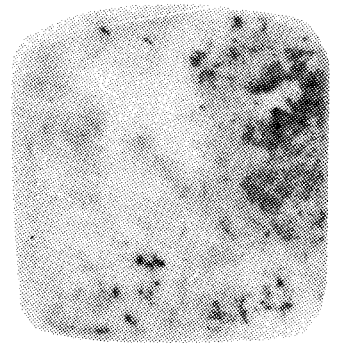
(1.47)



(1.97)



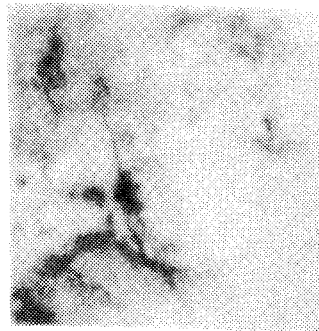
(1.95)



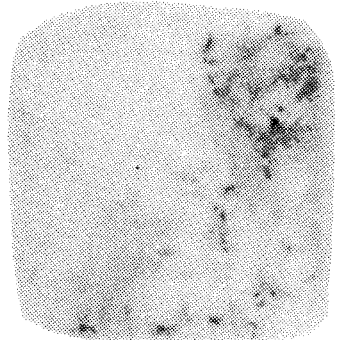
(2.53)



(2.89)



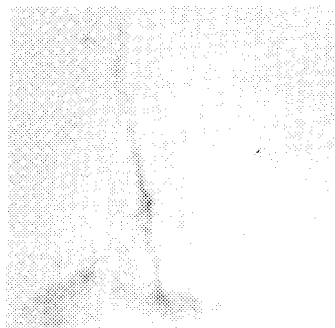
(3.13)



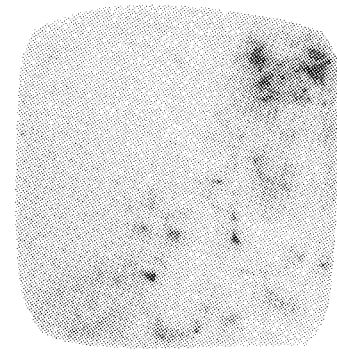
(3.38)



(3.74)



(4.05)



(4.83)

Cs, Fi7 318,8:1

Cs, Fi8 72,0:1

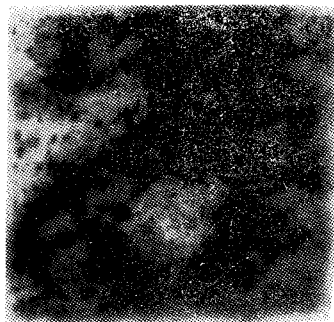
Cs, Fi8 358,1:1

Figure 6. Autoradiographs showing diffusion of ^{137}Cs and ^{241}Am in granite. Penetration depth in mm given in parenthesis.

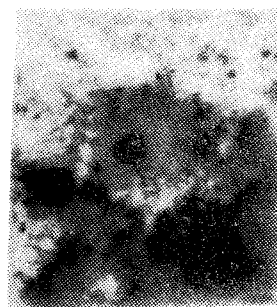
Autoradiographs



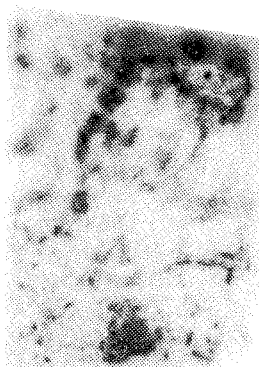
(1.09)



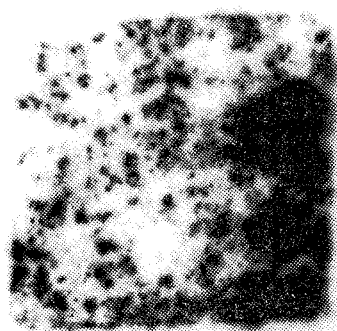
(0.10)



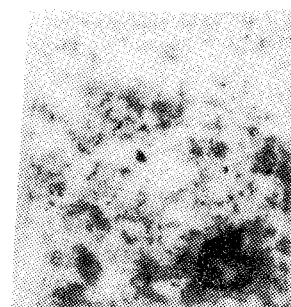
(0.87)



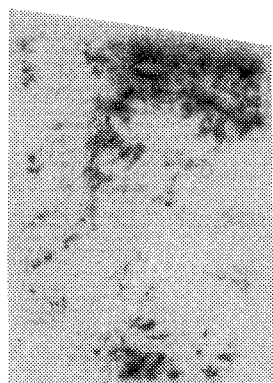
(2.08)



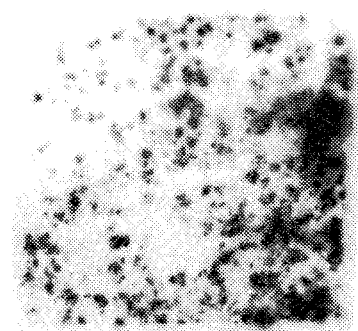
(0.62)



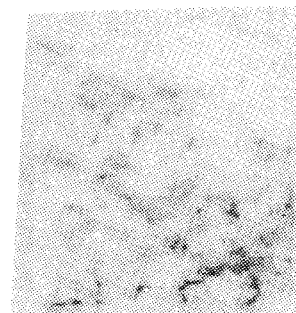
(1.69)



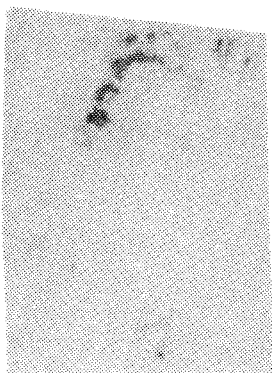
(2.56)



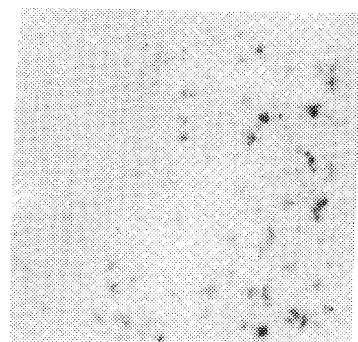
(1.07)



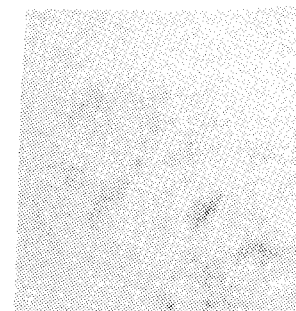
(2.92)



(3.39)



(2.14)



(4.97)

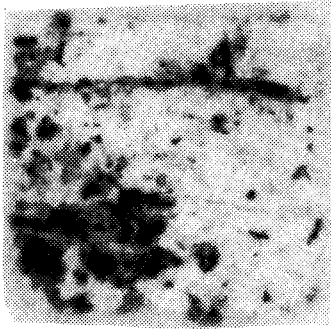
Cs, Stu 5,4:1

Cs, Stu 104,2

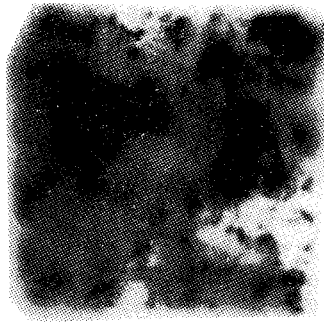
Cs, Stu 183,1:1

Figure 6. Cont.

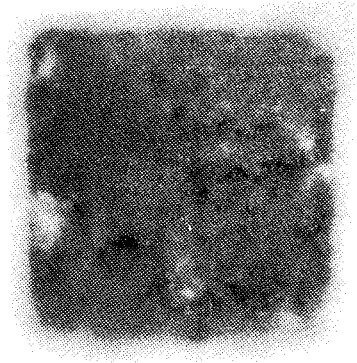
Autoradiographs



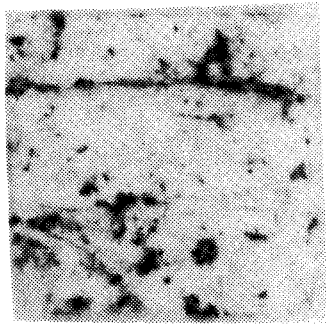
(0.28)



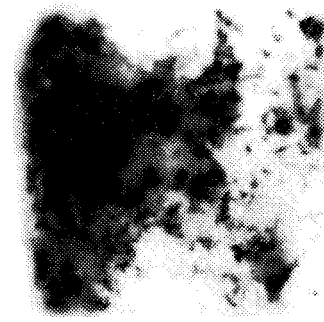
(0.51)



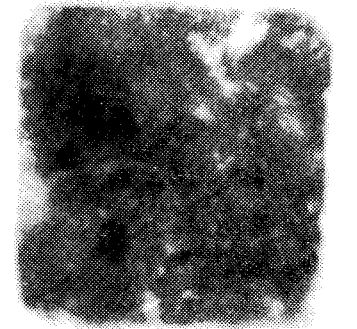
(0.50)



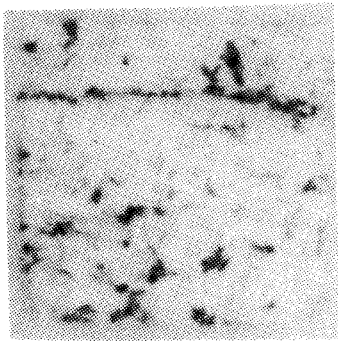
(0.66)



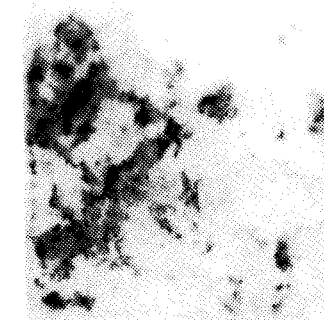
(1.12)



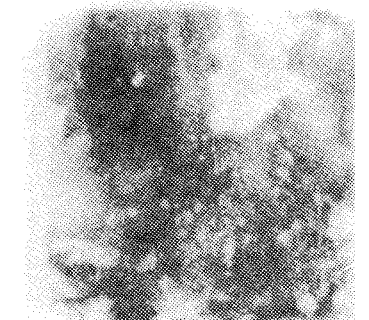
(0.96)



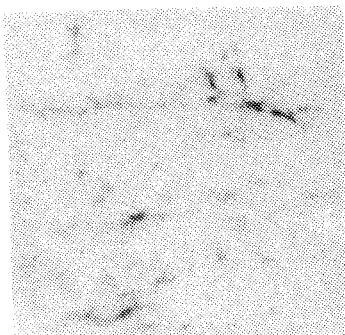
(0.90)



(1.50)

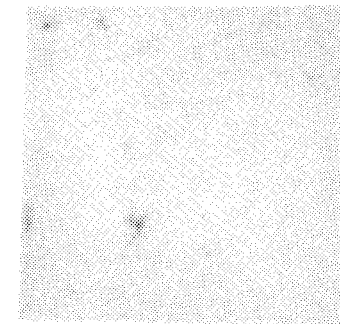


(1.32)



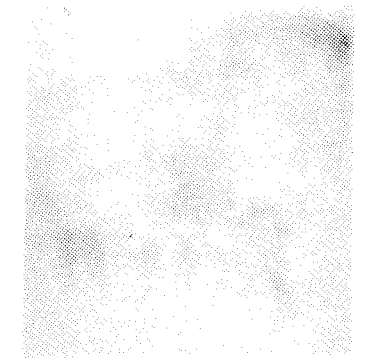
(1.64)

Cs, Str A:1vr



(2.32)

Cs, Str A:1N

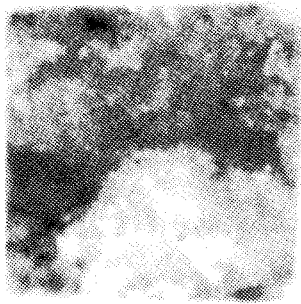


(2.19)

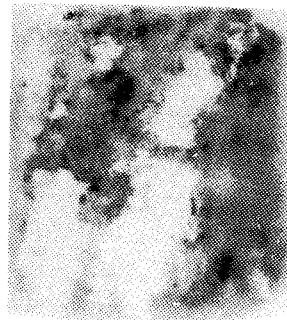
Cs, Str MS:2

Figure 6. Cont.

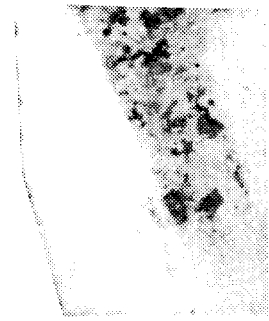
Autoradiographs



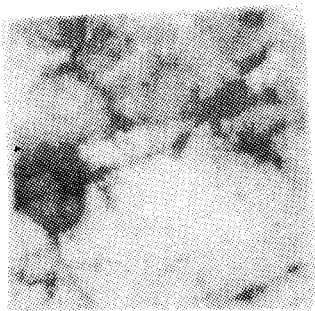
(0.64)



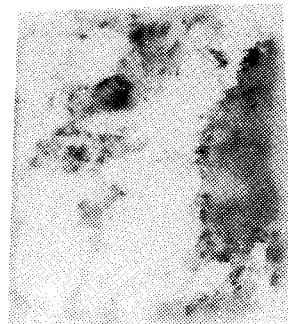
(0.48)



(0.10)



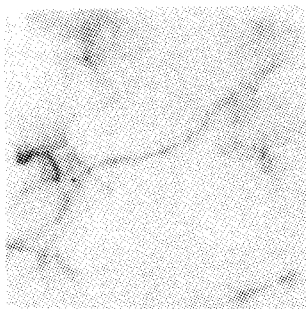
(1.43)



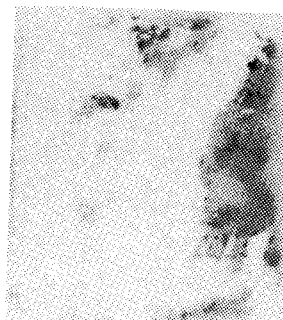
(0.70)



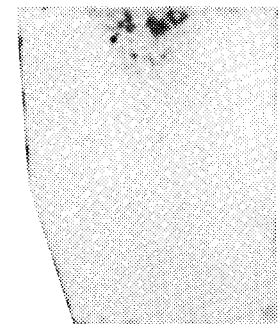
(0.15)



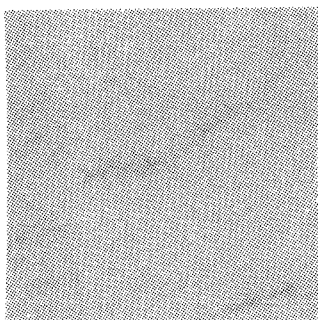
(1.93)



(0.89)

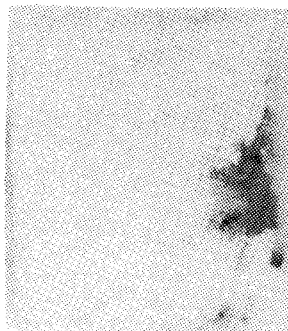


(0.25)



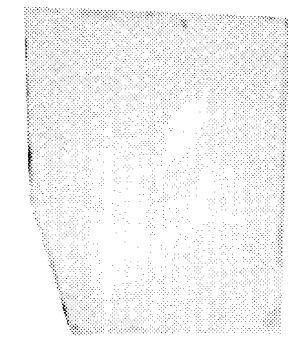
(2.69)

Cs, Str 1:1



(1.31)

Am, Fi7 526,5:3



(0.35)

Am, Stu 183.1:1r

minerals a sorption and subsequent transport inwards from the exposed surfaces will take place. For the transport through the rock in a porous, filled fracture or in a mineral vein containing high-capacity minerals the retention would roughly be given by

$$t = a Z^2/D_a \quad (3)$$

where t = retention time (s), Z = barrier thickness (m), D_a = apparent diffusivity (m^2/s) and a a constant. The retention time corresponding to a migration of e.g. 1 m would be around 10^4 years (assuming breakthrough of 5% of initial concentration, $a=0.1$).

7. CONCLUSIONS

Under oxidizing conditions, the transport of technetium (as TcO_4^-) will take place in water-bearing fractures and in micro-fissures in the bulk rock, and be very little affected by chemical interactions with the rock. (However, under reducing conditions technetium would be reduced to the tetravalent state (18), and would probably behave like americium).

Cesium would not be hydrolyzed or form complexes with the common anions in groundwaters. In fractures with minerals having a high CEC a large fraction of the cesium will be sorbed due to ion exchange reactions on the fracture surfaces and transported through high-capacity minerals further into the bulk rock, thus significantly decreasing the overall transport rate.

Americium would be highly hydrolyzed and exhibit a low total solubility in natural waters (8). Americium hydroxide species will be strongly sorbed on solid surfaces and be highly immobilized. In contrast to cesium there seems to be no indications of significant transport through high-capacity minerals into the rocks. The exposure time has been too short in the present experiments to allow significant uptake by the rock, except on the outer water exposed surface. Other experiments, with considerably longer exposure times, are in progress.

8. ACKNOWLEDGEMENTS

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