

POSIVA 98-05

Sorption of iodine on rocks from Posiva investigation sites

Seija Kulmala, Martti Hakanen
University of Helsinki
Department of Chemistry
Laboratory of Radiochemistry

Antero Lindberg
Geological Survey of Finland

May 1998

POSIVA OY

Mikonkatu 15 A, FIN-00100 HELSINKI, FINLAND

Phone (09) 2280 30 (nat.), (+358-9-) 2280 30 (int.)

Fax (09) 2280 3719 (nat.), (+358-9-) 2280 3719 (int.)

ISBN 951-652-043-X
ISSN 1239-3096

The conclusions and viewpoints presented in the report are those of author(s) and do not necessarily coincide with those of Posiva.

Radiokemian laboratorio
Kemian laitos
PL 55 (A.I.Virtasen aukio 1)
00014 Helsingin yliopisto

Helsinki, 8.5.1998

Tutkimussopimus POSIVA 9564/97/MVS

HEIKOSTI SORBOITUVIEN NUKLIDIEN SORPTIO

Raportti POSIVA-98-05

SORPTION OF IODINE ON ROCKS FROM POSIVA INVESTIGATION SITES

Helsingin yliopisto
Kemian laitos
Radiokemian laboratorio



Timo Jaakkola, professori

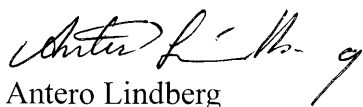


Seija Kulmala



Martti Hakanen

Geologian tutkimuslaitos



Antero Lindberg



Posiva-raportti – Posiva Report

Posiva Oy
Mikonkatu 15 A, FIN-00100 HELSINKI, FINLAND
Puh. (09) 2280 30 – Int. Tel. +358 9 2280 30

Raportin tunnus – Report code

POSIVA 98-05

Julkaisuaika – Date

May 1998

Tekijä(t) – Author(s) Seija Kulmala, Martti Hakanen, University of Helsinki Antero Lindberg, Geological Survey of Finland	Toimeksiantaja(t) – Commissioned by Posiva Oy
Nimeke – Title SORPTION OF IODINE ON ROCKS FROM POSIVA INVESTIGATION SITES	
Tiivistelmä – Abstract <p>Posiva Oy intends to dispose of the spent nuclear fuel produced in Finnish nuclear power plants in a repository deep in the Finnish bedrock. For the purpose, Posiva Oy has selected four areas in Finland for detailed investigation. These areas are Kivetty in Äänekoski, Romuvaara in Kuhmo, Olkiluoto in Eurajoki and Hästholmen in Loviisa.</p> <p>In the Finnish safety analysis (TILA-96), a K_d value of $2 \cdot 10^{-4} \text{ m}^3/\text{kg}$ is used for iodine in far-field conditions. The K_d value is based on experiments performed in Sweden and at the Department of Radiochemistry, University of Helsinki. The purpose of this study was to verify the values in new experiments.</p> <p>The sorption of iodine was studied in batch experiments and retardation in column experiments. The batch experiments with non-saline Allard water gave R_d values of 0.11-1.7 ml/g for Olkiluoto rocks, 0.13-0.55 ml/g for Romuvaara rocks, 0.23-2.3 ml/g for Kivetty rocks and 0.13-0.71 ml/g for Hästholmen rock. With saline OLSO water, the R_d values were ≤ 0.03 ml/g for both Olkiluoto and Hästholmen rock. The R_d values for non-saline water are almost the same as in earlier studies, but the R_d values for saline water are about one decade lower than the values for rapakivi granite and brackish water measured earlier.</p> <p>The column studies on iodine with relatively high flow rates showed no significant difference between the retardation of iodine and the retardation of tritiated water.</p>	
Avainsanat - Keywords iodine, sorption, retardation, batch experiments, retardation	
ISBN ISBN 951-652-043-X	ISSN ISSN 1239-3096
Sivumäärä – Number of pages 43	Kieli – Language English



Tekijä(t) – Author(s) Seija Kulmala, Martti Hakanen, Helsingin yliopisto Antero Lindberg, Geologian tutkimuskeskus	Toimeksiantaja(t) – Commissioned by Posiva Oy
Nimeke – Title JODIN SORPTIO POSIVAN TUTKIMUSALUEIDEN KIVIIN	
Tiivistelmä – Abstract <p>Posiva Oy suunnittelee suomalaisissa ydinvoimaloissa syntyvän käytetyn polttoaineen loppusijoittamista syväälle suomalaiseen kallioperään. Tätä tarkoitusta varten Posiva Oy on valinnut neljä paikkakuntaa eri puolilta Suomea tutkimuksia varten. Nämä paikkakunnat ovat Kivetty Äänekoskella, Romuvaara Kuhmossa, Olkiluoto Eurajoella ja Hästholmen Loviisassa.</p> <p>Suomalaisessa turvallisuusanalyysissä, TILA-96:ssa jodille kaukoalueella käytetään K_d-arvoa $2 \cdot 10^{-4} \text{ m}^3/\text{kg}$. Tämä K_d-arvo perustuu Ruotsissa ja Helsingin yliopiston Radiokemian laitoksella tehtyihin tutkimuksiin. Tämän tutkimuksen tarkoituksena oli varmistaa ko. arvon oikeellisuus uusilla kokeilla.</p> <p>Jodin sorptiota tutkittiin batch-kokeilla ja sen pidättymistä kolonnikokeilla. Jodin R_d-arvoiksi makeasta Allard-vedestä saatiin Olkiluodon kivimurskeille 0.11-1.7 ml/g, Romuvaaran kivimurskeille 0.13-0.55 ml/g, Kivetyyn kivimurskeille 0.23-2.3 ml/g ja Hästholmenin kivimurskeelle 0.13-0.71 ml/g. Suolaisesta OLSO-vedestä jodin R_d-arvoiksi saatiin $\leq 0.03 \text{ ml/g}$ sekä Olkiluodon että Hästholmenin kivimurskeelle. Makealla vedellä saadut tulokset ovat lähes samat kuin aikaisemminkin saadut. Suolaisessa vedessä sitä vastoin R_d-arvo oli kertalukua pienempi kuin mitä aikaisemmin on saatu rapakivelle Hästholmenin murtovedessä.</p> <p>Kolonnikokeissa jodin pidättyminen murskatulla kivellä täytetyssä kolonnissa ei poikennut merkittävästi tritoidun veden pidättymisestä käytetyillä melko suurilla virtausnopeuksilla.</p>	
Avainsanat - Keywords jodi, sorptio, pidättyminen, batch-kokeet, pidättyminen	
ISBN ISBN 951-652-043-X	ISSN ISSN 1239-3096
Sivumäärä – Number of pages 43	Kieli – Language Englanti

PREFACE

This study was commissioned by Posiva Oy and was carried out in the Laboratory of Radiochemistry, Department of Chemistry, University of Helsinki. The contact persons at Posiva Oy were Margit Snellman and Aimo Hautojärvi. The responsible researchers and authors were Seija Kulmala and Martti Hakanen from the Laboratory of Radiochemistry. The rock samples were chosen and identified by Antero Lindberg from the Geological Survey of Finland.

TABLE OF CONTENTS

Abstract

Tiivistelmä

1	INTRODUCTION	8
2	MATERIALS	10
	2.1 Rock samples	10
	2.2 Waters	12
	2.3 Radionuclides	13
	2.4 Radioactivity assay	13
3	METHODS	14
	3.1 Batch experiments	14
	3.2 Column experiments	18
4	RESULTS	19
	4.1 Batch experiments	19
	4.2 Column experiments	25
5	CONCLUSIONS	26
	REFERENCES	28
	APPENDIX A: Mineral compositions of the rock samples	30
	APPENDIX B: Results of the batch and column experiments	31

1 INTRODUCTION

Posiva Oy, the company in charge of managing the spent nuclear fuel from Finnish nuclear power plants, is surveying the possibility to dispose of the used nuclear fuel in the Finnish bedrock at a depth of about 500 m. For this purpose four areas in Finland were chosen for detailed investigation: Romuvaara in Kuhmo, Kivetty in Äänekoski, Olkiluoto in Eurajoki and Hästholmen in Loviisa.

This study is one part of a wider investigation into the migration behaviour of nuclear waste radionuclides under groundwater conditions in Finland.

The main chemical form of dissolved iodine in groundwaters is iodide (I⁻). The presence of iodate, IO₃⁻, due to oxidation by dissolved oxygen (Rancon 1988) or radiolysis (Vovk 1988), has also been suggested. Strong sorption of iodate-form iodine has been measured on minerals common on granitic rock fracture surfaces but no sorption of iodide (Ticknor & Cho 1990). Attachment of iodine to geological media by microbial activity and organic substances has been clearly demonstrated (Behrens 1982, Behrens 1985, Bors et al. 1988, Lieser & Steinkopff 1989). Strong abiotic sorption has been shown on minerals containing metals (Hg, Cu), which form low-solubility iodides (Huie et al. 1988, Rancon 1988). The anion exchange capacities of minerals common on fracture surfaces and of main minerals in bulk crystalline rocks are generally low but sorption of iodide to many minerals should be possible (Allard et al. 1983). Evidently there is competition in the sorption between iodide and other anionic species in groundwaters. Lieser & Steinkopff (1987) have concluded that selectivity of iodide is higher than that of chloride in sorption on (unspecified) mineral fraction of sediment. Sorption of oxyanions on oxide minerals by ligand exchange is clear (Stumm & Morgan 1996). Sorption of iodide on kaolinite and illite (Rancon 1988) and TiO₂ (Hakem et al. 1996) shows the same, though weaker, pH dependency as the sorption of oxyanions.

The relevant data for the sorption of iodine on crystalline rocks and marl have recently been reviewed by Stenhouse (1995).

The K_d values used in Finland for the sorption of iodine on crystalline rocks are based on experiments made in Sweden (Allard et al. 1979 and 1980) and at the Department of Radiochemistry, University of Helsinki (Hietanen et al. 1985, Pinnioja et al. 1986; see also the review by Hakanen & Hölttä, 1992). Koskinen et al. (1985) measured, for iodine, K_d values of 0.5-0.9 ml/g for crushed tonalite ($\phi < 2.0$ mm), 1.4-1.9 ml/g for crushed mica gneiss ($\phi < 2.0$ mm) and 0.28-0.42 ml/g for crushed rapakivi granite ($\phi < 2.0$ mm) in Allard water. Hietanen et al. (1985) measured an R_d value of 0.35 ± 0.07 ml/g for rapakivi granite in brackish Hästholmen groundwater and Allard et al. (1979) a value of 1 ml/g for granite in fresh groundwater.

The purpose of the present study was to verify the sorption values for iodine with new experiments.

2 MATERIALS

2.1 Rock samples

The rock samples were from the four areas under investigation by Posiva as possible locations for the repository of spent nuclear fuel. The rocks were mica gneiss and two types of granodiorite from Olkiluoto, granite, tonalite and mica gneiss from Romuvaara, porphyritic granite from Kivetty and rapakivi granite from Hästholmen.

Rock samples were cut from 42-mm diameter drill cores and crushed and sieved to grain size $\phi < 2.0$ mm. The rocks from Olkiluoto, Romuvaara and Kivetty are the same as those used in the sorption studies of protactinium (Kulmala et al. 1996). Mineral compositions of the rocks are presented in Table 1. The mineral compositions were determined from polished thin sections, one thin section per rock. More detailed information on the rock samples is given in Appendix A.

Table 1. Mineral composition of thin sections (Vol.-%) calculated by point counting method (500 points/sample). Rock names are "field names": the names in parenthesis are the names based on point counting.

Minerals	O11, mica gneiss	O12A, tonalite (granodiorite)	O12B, granite (granodiorite)	Ro1A, tonalite	Ro1B, leukotonalite (granite)	Ro1C, mica gneiss	Ki4A, porph. granite	Ki4B, porph. granodiorite (porph. granite)	HH2, rapakivi granite
K-feldspar	19.2	16.6	15.2	-	30.2	0.2	26.4	36.6	33.0
Plagioclase	14.8	38.6	38.4	25.4	24.8	27.0	23.6	25.4	19.4
Quartz	16.6	26.0	35.8	44.4	36.8	38.8	39.0	20.0	40.6
Biotite	32.4	6.6	0.2	16.4	2.6	31.2	5.4	12.0	3.2
Muscovite	2.4	3.6	9.2	12.2	3.8	0.2	0.8	0.8	2.0
Chlorite	-	6.4	0.2	+	0.6	0.2	-	-	0.4
Hornblende	-	-	-	-	+	-	4.0	4.4	1.4
Apatite	?	1.0	-	0.2	+	0.8	0.2	0.2	+
Epidote	+	0.4	+	1.0	1.2	0.6	-	0.2	+
Calcite	-	-	-	-	+	-	-	-	-
Sphene	-	+	-	+	-	-	-	0.2	-
Zircon	-	-	-	?	?	+	-	-	-
Garnet	-	0.6	+	-	-	-	-	-	-
Sillimanite	3.2	-	1.0	-	-	-	-	-	-
Cordierite	9.8	-	-	-	-	-	-	-	-
Fluorite	-	-	-	-	0.2	-	-	-	+
Opagues	1.6	0.2	-	0.4	+	1.0	0.6	0.2	+

- not observed
+ optically observed
? uncertain observation

2.2 Waters

The waters were synthetic, non-saline Allard water (Allard & Beall 1979) and saline OLSO water (Vuorinen et al. 1997). For sorption studies at different pH, four Allard waters of different pH were prepared. All waters were prepared from ultra pure grade water with use of analytical grade reagents. The chemical compositions of the synthetic, simulated groundwaters are presented in Table 2.

Table 2. Chemical compositions of the synthetic, simulated groundwaters. The four first waters are so-called pH-Allard waters and the waters with pH value given in parenthesis are commonly used Allard water and OLSO water. (Amounts of added chemicals, only pH was measured after preparation)

	pH 5 Allard	pH 6 Allard	pH 7 Allard	pH 10 Allard	Allard (pH 8.2)	OLSO (pH 7.2)
Na (mg/l)	52.4	52.4	52.4	56.0	56.0	4800
K (mg/l)	3.91	3.91	3.91	3.91	3.9	21
Ca (mg/l)	17.9	17.9	17.9	17.9	18	4000
Mg (mg/l)	4.32	4.32	4.32	4.32	4.3	56
SiO ₂ (mg/l)	6.44	6.44	6.44	12	12	2.5
SO ₄ (mg/l)	9.61	9.61	9.61	9.61	9.6	4.2
Cl (mg/l)	115	105	65.6	55	55	14600
HCO ₃ (tot) (mg/l)	123	123	123	123	123	10
Sr (mg/l)						35
Br (mg/l)						105
F (mg/l)						1.2
I (mg/l)						0.9
B (mg/l)						0.9
pH (measured)	4.4	6.0	7.0	10.1	8.2	7.2

2.3 Radionuclides

The radionuclides were I-125 (Amersham International, NaI in dilute NaOH) and H-3 (Amersham International). I-125 was used in batch sorption studies and in column experiments, while H-3 tritiated water was used in column experiments as a flow rate monitor.

2.4 Radioactivity assay

Iodine was measured by γ -counting with Wallac Ultrogamma 1280 (well-type NaI(Tl) detector) and tritium by liquid scintillation counting with LSC Wallac 1217 Rackbeta.

3 METHODS

Sorption of iodine was studied by batch method and retardation in column experiments. Both experiments were made at room temperature and under ambient atmospheric conditions.

3.1 Batch experiments

The batch experiments were made in 50-ml polypropylene centrifuge tubes. The solid/water (m/V) ratio was 1/10. The samples (three or four parallel) consisted of 3.5 g of crushed rock and 35 ml of water. Allard-water was used for all rocks, OLSO only for Olkiluoto and Hästholmen rock samples. The samples were mixed for two weeks (a 15-minute period every hour) to achieve a steady state between the rock and the water. During this time the water was changed four times, which in previous experiments (Huitti et al. 1998) was observed to be sufficient. Spiking was done by adding 20 ± 0.1 μl of I-125 solution to the sample after the final water change (the error in amounts of rock and water by weight is less than 0.1%. The error in volume of the I-125 spiking solution was smaller than 0.5%). Mixing was continued for two weeks. The samples were then centrifuged (4200 G, 30 min) and a 5-ml subsample was taken for radioactivity measurement.

For desorption studies the whole sorption study solution was removed, the rock sample was weighed and 35 ml of fresh Allard or OLSO water was added to the sample. The samples were mixed as before for three weeks, centrifuged, and a 5-ml subsample was taken for measurement. The procedure is schematically presented in Figure 1.

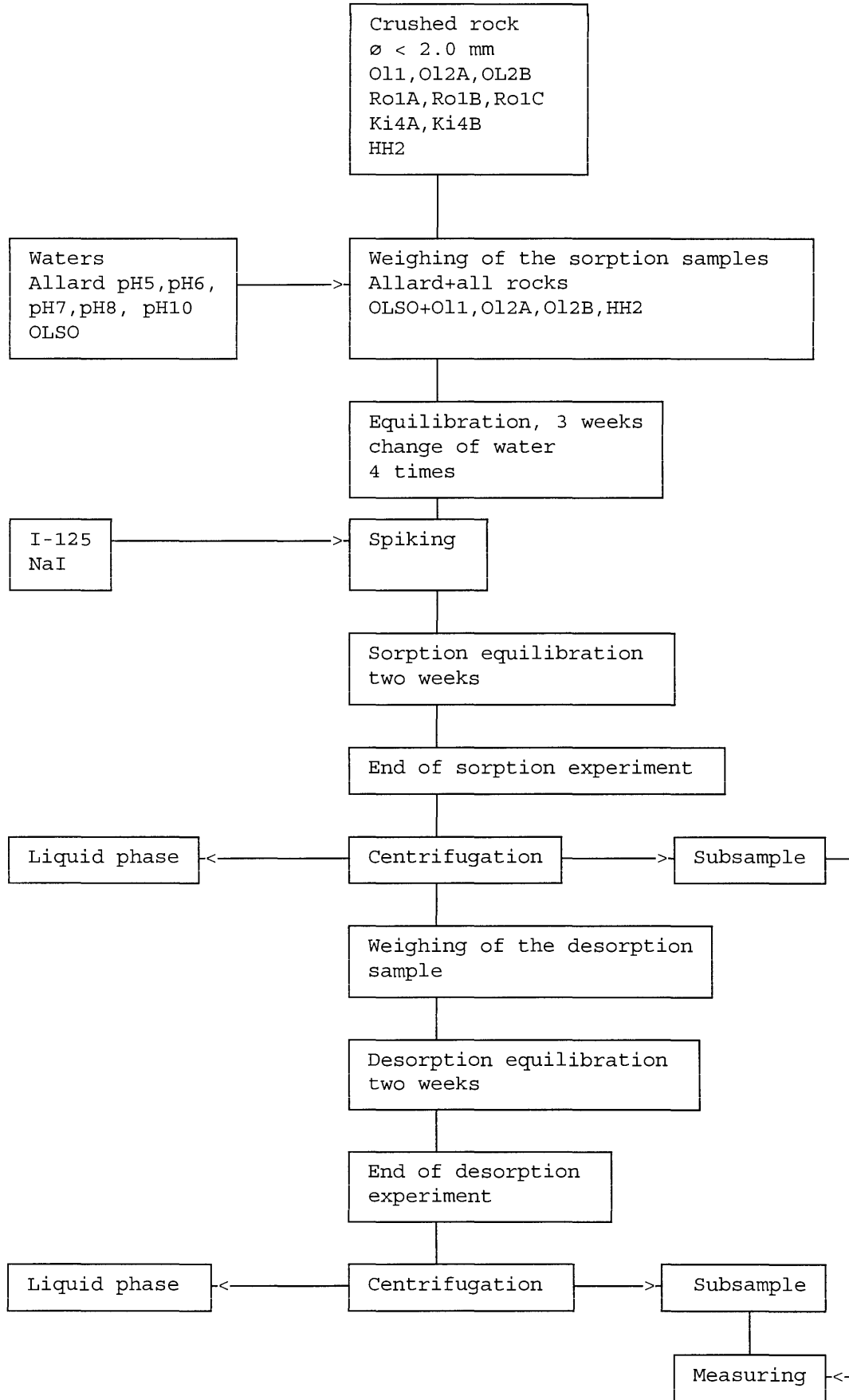


Figure 1. Experimental procedure for sorption batch experiments.

The sorption percentages, $S(\%)$, were calculated from the measurements with Equation 1:

$$S(\%) = \frac{\frac{A_{tracer} - A_{sample}}{V_{total} - V_{sample}}}{\frac{A_{tracer}}{V_{total}}} * 100\% \quad 1$$

where A_{tracer} = activity of the spiking solution
 V_{total} = total volume of the sample
 A_{sample} = activity of the measured subsample
 V_{sample} = volume of the measured subsample

The sorption ratio, R_d , was calculated with Equation 2:

$$R_d = \frac{S(\%)}{100 - S(\%)} * \frac{V}{m} \quad 2$$

where $S(\%)$ = sorption percentage
 V = volume of water in the sample (m^3)
 m = mass of the solid in the sample (kg)

$$R_d(m^3/kg) = \frac{R_d(ml/g)}{10^3} \quad 3$$

When the liquid phase is removed from the sorption sample, some of the spiking solution is unavoidably left in the crushed rock. This is taken into account in calculating the sorption percentages in desorption experiments.

$$S_{de} (\%) = \frac{A_{tracer} * S - \left[\frac{A_{de,sample}}{V_{de,sample}} * V_{de,total} - \frac{V_r * A_{tracer}}{V_{s,total}} * (1 - S) \right]}{A_{tracer} * S + \frac{V_r * A_{tracer}}{V_{s,total}} * (1 - S)} * 100\% \quad 4$$

where A_{tracer} = activity of the spiking solution
 S = sorption percentage/100
 V_r = volume of spiking solution left with the solid phase
 $A_{de,sample}$ = activity of the measured desorption subsample
 $V_{de,sample}$ = volume of the measured desorption subsample
 $V_{de,total}$ = total volume of the desorption sample
 $V_{s,total}$ = total volume of the sorption sample

With some samples a portion of the remaining crushed rock was removed from the experiment at vessel to a measuring tube and analysed for I-125.

To avoid the growth of micro-organisms, which may affect the sorption of iodine, some of the samples were prepared sterile by irradiating (25 kGy) the crushed rock with a Co-60 source. The Allard water used for this experiment was freshly prepared and filtered through 0.22 μ m Millipore filter. The centrifuge tubes, as well as other containers, were rinsed with ethanol before use.

3.2 Column experiments

The retardation of iodine was studied in column experiments. The columns were prepared in polyacrylic tubes (inner $\varnothing = 4$ mm, length 100 mm) by filling them with the crushed rock (porosity about 30%). Allard water was then pumped through the columns until equilibrium between water and rock would be achieved. After about two days pumping, a 1- μ l sample of iodide or tritiated water spike was injected to the column via a Rheodyne injector. The outflowing water was collected as 5-drop (≈ 0.1 g) fractions. Each fraction was weighed, and the time expired between spiking and the collection of the fraction was recorded. The equipment is described in Figure 2.

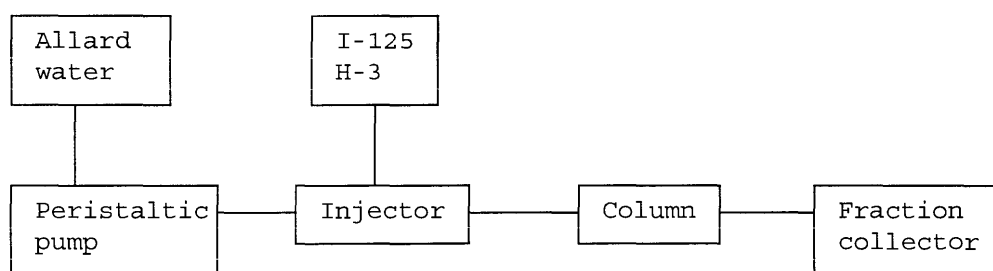


Figure 2. Diagram of the column experiments.

4 RESULTS

4.1 Batch experiments

The sorption percentages and R_d values of iodine from the batch experiments with Allard water (pH 8.2) and OLSO water (pH 7.2) are given in Table 3. The results for desorption studies are presented in Table 4. The sorption percentages and R_d values for pH-Allard waters are presented in Table 5 and the results for desorption studies in Table 6. The results in Tables 3 and 4 are the ranges between minimum and maximum values of four parallel samples and those in Tables 5 and 6 the ranges for three parallel samples. The R_d values are also presented in Figures 3-6. A few rock samples were measured as well. The rock samples were measured after the desorption experiments and only the measured cpm/g values instead of R_d are given in Table 7 and Figure 7. The results for individual samples are presented in Appendix B.

Table 3. Sorption of iodine on rock samples in Allard and OLSO waters.

Rock sample	Allard water		OLSO water	
	R_d (ml/g)	pH (end)	R_d (ml/g)	pH (end)
O11, mica gneiss	0.91 - 1.7	8.45	≤ 0.03	7.69
O12A, tonalite	0.68 - 1.2	8.43	≤ 0.03	7.87
O12B, granite	0.11 - 0.27	8.28	≤ 0.03	7.30
Ro1A, tonalite	0.19 - 0.37	8.46		
Ro1B, granite	0.14 - 0.55	8.47		
Ro1C, mica gneiss	0.13 - 0.20	8.60		
Ki4A, porph. granite	0.23 - 0.98	8.35		
Ki4B, porph. granodiorite	0.59 - 2.3	8.44		
HH2, rapakivi granite	0.13 - 0.71	8.33	≤ 0.03	7.45

Table 4. Desorption of iodine from rock samples in Allard water.

Rock sample		
	R _d (ml/g)	pH (end)
O11, mica gneiss	4.4 - 17	8.36
O12A, tonalite	9.1 - 25	8.35
O12B, granite	1.2 - 6.8	8.14
Ro1A, tonalite	4.6 - 9.8	8.36
Ro1B, granite	5.9 - 6.3	8.33
Ro1C, mica gneiss	0.60 - 1.8	8.48
Ki4A, porph. granite	4.7 - 21	8.18
Ki4B, porph. granodiorite	7.6 - 13	8.30
HH2, rapakivi granite	1.3 - 21	8.30

Table 5. Sorption of iodine on rock samples in pH-Allard waters.

Rock sample	PH 5 Allard (initial pH 4.4)		pH 6 Allard (initial pH 6.0)		pH 7 Allard (initial pH 7.0)		pH 10 Allard (initial pH 10.1)	
	R _d (ml/g)	pH (end)	R _d (ml/g)	pH (end)	R _d (ml/g)	pH (end)	R _d (m ³ /kg)	pH (end)
O11, mica gneiss	2.1-5.5	6.20	0.81-2.9	6.73	0.22-0.43	7.80	0.027-0.17	9.63
O12A, tonalite	2.4-3.4	5.90	1.0-2.6	6.68	0.47-0.92	7.69	≤ 0.03	9.65
O12B, granite	2.4-2.6	5.71	0.20-0.93	6.63	0.27-0.91	7.64	0.0205	9.68
Ro1A, tonalite	1.4-1.8	7.51	0.66-2.8	7.65	0.31-0.84	8.04	0.018-0.24	9.65
Ro1B, granite	2.5-11	6.82	2.5-8.8	7.28	0.57-0.93	8.05	0.059-0.18	9.66
Ro1C, mica gneiss	0.54-3.8	7.65	0.13-0.70	7.86	0.077-0.070	8.13	0.027-0.82	9.65
Ki4A, porph. granite	2.5-8.5	6.34	0.34-5.5	6.97	0.28-0.85	7.93	0.02-0.2	9.64
Ki4B, porph. granodiorite	2.5-11	6.38	0.89-3.0	6.91	1.20	7.96	1.7-69	9.61
HH2, rapakivi granite	73-750	6.30	1.6-3.0	6.90	0.45-1.5	7.91	0.076	9.59

Table 6. Desorption of iodine from rock samples in pH-Allard waters.

Rock sample	pH 5 Allard		pH 6 Allard		pH 7 Allard	
	R _d (ml/g)	pH	R _d (ml/g)	pH	R _d (ml/g)	pH (end)
O11, mica gneiss	15 - 31	6.51	8.1 - 16	6.93	1.9 - 5.4	7.90
O12A, tonalite	21 - 35	6.09	5.7 - 35	6.91	7.7 - 211	7.80
O12B, granite	18 - 26	5.74	1.6 - 14	6.84	1.8 - 12	7.74
Ro1A, tonalite	4.6 - 11	7.74	15 - 54	7.91	3.4 - 34	8.15
Ro1B, granite	12 - 41	6.81	38 - 110	7.39	7.4 - 55	8.20
Ro1C, mica gneiss	7.3 - 29	7.86	16	8.00	-	8.24
Ki4A, porph. granite	46 - 77	6.33	7.0 - 62	7.09	4.6 - 21	8.04
Ki4B, porph. granodiorite	12 - 37	6.38	10 - 285	7.11	30 - 81	8.07
HH2, rapakivi granite	73 - 690	6.36	3.2 - 31	7.05	7.5 - 81	8.10

Table 7. Cpm results for measured crushed rock samples after desorption (cpm/g rock $\pm \sigma_{n-1}$).

Sample	Allard	OLSO	pH 5 Allard	pH 6 Allard	pH 7 Allard	pH 10 Allard
O11, mica gneiss	95 \pm 65	4.3 \pm 1.8	840 \pm 270	520 \pm 220	120 \pm 70	12 \pm 4
Ro1B, granite	52 \pm 34		1270 \pm 370			
Ki4A, porph. granite	72 \pm 28		1500 \pm 500			
HH2, rapakivi granite	110 \pm 20	5.0 \pm 0.7	3400 \pm 300			

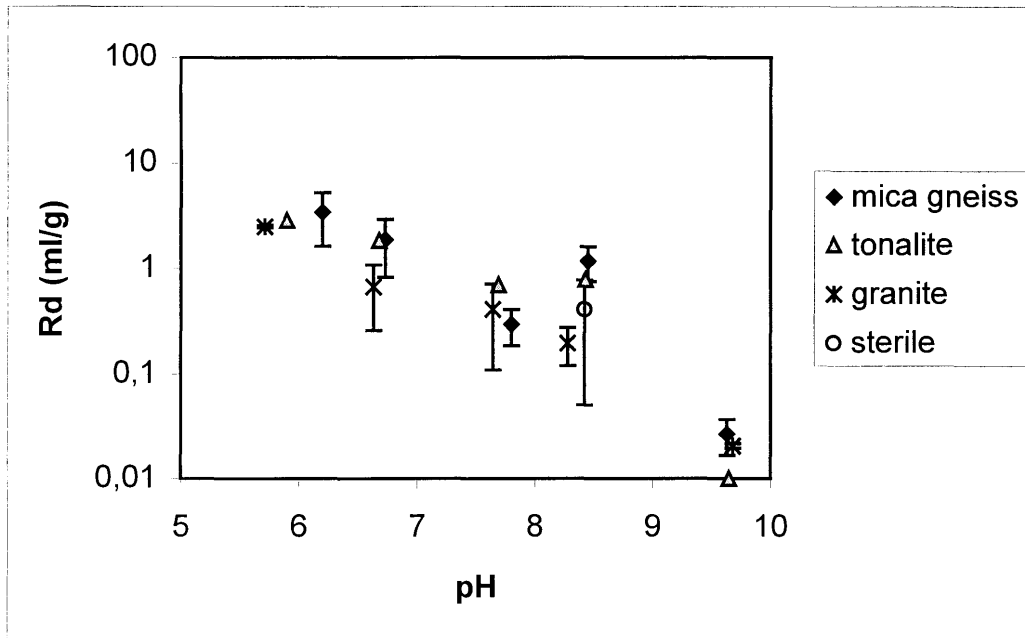


Figure 3. Sorption of iodine on Olkiluoto crushed rock from Allard waters of different pH. The pH value was measured at the end of the sorption experiment. The sterile sample was Ol2A granite.

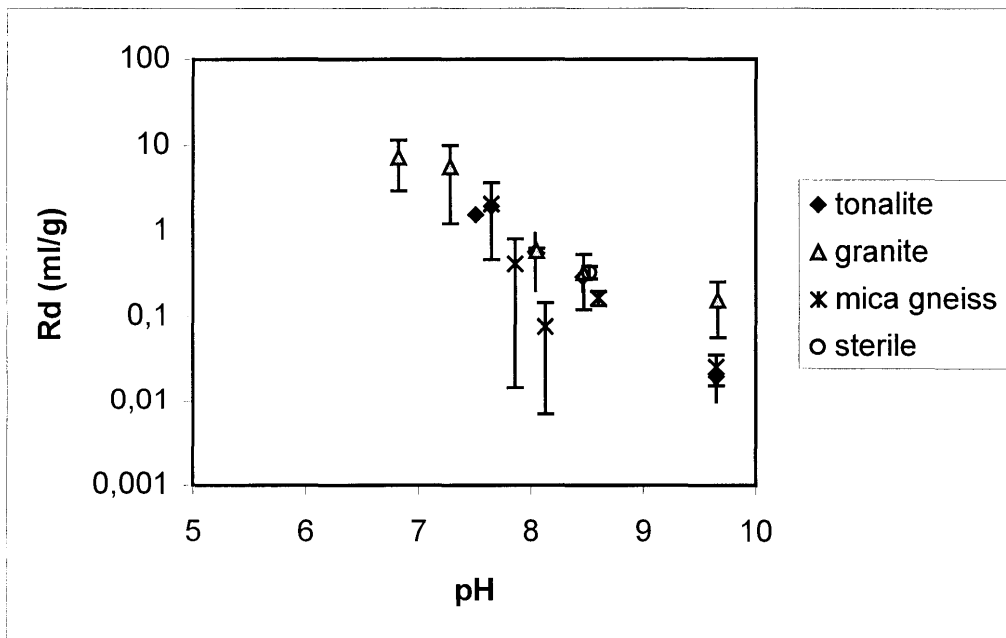


Figure 4. Sorption of iodine on Romuvaara crushed rock from Allard waters of different pH. The pH value was measured at the end of the sorption experiment. The sterile rock sample was Ro1A tonalite.

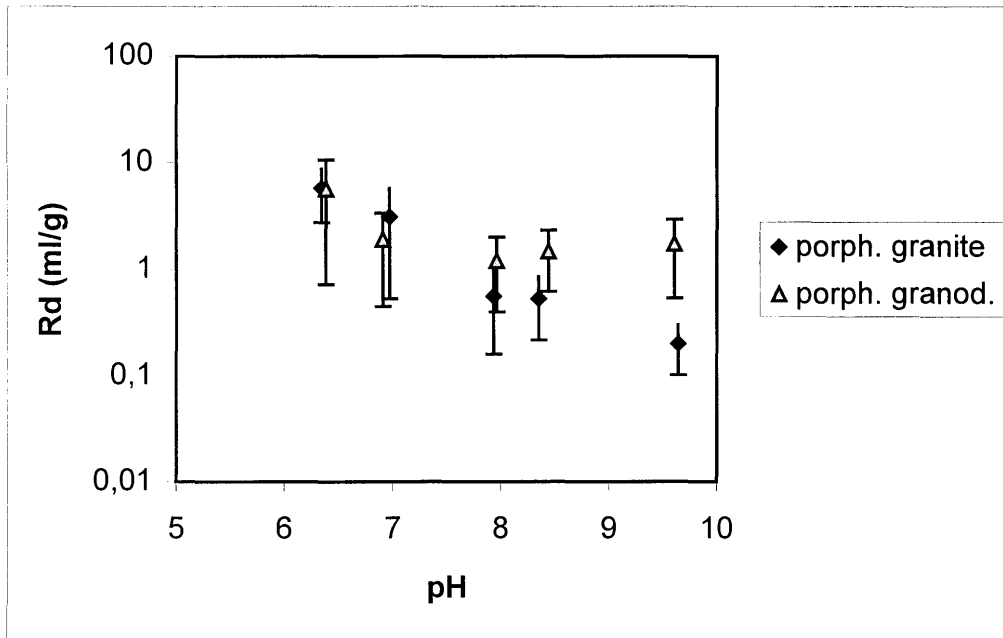


Figure 5. Sorption of iodine on Kivetty crushed rock from Allard waters of different pH. The pH value was measured at the end of the sorption experiment.

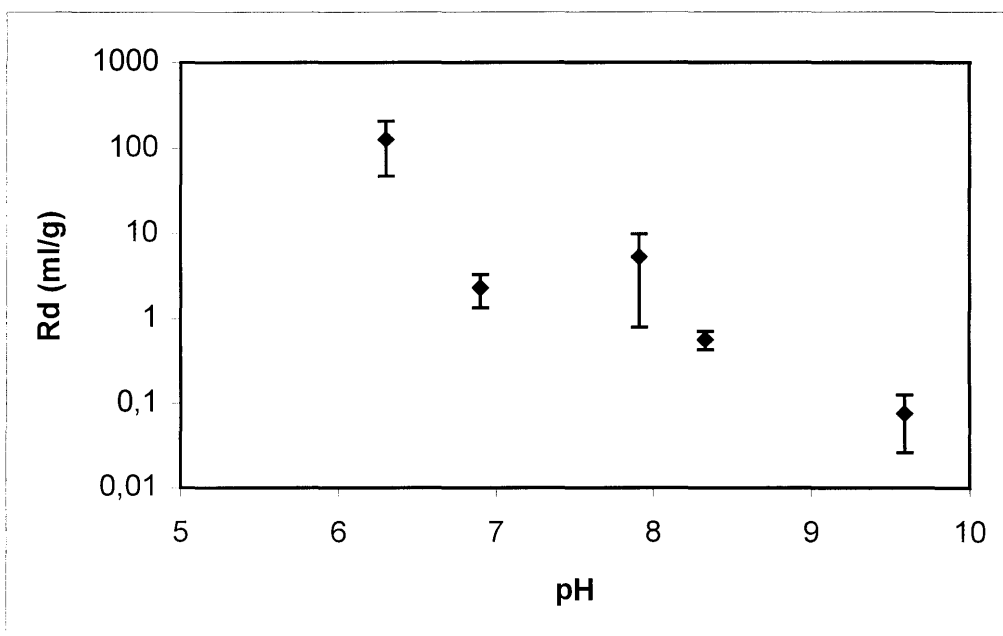


Figure 6. Sorption of iodine on Hästholmen crushed rapakivi granite from Allard waters of different pH. The pH value was measured at the end of the experiment.

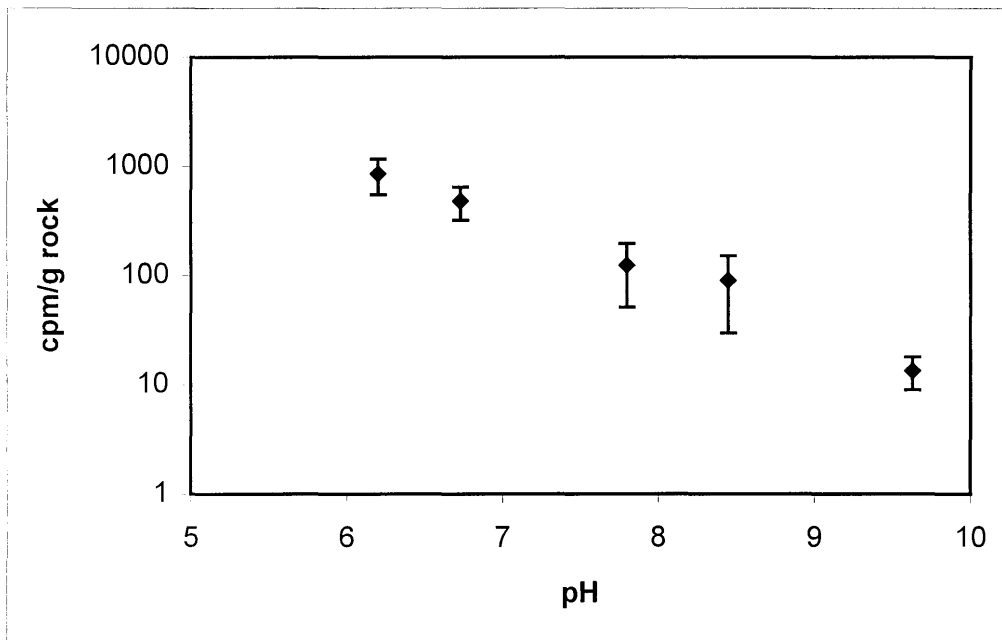


Figure 7. Cpm results of measured Ol1 mica gneiss crushed rock (cpm/g) with the pH of the removed water.

Experiments were also made with sterile samples (see Table 8). The bacteria-free samples were prepared only with Ol2B granite and Ro1A tonalite in Allard water (pH 8.2). The sorption was at the same level as in other experiments (e.g. Figures 3 and 4).

Table 8. Sorption of iodine in sterile rock-water conditions.

Rock	R_d (ml/g)
Ol2B, granite	0.12 - 0.82
Ro1A, tonalite	0.28 - 0.39

The detailed results, i.e. sorption percentages and R_d values of individual samples, are presented in Appendix B.

4.2 Column experiments

The retardation of iodine was studied in column experiments. The porosity of the rock columns was about 30%. The elution curves of iodine and tritiated water for two rocks, O11 mica gneiss and Ro1A tonalite, are presented in Figures 8 and 9. Flow rates in the experiments were high, 18 - 23 mg/min, or about 4.5 mm/min.

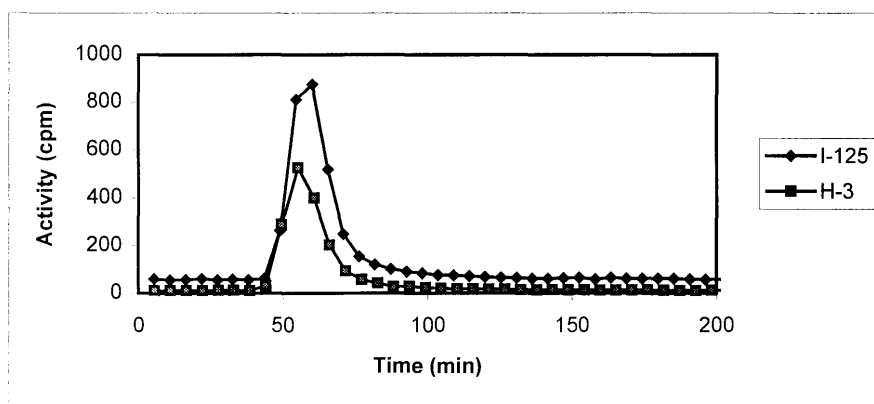


Figure 8. Elution curves of iodine and HTO for O11 mica gneiss crushed rock column. The flow rate of Allard water was 17.7 ml/min.

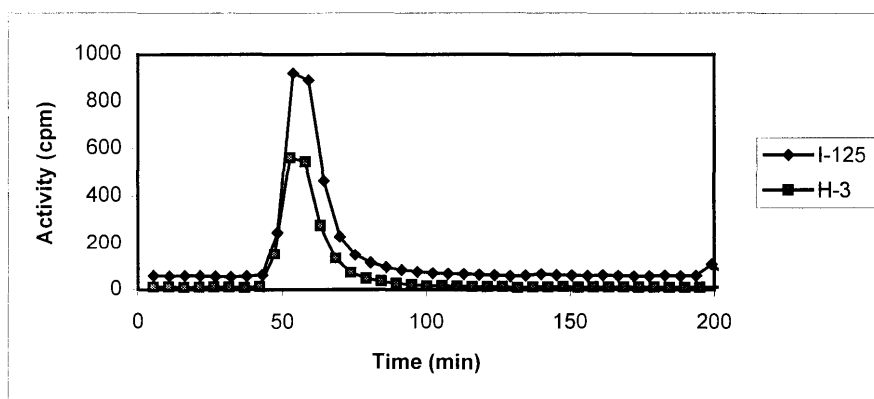


Figure 9. Elution curves of iodine and HTO for Ro1A granite crushed rock column. The flow rate of Allard water was 19.5 mg/min.

As can be seen from the figures, iodine and is not significantly retarded as compared with tritiated water. The results indicate that sorption kinetics of iodine is slow.

5 CONCLUSIONS

Anionic waste nuclides, like iodine in iodide form, are regarded as weakly or non-sorbing in crystalline rock environments. In this study the R_d values measured in batch experiments carried out were 0.11-1.7 ml/g for Olkiluoto crushed rock, 0.13-0.55 ml/g for Romuvaara crushed rock, 0.23-2.3 ml/g for Kivetty crushed rock and 0.13-0.71 ml/g for Hästholmen crushed rock in low-salinity Allard water (initial pH 8.2). In saline OLSO water (initial pH 7.2), the R_d values were ≤ 0.03 ml/g for both Olkiluoto and Hästholmen crushed rock samples.

The surface charge of many natural oxide minerals is positive at slightly acidic to neutral pH. Increased sorption of anions with decreasing pH is expected (Allard et al. 1983). When the pH of the Allard water was made slightly acidic with hydrochloric acid, the R_d values for iodine increased with decreasing pH (Figures 3-6, pp 21-22). The R_d values for Olkiluoto rocks were 2.1-5.5 ml/g at pH 5.7-6.2, 0.20-2.9 ml/g at pH 6.63-6.73, 0.22-0.92 ml/g at pH 7.64-7.80 and 0.02-0.17 ml/g at pH 9.63-9.68. The highest increase in sorption with decreasing pH was measured for Hästholmen crushed rapakivi granite, where the R_d value was 73-750 ml/g at pH 6.30, 1.6-3.0 ml/g at pH 6.90, 0.45-1.5 ml/g at pH 7.91 and 0.076 ml/g at pH 9.59. The results are in agreement with the expected general trend in surface complexation sorption of anions to oxide minerals.

Sorption was lower from saline OLSO water than from Allard water; almost all R_d values were ≤ 0.03 ml/g. There was wide variation in the values of individual samples, however, and the variation was wider than for the non-saline Allard water. The values for the saline water are about one order of magnitude lower than those determined earlier for rapakivi granite in brackish groundwater (Hietanen et al. 1985).

The R_d values measured earlier (Hietanen et al. 1985) for iodine in Hästholmen brackish groundwater (Y1 122-138 m) were 0.35 ± 0.07 ml/g. When analyzed, the initial pH value was 7.0. The pH value at the end of the experiment was not measured. In rapakivi granite/brackish groundwater experiments performed later (Huitti et al. 1996) the initial pH at ambient atmosphere was 7.1 - 7.2. The pH values at the end of the experiments varied from 7.3 to 7.7. The iodine concentration presented in review report of Hästholmen

groundwaters (Snellman & Helenius 1992) was 0.05 mg/l, the concentration of bromine 19 - 28 mg/l and the concentration of chlorine about 5000 mg/l.

The simulated saline groundwater OLSO, contains 0.9 mg/l of iodine, 105 mg/l of bromine and 14 600 mg/l of chlorine. The pH values at the end of the experiments were 7.3 - 7.7. The OLSO water is thus significantly more saline than the Hästholmen brackish water. Also the concentration of iodine in OLSO water ($7 \cdot 10^{-6}$ mol/l) is much higher than the amount of iodine used for spiking the samples ($7.7 \cdot 10^{-9}$ mg/35 ml = $2 \cdot 10^{-12}$ mol/l).

R_d values were higher in the desorption studies than the sorption studies. As indicated by the column experiments the sorption of iodine on the rocks was either partly irreversible or very slow. This was also confirmed by the measurement of selected rock samples after the desorption experiment was completed (Table 7 page 20, Figure 7 page 23). Irreversibility in sorption of iodide has also been reported by Lieser & Steinkopff (1989).

Micro-organisms can cause transformation between inorganic and organic forms of iodine, which may also effect the sorption of iodine (Behrens 1982, Behrens 1985, Bors et al. 1988, Lieser & Steinkopff 1989). The R_d values for the experiments with sterile materials did not differ from the other R_d values (Table 8, p. 23). The observed sorption was thus due to abiotic reactions.

The retardation of iodine was studied column experiments, and the results showed no significant difference between the retardation of iodine and tritiated water. The porosity of the columns filled with the crushed rock was about 30%, which can be assumed sufficient not to have affected the flow rate of HTO. The flow rates in the experiments were quite high (18 - 23 mg/min, about 4.5 mm/min). In comparison with the two R_d values, the results indicate that the sorption kinetics of iodine is slow. It might be useful therefore to study the retardation at much lower flow rates.

REFERENCES

Allard, B., Beall, J., 1979. Sorption of americium on geologic media. *Environm. Sci and Health*. Vol 6, p 507.

Allard, B., Kipatsi, H., Torstenfelt, R., Rydberg, J., 1979. Nuclide transport by ground water in Swedish bedrock. In: McCarthy, G.J. (ed.). *Scientific Basis for Nuclear Waste Management I*. New York, USA: Plenum Press, pp 403-410. ISBN 0-306-40181-9.

Allard, B., Torstenfelt, B., Andersson, K., Rydberg, J., 1980. Possible retention of iodine in the ground. In: Northrup, C.J.M.Jr (ed.). *Scientific Basis for Nuclear Waste Management II*. New York, USA: Plenum Press, pp 673-680. ISBN 0-306-40550-4.

Allard, B., Karlsson, M., Tullborg, E.-L., Larson, S.Å., 1983. Ion exchange capacities and surface areas of some major componenets and common fracture filling materials of igneous rocks. SKBF/KBS Technical Report 83-64. ISSN 0348-7504.

Behrens, H., 1982 New insights into the chemical behaviour of radioiodine in aquatic environments. *Environmental migration of long-lived radionuclides*. IAEA, STI/PUB/597, pp. 27-40.

Behrens, H., 1985. Speciation of radioiodine in aquatic and terrestrial systems under the influence of biochemical processes. In: Bulman, R.A., Cooper, J.R. (ed.). *Speciation of Fission and Activation Products in the Environment*. London: Elsevier Applied Science Publishers, pp 223-230.

Bors, J., Martens, R., Kühn, W., 1988. Studies on the role of natural and anthropogenic organic substances in the mobility of radioiodine in soils. *Radiochimica Acta*, Vol 44/45, pp 201-206, ISSN 0033-8230.

Hakanen, M., Hölttä, P., 1992. Review of sorption and diffusion parameters for TVO-92. Nuclear Waste Commission of Finnish Power Companies, Report YJT-92-14, ISSN 0359-458X.

Hietanen, R., Alaluusua, M., Jaakkola, T., 1985. Sorption of cesium, strontium, iodine, nickel and carbon in mixtures of concrete, crushed rock and bitumen. Nuclear Waste Commission of Finnish Power Companies, Report YJT-85-38. ISSN 0359-548X.

Huie, Z., Jishu, Z., Lanying, Z., 1988. Sorption of radionuclides technetium and iodine on minerals. *Radiochimica Acta*, Vol. 44/45, pp. 143-145, ISSN 0033-8230.

Huitti, T., Hakanen, M., Lindberg, A., 1996. Sorption of cesium, radium, protactinium, uranium, neptunium and plutonium on rapakivi granite. Report POSIVA-96-23, 57 pp, ISBN 951-652-022-7.

- Koskinen, A., Alaluusua, M., Pinnioja, S., Jaakkola, T., Lindberg, A. 1985. Sorption of iodine, neptunium, technetium, thorium, and uranium on rocks and minerals. Nuclear Waste Commission of Finnish Power Companies, Helsinki, Report YJT-85-36. ISSN 0359-548X.
- Kulmala, S., Hakanen, M., Lindberg, A. 1996. Sorption of protactinium on rocks in groundwaters from Posiva investigation sites. Report Posiva-96-18, Appendix A p. 2(3). ISBN 951-652-017-0.
- Lieser, K.H., Steinkopff, Th., 1989. Chemistry of radioactive iodine in the hydrosphere and in the geosphere. *Radiochimica Acta*, Vol. 46, pp 49-55, ISSN 0033-8230.
- Pinnioja, S., Hietanen, R., Alaluusua, M., 1986. Review of the sorption data of the main radionuclides in low and intermediate level nuclear wastes for Finnish bedrock. Nuclear Waste Commission of Finnish Power Companies, Report YJT-86-02, ISSN 0359-548X.
- Rancon, D., 1988. Comparative study of radioactive iodine behaviour in soils under various experimental natural conditions. *Radiochimica Acta*, Vol. 44/45, pp 187-193, ISSN 0033-8230.
- Snellman, M., Helenius, J., 1992. Groundwater chemistry of Hästholmen, Loviisa - review of research in years 1980 - 1992. Nuclear Waste Commission of Finnish Power Companies, Report YJT-92-27, ISSN 0359-548X.
- Stenhouse, M.J., 1995. Sorption databases for crystalline, marl and bentonite for performance assessment. Nagra Technical Report 93-06.
- Stumm, W., Morgan, J.J., 1996. *Aquatic Chemistry, chemical equilibria and rates in natural waters*. 3rd ed. New York, USA: John Wiley & Sons, Inc. 1022 p, ISBN 0-471-51185-4.
- Ticknor, K.V., Cho, Y.-H., 1990. Interaction of iodide and iodate with granitic fracture-filling minerals. *J. Radioanal. Nucl. Chem., Articles*, Vol. 140, No. 1, pp. 75-90, ISSN 0236-5731.
- Vieno, T., Nordman, H., 1996. Interim report on safety assessment of spent fuel disposal TILA 96. Report POSIVA-96-17, ISBN 951-652-016-2, ISSN 1239-3096.
- Vovk, I.F., 1988. Radiolysis as a factor influencing migration behaviour of iodine in the geosphere. *Radiochimica Acta*, Vol. 44/45, pp. 195-199, ISSN 0033-8230.
- Vuorinen, U., Ollila, K., Snellman, M. 1997. Groundwater chemistry of Olkiluoto - saline and brackish water - recipe for saline reference water. Working Report Posiva-97-25.

APPENDIX A: Mineral compositions of the rock samples (Table 1, p. 11)

O11/11 is relatively fresh, even-grained mica gneiss with clear schistosity.

O12A/9 is medium-grained (0.6 - 3 mm) granodiorite which is slightly schistose: plagioclase alteration to sericite has been relatively strong.

OL2B/9 comes from a white granite vein (granodioritic composition on the basis of point counting), which contains muscovite as dark mineral and some sillimanite and garnet marking the metamorphic origin and influence of sedimentary content.

Ro1A/9 is a medium grained (0.5 - 1.0 mm) tonalite, with some indications of deformation and recrystallization.

Ro1B/10 is from a leucocratic tonalite containing abundant potassium feldspar, which puts it in the granite field in the IUGS classification. Relatively coarse muscovite flakes and very fine-grained (cloudy) sericite are abundant.

Ro1C/11 is a fine-grained (0.2 - 0.6 mm) unaltered mica gneiss with clear schistosity.

Ki4A/6 is a porphyritic granite with grain size 1 to 7 mm. The texture shows some deformation but no alteration. Deformation has produced recrystallized, fine-grained groundmass.

Ki4B/4 is very similar to the Ki4A/6 sample, but here large potassium feldspar phenocrysts make the composition more granitic (quartz monzonite). Deformation has been stronger and plagioclase is moderately sericitized.

HH2 the average grain size varies from 1.5 to 3 mm with some coarse (7-12 mm) quartz, K-feldspar and plagioclase grains. Sericite content is result from the slight alteration of plagioclase and K-feldspar. Hornblende has partly altered to biotite and biotite to chlorite.

APPENDIX B: Results of the batch and column experiments.

Table 1. Sorption of iodine from non-saline Allard water.	31
Table 2. Sorption of iodine from saline OLSO water.	32
Table 3. Sorption of iodine from pH 5 Allard water (initial pH 4.4).	33
Table 4. Sorption of iodine from pH 6 Allard water (initial pH 6.0).	34
Table 5. Sorption of iodine from pH 7 Allard water (initial pH 7.0).	35
Table 6. Sorption of iodine from pH 10 Allard water (initial pH 10.1).	36
Table 7. Sorption of iodine in sterile materials.	37
Table 9. Cpm results for measured crushed rock samples after desorption.	37
Figure 1. Elution curve of iodine and HTO for O11 mica gneiss crushed rock column. The flow rate of the Allard water (initial pH 8.2) was 17.7 mg/min	38
Figure 2. Elution curve of iodine and HTO for O12A tonalite crushed rock column. The flow rate of the Allard water was 23.1 mg/min.	38
Figure 3. Elution curve of iodine and HTO for OL2B granite crushed rock column. The flow rate of Allard water was 23.1 mg/min.	39
Figure 4. Elution curve of iodine and HTO for Ro1A tonalite crushed rock column. The flow rate of Allard water was 19.1 mg/min.	39
Figure 5. Elution curve of iodine and HTO for Ro1B granite crushed rock column. The flow rate of Allard water was 19.5 mg/min.	40
Figure 6. Elution curve of iodine and HTO for Ro1C mica gneiss crushed rock column. The flow rate of Allard water was 23.4 mg/min.	40
Figure 7. Elution curve of iodine and HTO for Ki4A porphyritic granite crushed rock column. The flow rate of Allard water was 19.2 mg/min.	41
Figure 8. Elution curve of iodine and HTO for Ki4B porphyritic granodiorite crushed rock column. The flow rate of Allard was 19.3 mg/min.	41
Figure 9. Elution curve of iodine and HTO for HH2 rapakivi granite crushed rock column. The flow rate of Allard water was 19.4 mg/min.	42

Table 1. Sorption of iodine from non-saline, Allard water.

Rock	Sorption(%)	R _d (ml/g)	pH (end)
O11, mica gneiss			
sample 1	8,24 ± 0,58	0,909	8,50
sample 2	14,0 ± 0,6	1,65	8,46
sample 3	8,63 ± 0,58	0,967	8,42
sample 4	1,75 ± 0,62	0,180	8,43
O12A, tonalite			
sample 1	6,31 ± 0,60	0,679	8,42
sample 2	10,5 ± 0,6	1,18	8,37
sample 3	-		8,47
sample 4	4,62 ± 0,61	0,489	8,45
O12B, granite			
sample 1	2,56 ± 0,62	0,265	8,29
sample 2	0,625 ± 0,623	0,0632	8,29
sample 3	1,13 ± 0,62	0,114	8,27
sample 4	2,05 ± 0,62	0,212	8,26
Ro1A, tonalite			
sample 1	2,97 ± 0,61	0,322	8,47
sample 2	3,54 ± 0,62	0,373	8,49
sample 3	1,81 ± 0,63	0,191	8,46
sample 4	0,597 ± 0,597		8,43
Ro1B, granite			
sample 1	1,37 ± 0,63	0,136	8,48
sample 2	0,701 ± 0,624	0,0714	8,44
sample 3	5,27 ± 0,60	0,549	8,44
sample 4	2,97 ± 0,62	0,305	8,50
Ro1C, mica gneiss			
sample 1	1,89 ± 0,62	0,194	8,60
sample 2	0,256 ± 0,256	0,0263	8,62
sample 3	1,69 ± 0,62	0,175	8,60
sample 4	1,31 ± 0,62	0,133	8,57
Ki4A, porph. granite			
sample 1	4,29 ± 0,61	0,475	8,34
sample 2	2,16 ± 0,63	0,229	8,36
sample 3	8,51 ± 0,59	0,974	8,34
sample 4	4,26 ± 0,61	0,451	8,34
Ki4B, porph. granodiorite			
sample 1	5,26 ± 0,60	0,591	8,43
sample 2	13,0 ± 0,6	1,55	8,39
sample 3	17,6 ± 0,6	2,27	8,47
sample 4	-		8,46
HH2, rapakivi granite			
sample 1	6,55 ± 0,55	0,709	8,27
sample 2	4,00 ± 0,56	0,430	8,36
sample 3	1,22 ± 0,57	0,128	8,37
sample 4	5,49 ± 0,56	0,591	8,31

Table 2. Sorption of iodine from saline, OLSO water.

Rock sample	Sorption(%)	R _d (ml/g)	pH (end)
Ol1, mica gneiss			
sample 1	0,261 ± 0,261	0.026	7,76
sample 2	< 0,1	< 0.01	7,57
sample 3	-	-	7,73
sample 4	< 0,1	< 0.01	7,71
Ol2A, tonalite			
sample 1	< 0,1	< 0.01	7,78
sample 2	5,38 ± 0,46	0.57	7,92
sample 3	< 0,1	< 0.01	7,87
sample 4	0,0291 ± 0,0291	< 0.01	7,91
Ol2B, granite			
sample 1	1,72 ± 0,49	0.18	7,31
sample 2	0,243 ± 0,241	0.025	7,29
sample 3	1,11 ± 0,49	0.12	7,28
sample 4	0,0226 ± 0,0226	< 0.01	7,33
HH2, rapakivi granite			
sample 1	< 0,1	< 0.01	
sample 2	0,0863 ± 0,0863	< 0.01	
sample 3	< 0,1	< 0.01	
sample 4	< 0,1	< 0.01	

Table 3. Sorption of iodine from pH 5 Allard water (initial pH 4.4).

Rock sample	Sorption(%)	R _d (ml/g)	pH (end)
Ol1, mica gneiss			
sample 1	20,7 ± 2,0	2,70	6,24
sample 2	34,6 ± 1,7	5,46	6,04
sample 3	17,0 ± 2,1	2,09	6,32
Ol2A, tonalite			
sample 1	19,3 ± 2,1	2,44	5,85
sample 2	20,5 ± 2,1	2,65	5,90
sample 3	24,8 ± 1,9	3,38	5,94
Ol2B, granite			
sample 1	19,4 ± 2,1	2,48	5,67
sample 2	19,4 ± 2,1	2,40	5,78
sample 3	19,8 ± 2,1	2,53	5,67
Ro1A, tonalite			
sample 1	6,66 ± 2,35	1,728	7,50
sample 2	3,80 ± 2,42	1,400	7,48
sample 3	5,69 ± 2,37	1,603	7,56
Ro1B, granite			
sample 1	19,7 ± 2,1	2,51	6,88
sample 2	51,7 ± 1,3	10,9	6,79
sample 3	45,3 ± 1,5	8,42	6,80
Ro1C, mica gneiss			
sample 1	5,04 ± 2,39	0,539	7,70
sample 2	27,1 ± 1,9	3,76	7,65
sample 3	15,8 ± 2,2	1,99	7,61
Ki4A, porph. granite			
sample 1	38,0 ± 1,7	6,30	6,43
sample 2	20,1 ± 2,1	2,53	6,28
sample 3	46,2 ± 1,5	8,47	6,31
Ki4B, porph. granodiorite			
sample 1	23,6 ± 2,0	3,16	6,33
sample 2	19,5 ± 2,1	2,46	6,43
sample 3	52,6 ± 1,3	11,3	6,40
HH2, rapakivi granite			
sample 1	91,8 ± 0,3	115	6,30
sample 2	87,8 ± 0,4	73,1	6,29
sample 3	98,6 ± 0,1	747	6,32

Table 4. Sorption of iodine from pH 6 Allard water (initial pH 6.0).

Rock sample	Sorption(%)	R _d (ml/g)	pH (end)
Ol1, mica gneiss			
sample 1	7,33 ± 2,34	0,814	6,67
sample 2	21,9 ± 2,0	2,89	6,73
sample 3	15,7 ± 2,2	1,88	6,78
Ol2A, tonalite			
sample 1	20,2 ± 2,1	2,56	6,67
sample 2	8,77 ± 2,30	1,01	6,68
sample 3	15,6 ± 2,2	1,90	6,68
Ol2B, granite			
sample 1	7,98 ± 2,32	0,874	6,59
sample 2	8,18 ± 2,31	0,931	6,63
sample 3	1,91 ± 2,46	0,198	6,66
Ro1A, tonalite			
sample 1	21,6 ± 2,0	2,83	7,59
sample 2	6,14 ± 2,36	0,664	7,70
sample 3	20,0 ± 2,2	2,45	7,65
Ro1B, granite			
sample 1	45,9 ± 1,5	8,77	7,35
sample 2	20,0 ± 2,1	2,53	7,25
sample 3	1,64 ± 2,54		7,23
Ro1C, mica gneiss			
sample 1	2,54 ± 2,51		7,82
sample 2	1,26 ± 1,26	0,133	7,91
sample 3	6,52 ± 2,34	0,703	7,85
Ki4A, porph. granite			
sample 1	3,20 ± 2,43	0,340	7,04
sample 2	34,2 ± 1,7	5,46	6,93
sample 3	25,9 ± 1,9	3,56	6,95
Ki4B, porph. granodiorite			
sample 1	22,4 ± 2,0	2,96	6,93
sample 2	96,5 ± 0,3	280	6,90
sample 3	7,99 ± 2,32	0,888	6,89
HH2, rapakivi granite			
sample 1	82,1 ± 0,6	46,8	6,89
sample 2	22,5 ± 2,0	2,99	6,90
sample 3	13,9 ± 2,2	1,63	6,92

Table 5. Sorption of iodine from pH 7 Allard water (initial pH 7.0).

Rock sample	Sorption(%)	R _d (ml/g)	pH (end)
O11, mica gneiss			
sample 1	2,16 ± 2,16	0,225	7,87
sample 2	3,89 ± 2,42	0,423	7,77
sample 3	2,28 ± 2,28	0,241	7,75
O12A, tonalite			
sample 1	8,41 ± 2,31	0,916	7,76
sample 2	87,9 ± 0,5	77,3	7,65
sample 3	4,51 ± 2,39	0,475	7,66
O12B, granite			
sample 1	2,55 ± 2,44	0,268	7,60
sample 2	0,831 ± 0,831	0,0850	7,65
sample 3	8,09 ± 2,31	0,909	7,68
Ro1A, tonalite			
sample 1	7,72 ± 2,32	0,841	8,08
sample 2	3,02 ± 2,43	0,313	8,03
sample 3	75,5 ± 0,7	31,0	8,02
Ro1B, granite			
sample 1	21,7 ± 2,0	2,75	8,10
sample 2	5,85 ± 2,37	0,625	8,03
sample 3	5,29 ± 2,38	0,566	8,02
Ro1C, mica gneiss			
sample 1	0,706 ± 0,706		8,22
sample 2	1,51 ± 1,51		8,07
sample 3	0,0765 ± 0,0765		8,09
Ki4A, porph. granite			
sample 1	7,77 ± 2,32	0,847	7,93
sample 2	2,65 ± 2,44	0,277	7,96
sample 3	0,102 ± 0,102	0,0102	7,91
Ki4B, porph. granodiorite			
sample 1	10,4 ± 2,3	1,20	8,03
sample 2	87,2 ± 0,5	69,2	7,91
sample 3	94,1 ± 0,2	165,7	7,93
HH2, rapakivi granite			
sample 1	4,26 ± 2,41	0,445	7,95
sample 2	12,0 ± 2,3	1,47	7,91
sample 3	87,6 ± 0,5	71,7	7,87

Table 6. Sorption of iodine from pH 10 Allard water (initial pH 10.1).

Rock sample	Sorption(%)	R _d (ml/g)	pH (end)
O11, mica gneiss			
sample 1	< 0,1	< 0.01	9,60
sample 2	1,70 ± 1,70	0.17	9,63
sample 3	0,265 ± 0,265	0.027	9,65
O12A, tonalite			
sample 1	7,21 ± 2,33	0,788	9,64
sample 2	< 0,1	< 0.01	9,65
sample 3	< 0,1	< 0.01	9,66
O12B, granite			
sample 1	5,99 ± 2,36	0,657	9,67
sample 2	0,205 ± 0,205	0.021	9,69
sample 3	< 0,1	< 0.01	9,69
Ro1A, tonalite			
sample 1	2,36 ± 2,36	0.24	9,63
sample 2	0,172 ± 0,172	0,0172	9,66
sample 3	< 0,1	< 0.01	9,67
Ro1B, granite			
sample 1	2,30 ± 2,30	0.24	9,65
sample 2	1,76 ± 1,76	0,178	9,68
sample 3	0,596 ± 0,596	0,0594	9,66
Ro1C, mica gneiss			
sample 1	0,236 ± 0,236	0.024	9,63
sample 2	7,46 ± 2,33	0,823	9,65
sample 3	0,278 ± 0,278	0.028	9,66
Ki4A, porph. granite			
sample 1	0,192 ± 0,192	0,0199	9,63
sample 2	1,98 ± 1,98	0,201	9,66
sample 3	1,28 ± 1,28	0.13	9,64
Ki4B, porph. granodiorite			
sample 1	55,6 ± 1,2	12,9	9,61
sample 2	14,6 ± 2,2	1,74	9,61
sample 3	87,5 ± 0,5	69,0	9,62
HH2, rapakivi granite			
Näyte 1	67,8 ± 0,9	21,9	9,58
Näyte 2	0,753 ± 0,753	0,0761	9,59
Näyte 3	< 0,1	< 0.01	9,59

Table 7. Sorption of iodine in sterile materials.

Rock sample	Sorption %	R _d (ml/g)
O12B, granite		
Sample 1	7.63 ± 0.39	0.822
Sample 2	1.07 ± 0.06	0.110
Sample 3	2.85 ± 0.15	0.297
Ro1A, tonalite		
Sample 1	2.71 ± 0.14	0.282
Sample 2	3.20 ± 0.16	0.332
Sample 3	3.78 ± 0.19	0.392

Table 8. Cpm results of measured crushed rock samples after desorption.

Sample	Allard	OLSO	pH 5	pH 6	pH 7	pH 10
O11, mica gneiss						
Sample 1	150	5.65	749	345	204	8.54
Sample 2	58.9	4.34	1150	769	89.2	11.1
Sample 3	150	1.84	633	437	67.6	16.5
Sample 4	21.9	5.47				
Ro1B, granite						
Sample 1	17.7		856			
Sample 2	28.1		1360			
Sample 3	85.4		1590			
Sample 4	75.2					
Ki4A, porph. granite						
Sample 1	77.7		1650			
Sample 2	44.7		972			
Sample 3	109		1880			
Sample 4	57.4					
HH2, rapakivi granite						
Sample 1	124	5.05	3730			
Sample 2	128	4.39	3240			
Sample 3	90.5	5.96	3230			
Sample 4	84.1	4.53				

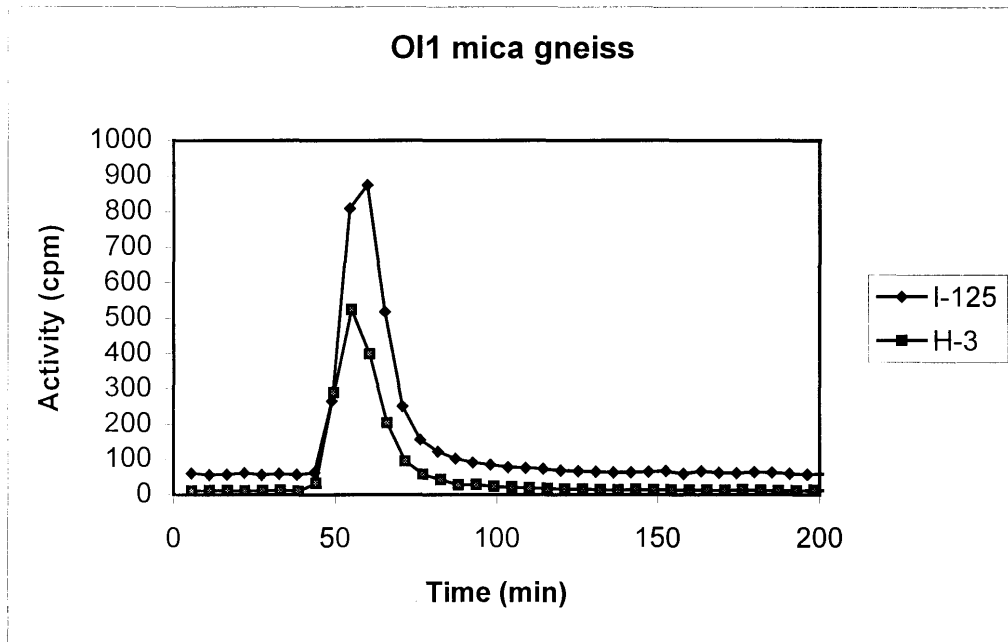


Figure 1. Elution curve of iodine and HTO for OI1 mica gneiss crushed rock column. The flow rate of the Allard water (initial pH 8.2) was 17.7 mg/min.

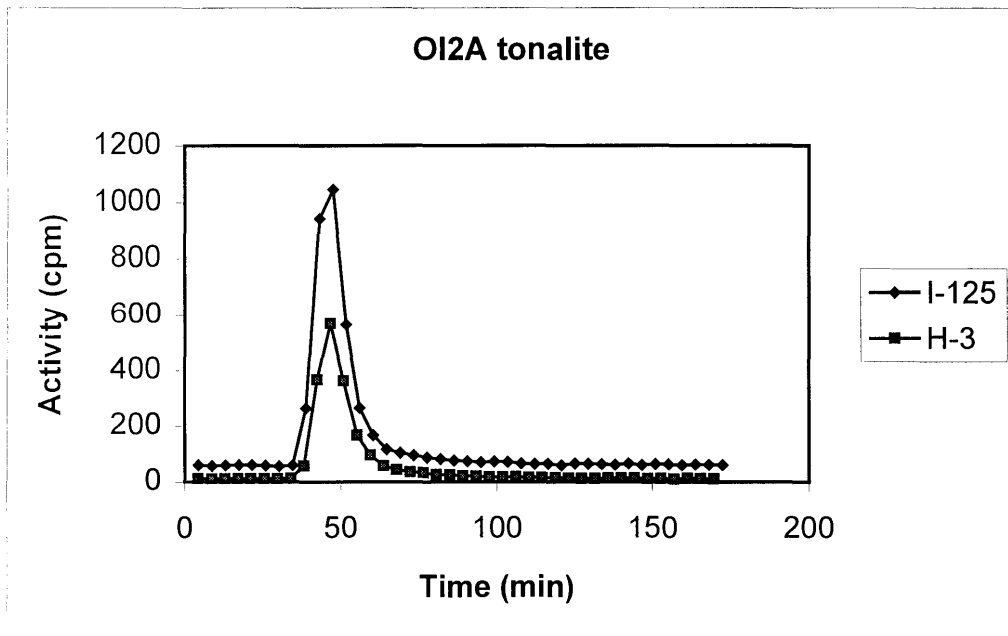


Figure 2. Elution curve of iodine and HTO for OI2A tonalite crushed rock column. The flow rate of Allard water was 23.1 mg/min.

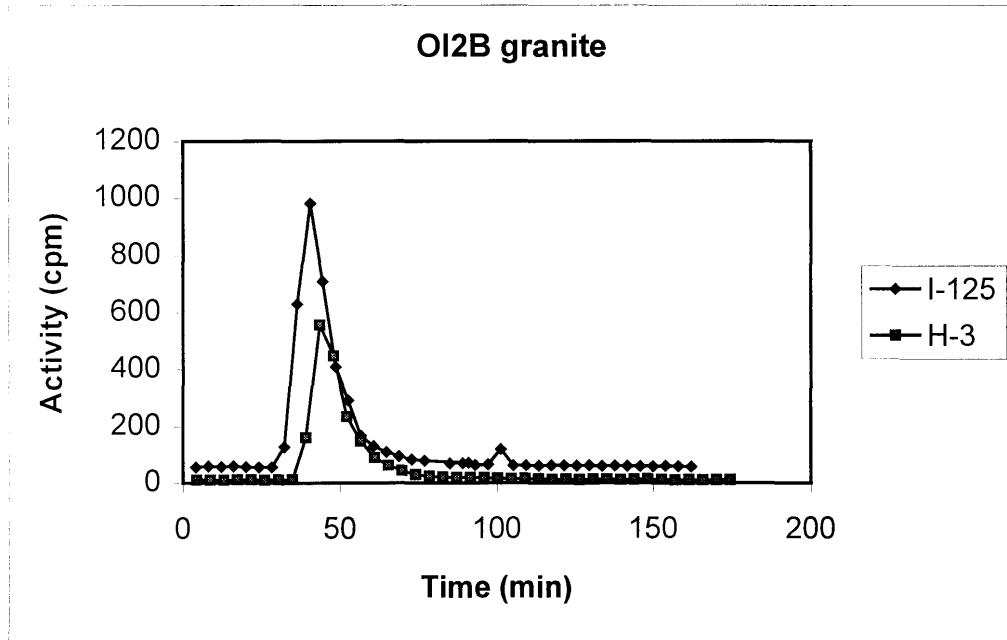


Figure 3. Elution curve of iodine and HTO for Ol2B granite crushed rock column. The flow rate of Allard water was 23.1 mg/min.

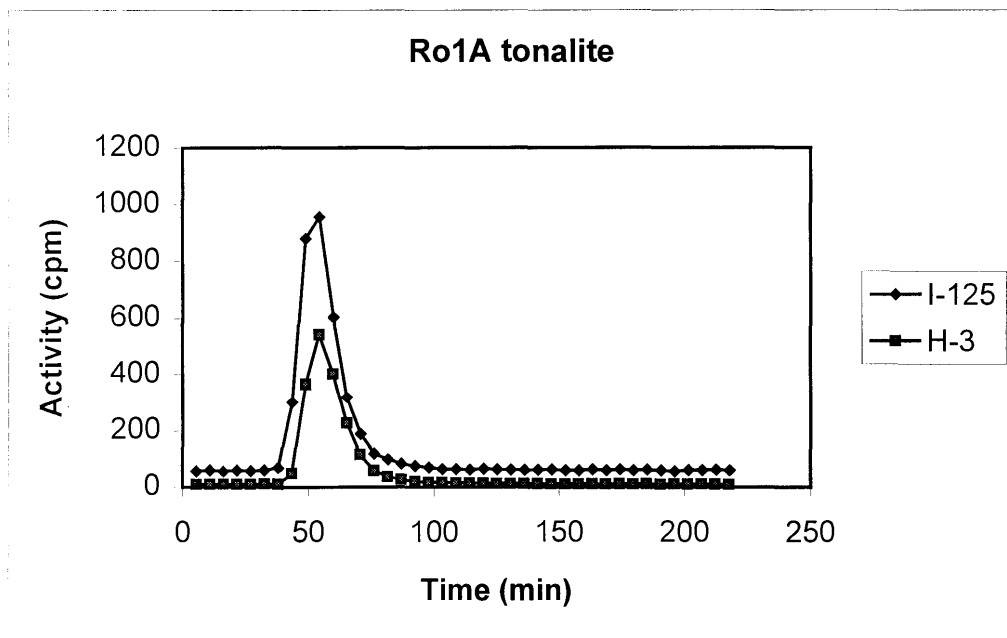


Figure 4. Elution curve of iodine and HTO for Ro1A tonalite crushed rock column. The flow rate of Allard water was 19.1 mg/min.

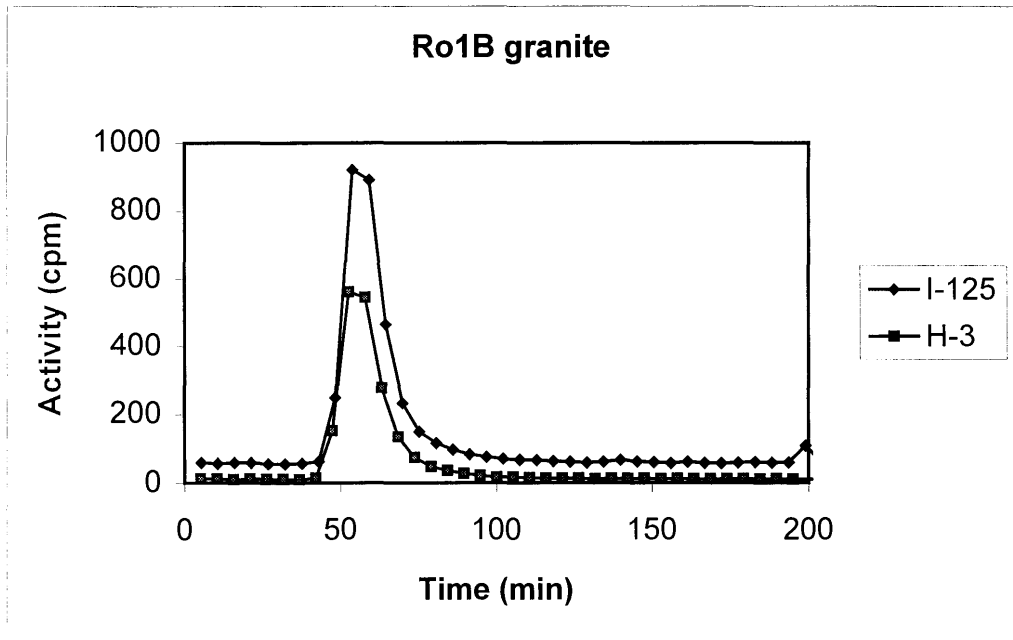


Figure 5. Elution curve of iodine and HTO for Ro1B granite crushed rock column. The flow rate of Allard water was 19.5 mg/min.

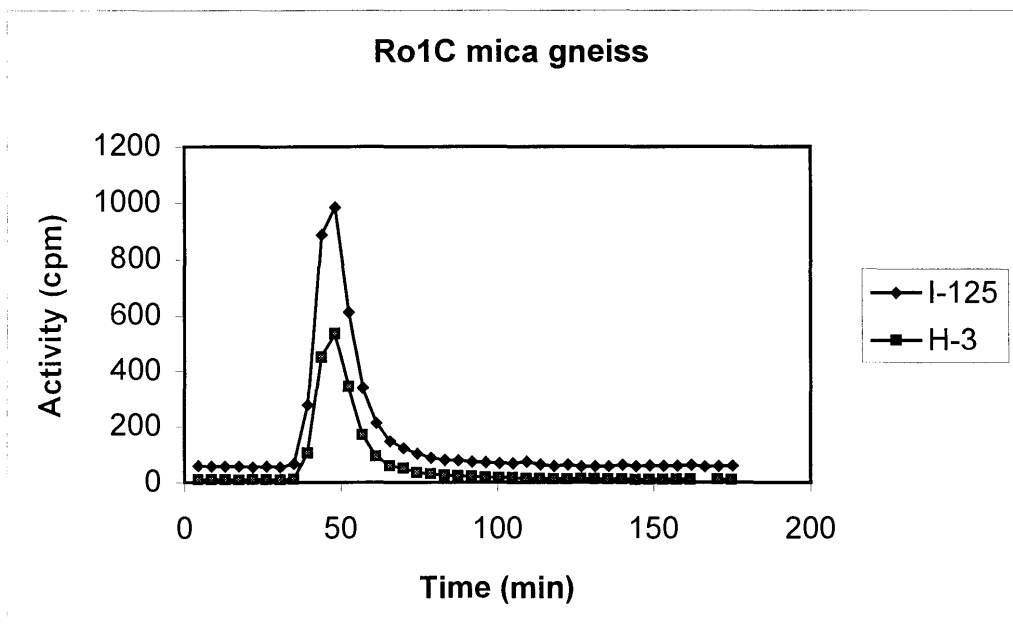


Figure 6. Elution curve of iodine and HTO for Ro1C mica gneiss crushed rock column. The flow rate of Allard water was 23.4 mg/min.

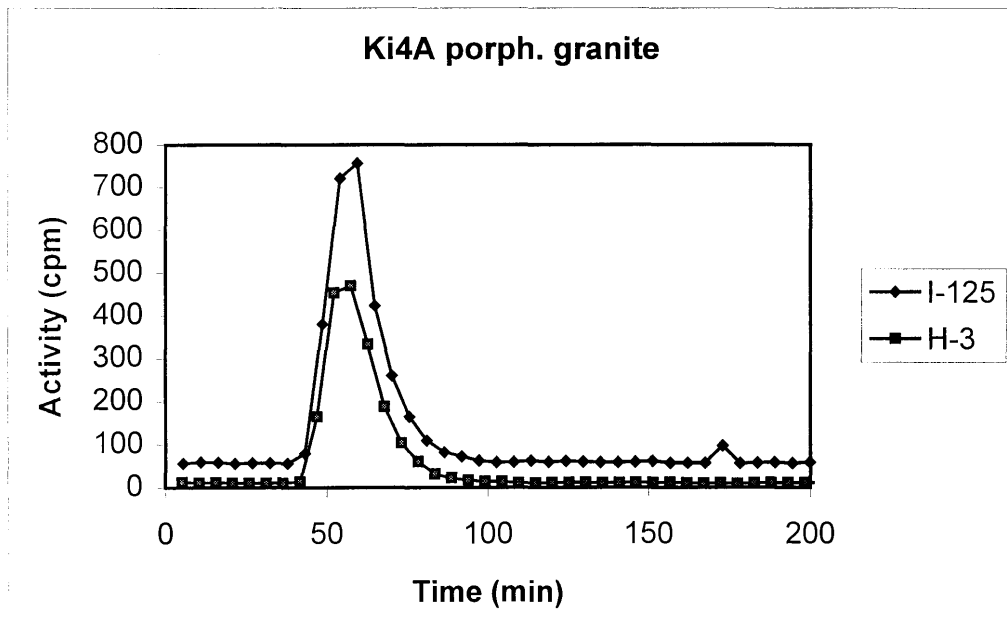


Figure 7. Elution curve of iodine and HTO for Ki4A porphyritic granite crushed rock column. The flow rate of Allard water was 19.2 mg/min.

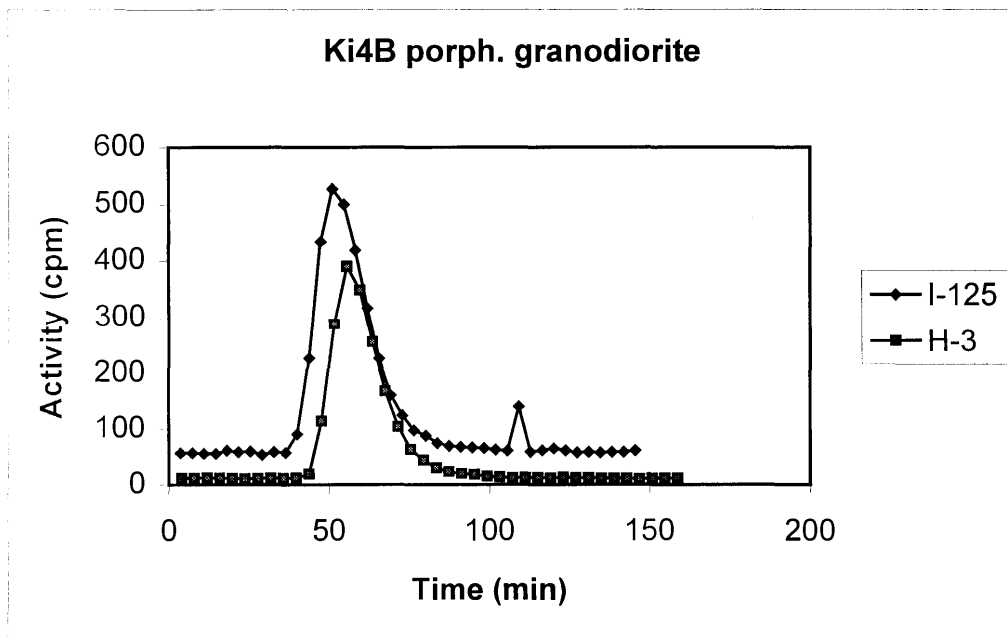


Figure 8. Elution curve of iodine and HTO for Ki4B porphyritic granodiorite crushed rock column. The flow rate of Allard water was 19.3 mg/min.

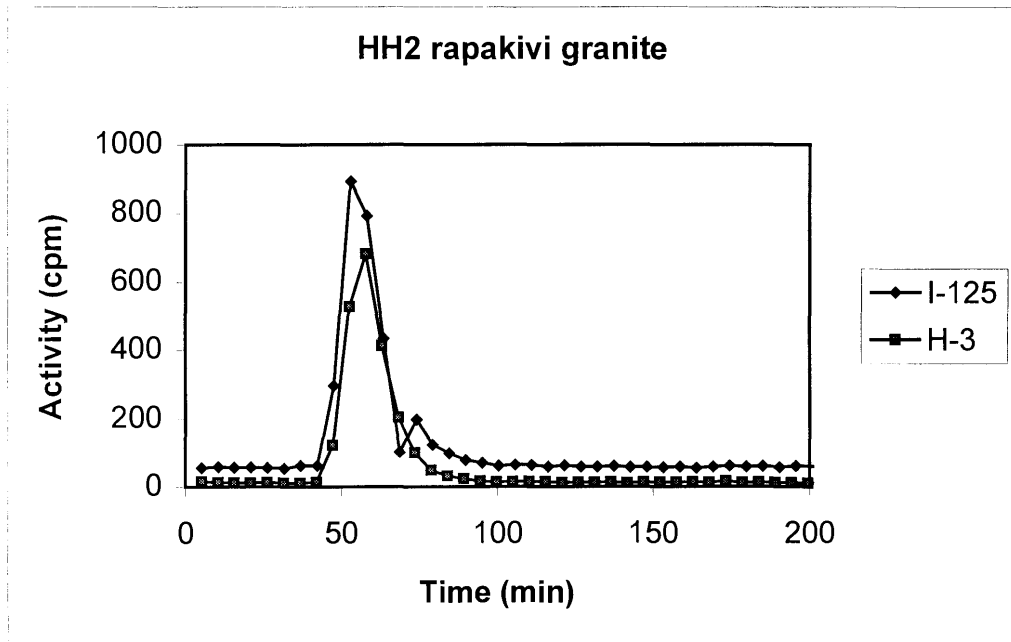


Figure 9. Elution curve of iodine and HTO for HH2 rapakivi granite crushed rock column. The flow rate of Allard water was 19.4 mg/min.

POSIVA REPORTS 1998, situation 5/98

- POSIVA 98-01 Bentonite swelling pressure in strong NaCl solutions
- Correlation of model calculations to experimentally determined data
Ola Karnland
Clay Technology, Lund, Sweden
January 1998
ISBN 951-652-039-1
- POSIVA 98-02 A working groups conclusions on site specific flow and
transport modelling
Johan Andersson
Golder Associates AB, Sweden
Henry Ahokas
Fintact Oy
Lasse Koskinen, Antti Poteri
VTT Energy
Auli Niemi
Royal Institute of Technology, Hydraulic Engineering, Sweden
(permanent affiliation: VTT Communities and Infrastructure, Finland)
Aimo Hautajärvi
Posiva Oy
March 1998 (to be published)
ISBN 951-652-040-5
- POSIVA 98-03 EB-welding of the copper canister for the nuclear waste disposal –
Final report of the development programme 1994-1997
Harri Aalto
Outokumpu Poricopper Oy
April 1998 (to be published)
ISBN 951-652-041-3
- POSIVA 98-04 An isotopic and fluid inclusion study of fracture calcite from borehole
OL-KR1 at the Olkiluoto site, Finland
Alexander Blyth, Shaun Frape
University of Waterloo, Waterloo, Ontario, Canada
Runar Blomqvist, Pasi Nissinen
Geological Survey of Finland
Robert McNutt
McMaster University, Hamilton, Ontario, Canada
April 1998
ISBN 951-652-042-1
- POSIVA 98-05 Sorption of iodine on rocks from Posiva investigation sites
Seija Kulmala, Martti Hakanen
Laboratory of Radiochemistry
Department of Chemistry
University of Helsinki
Antero Lindberg
Geological Survey of Finland
May 1998
ISBN 951-652-043-X