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Sorption of protactinium on rocks in groundwaters from Posiva investigation sites

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Tiivistelmä – Abstract <p>Posiva Oy intends to dispose of the spent fuel produced in Finnish nuclear power plants in a repository deep in the Finnish bedrock. For the purpose Posiva Oy selected three areas for detailed investigations in 1992. These areas are Kivetty in Äänekoski, Romuvaara in Kuhmo and Olkiluoto in Eurajoki. This study examines the sorption behaviour of protactinium on crushed rock and thin section samples of rocks from these areas. In January 1997 detailed investigations will also be initiated at Hästholmen in Loviisa.</p> <p>The sorption of protactinium was high on all rocks under both oxic and anoxic conditions. The main difference between the oxic and anoxic conditions was the solubility of the spiking solution. The spiking solution was dried into a teflon platelet and the teflon platelet put into the sample. Under anoxic conditions the undissolved fraction was 4.70-6.7% and under oxic conditions it was < 1.0%.</p> <p>The sorption of protactinium was lowest on Romuvaara rock samples from RO-KR5 groundwater under anoxic conditions. The R_d values were 0.07-2.3 m³/kg. Under oxic conditions the R_d values were 0.20-1.8 m³/kg. From KI-KR4 and OL-KR5 groundwaters under anoxic conditions the R_d values for Romuvaara rock samples were 0.70-8.6 m³/kg and 0.35-1.7 m³/kg, respectively.</p> <p>The sorption of protactinium on Kivetty and Olkiluoto rock samples was almost the same. The R_d values for Kivetty rock samples from KI-KR4 groundwater were 0.51-1.7 m³/kg and 1.7-12 m³/kg under oxic and anoxic conditions, respectively. The corresponding R_d values for Olkiluoto rock samples were 0.71-3.1 m³/kg and 0.31-2.4 m³/kg.</p>	
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Tiivistelmä – Abstract <p>Posiva Oy suunnittelee suomalaisissa ydinvoimaloissa syntyvän käytetyn ydinpolttoaineen loppusijoittamista syvälle suomalaiseen kallioperään. Yksityiskohtaisia tutkimuksia varten Posiva Oy on valinnut vuonna 1992 kolme aluetta eri paikkakunnilta: Kivetyn Äänekoskelta, Romuvaaran Kuhmosta ja Olkiluodon Eurajoelta. Tässä tutkimuksessa on käsitelty protaktiniumin sorptiota näiltä paikkakunnilta valittuihin kivimurskeisiin ja -hieisiin. Tammikuussa 1997 Loviisan Hästholmen liitetään mukaan käytetyn polttoaineen sijoituspaikkatutkimuksiin.</p> <p>Protaktiniumin sorptio oli korkea kaikilla kivillä sekä hapellisissa että hapettomissa olosuhteissa. Suurin ero hapellisten ja hapettomien olosuhteiden välillä oli leimaliuoksen liukoisuudessa. Leimaliuos kuivattiin teflonpalalle, joka laitettiin näytteeseen. Hapettomissa olosuhteissa teflonpalalle jäi 4,70-16,7 %. Hapellisissa olosuhteissa liukenemattoman leiman osuus oli < 1,0 %.</p> <p>Protaktiniumin sorptio oli heikointa Romuvaaran kiviin RO-KR5-pohjavedestä hapettomissa olosuhteissa. Massajakaantumiskertoimet, R_d-arvot, olivat 0,07-2,3 m³/kg. Hapellisissa olosuhteissa R_d-arvot olivat 0,20-1,8 m³/kg. KI-KR4- ja OL-KR5-pohjavesistä hapettomissa olosuhteissa R_d-arvot Romuvaaran kiviäytteille olivat 0,70-8,6 m³/kg KI-KR4-pohjavedellä ja 0,35-1,7 m³/kg OL-KR5-pohjavedellä.</p> <p>Protaktiniumin sorptio Kivetyn ja Olkiluodon kiviäytteisiin vastaavista pohjavesistä oli miltei samaa. R_d-arvot Kivetyn kiviäytteille KI-KR4-pohjavedestä olivat 0,51-1,7 m³/kg hapellisissa olosuhteissa ja 1,7-12 m³/kg hapettomissa olosuhteissa. Vastaavat R_d-arvot Olkiluodon kiviäytteille OL-KR5-pohjavedestä olivat 0,71-3,1 m³/kg ja 0,31-2,4 m³/kg.</p>	
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PREFACE

This study was commissioned by Teollisuuden Voima Oy (after 1.1.1996 Posiva Oy) and was carried out in the Laboratory of Radiochemistry, Department of Chemistry, University of Helsinki. The contact persons from Posiva were Lauri Pöllänen and Margit Snellman. 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.

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APPENDIX B

1. INTRODUCTION

Since 1983 Teollisuuden Voima Oy (TVO) has been surveying Finnish bedrock for the final disposal of the spent nuclear fuel. Since January 1996 the work has been continued by Posiva Oy. Five sites were studied during the preliminary site investigation phase in 1987–1992. Detailed site investigations were initiated in 1993 in three of these sites: Romuvaara in Kuhmo, Kivetty in Äänekoski and Olkiluoto in Eurajoki.

This study is one part of a broad investigation into the migration behaviour of nuclear waste radionuclides under groundwater conditions in Finland. Here the sorption of protactinium was studied, under both oxic and anoxic conditions. Most samples were crushed rock, the others polished rock thin sections.

2. MATERIALS

2.1 Rock samples

The rock samples were from the three areas under investigation by Posiva as possible locations for the repository of spent nuclear fuel. The samples were mica gneiss and granodiorite from Olkiluoto, granite, tonalite and mica gneiss from Romuvaara and porphyritic granite from Kivetty. All samples were cut from 42-mm diameter drill core samples.

Parts of the samples were crushed and sieved to grain size $\phi < 2.0$ mm. In addition, some polished thin sections were prepared for sorption experiments. Detailed information on the rock samples is given in Table 1, where also the specific areas of the crushed rock samples are presented. The specific areas were measured at the Helsinki University of Technology by N_2 /BET method. One thin section per rock sample was investigated by point counting method to gain an idea of the composition and texture of these heterogeneous rock samples. Mineral compositions of the thin sections are shown in Table 2.

For comparison, experiments were made with two rocks from the Palmottu natural analogue study site: R373/139.70 (drill hole/depth) and R373/146.70. Both these rocks were mica gneiss with considerable pyrite content. The specific areas of these rocks were not measured, neither were the cation exchange capacities of these samples.

Table 1. Rock samples used in the study. Crushed rock samples and polished thin sections were prepared at the Geological Survey of Finland and specific areas were measured at the Helsinki University of Technology, Laboratory of Mechanical Processing and Recycling. Ol=Olkiluoto study site, Ro=Romuvaara study site and Ki=Kivetty study site.

Rock	Depth of the rock thin section (m)	Depth of the crushed rock sample (m)	Specific area of the crushed rock (m ² /g)
Ol1, mica gneiss	561	520–586	0.95
Ol2A, granodiorite (tonalite)	355	346–390	0.22
Ol2B, granodiorite (granite)	417	417–455	0.36
Ro1A, tonalite	337	335–412	0.28
Ro1B, granite (leukotonal.)	559	547–654	0.23
Ro1C, mica gneiss	753	724–793	0.18
Ki4A, porph. granite	301	255–328	0.21
Ki4B, porph. granite (granod.)	385, 439	371–442	0.22

rock names based on point counting; names in parentheses are "field names"

Table 2. Mineral compositions of thin sections (Vol.-%) calculated by point counting method (500 points/sample).

Minerals	Ol1/11	Ol2A/9	Ol2B/9	Ro1A/9	Ro1B/10	Ro1C/11	Ki4A/6	Ki4B/4
K-feldspar	19.2	16.6	15.2	-	30.2	0.2	26.4	36.6
Plagioclase	14.8	38.6	38.4	25.4	24.8	27.0	23.6	25.4
Quartz	16.6	26.0	35.8	44.4	36.4	38.8	39.0	20.0
Biotite	32.4	6.6	0.2	16.4	2.6	31.2	5.4	12.0
Muscovite	2.4	3.6	9.2	12.2	3.8	0.2	0.8	0.8
Chlorite	-	6.4	0.2	+	0.6	0.2	-	-
Hornblende	-	-	-	-	+?	-	4.0	4.4
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	-	-	-
Opakes	1.6	0.2	-	0.4	+	1.0	0.6	0.2

Some detailed information of the thin sections is presented in Appendix A, Table 1.

2.2 Groundwaters

The waters were natural groundwaters collected from the same areas and depths as the rock samples. The concentrations of selected solutes in the groundwaters are presented in Table 3 and the detailed chemical compositions of the groundwaters in Appendix A.

Table 3. Chemical composition of the natural groundwaters used in this study (Imatran Voima Oy, Chem. Lab.

	Ro-KR5/T1 13.11.1995	Ki-KR4/T1 20.11.1995	OI-KR5/T1 30.10.1995
pH	9.3	8.2	8.2
Eh(Pt) (mV) *	300	-75	-270
Conductivity 25°C (mS/m)	18.0	17.0	2330
Alkalinity (meq/l)	0.28	0.24	0.10
KMnO ₄ (mg/l)	2.8	2.9	80.0
SiO ₂ (mg/l)	13.0	13.0	6.3
CO _{2,tot} (mg/l)	53.8	58.2	4.20
Br (mg/l)	0.10	0.03	58.0
Cl (mg/l)	4.3	3.2	9200
F (mg/l)	-	3.0	1.30
PO ₄ (mg/l)	0.44	0.02	<0.01
SO ₄ (mg/l)	3.9	2.00	3.30
Al (mg/l)	0.071	0.030	0.032
Ca (total, mg/l)	2.1	13.0	2100
Fe (total, mg/l)	0.059	0.520	0.230
K (mg/l)	0.69	0.87	15.0
Mg (mg/l)	0.08	3.0	67.0
Mn (mg/l)	0.001	0.210	0.360
Na (mg/l)	39.0	18.0	3330
Cs (mg/l)	<0.005	<0.005	0.008
H-3 (TU)	0.9	1.9	2.0
U-238 (H ₂ O, µg/l)	0.439	3.190	0.026
ionic strength	1.5·10 ⁻³	1.9·10 ⁻³	3.1·10 ⁻¹

* Eh(Pt) (mV) field measurement

Other values are measured at the laboratory

2.3 Chemicals and solutions for measurement of cation exchange capacity

A silver-thiourea (AgTU) solution was prepared to determine the cation exchange capacity (CEC) of the crushed rock samples [1]. The solution contained

7.79 g thiourea (Pro analysi (p.a.), Merck)/240 ml ultra pure grade water
and 7.82 g NH₄Ac (0.1 M, p.a., Merck)
and 500 ml 0.02 M AgNO₃ (1.71 g AgNO₃ (p.a., Merck) and 500 ml ultra pure grade water)
diluted to one litre

Another experiment to determine the CEC value was made with the same AgTU solution but with the pH adjusted to pH 1.0 with 1.0 M HNO₃ solution.

The caesium sorption isotherm was also measured. Four different Cs solutions were prepared in ultra pure grade water. The Cs concentration was obtained with nonradioactive CsCl (p.a. Merck), and 100 µl of Cs-134 solution/sample was added as tracer. The concentrations of caesium in the solutions were:

Cs1 $1.11 \cdot 10^{-2}$ mol/l, Cs2 $1.16 \cdot 10^{-4}$ mol/l

Cs3 $1.22 \cdot 10^{-6}$ mol/l, Cs4 $1.16 \cdot 10^{-7}$ mol/l

2.4 Radionuclides and carrier solutions

The radionuclides were Pa-231 ($T_{1/2} = 32\ 760$ a) and Pa-233 ($T_{1/2} = 27$ d). Pa-231 was separated from uraninite and Pa-233 from Np-237 solution at the Laboratory of Radiochemistry. Pa-233 was in 9 M HCl + 5% HF solution and Pa-231 in 8 M HCl + 1 M HF solution. Pa-233 was used as radioactive tracer while Pa-231 was used to achieve higher concentration of Pa. The two Pa concentrations of the spiked groundwaters are set out in Table 4. It was assumed in the calculations that all the spike dried onto the teflon platelet used for spiking became dissolved in the groundwater. Tc-99 solution was used to follow the oxidation/reduction equilibrium of the samples under anoxic conditions.

Table 4. Concentrations of protactinium in the sample (mol/l) assuming that all the spiking solution dried into the teflon platelet has dissolved.

Nuclide	Decay mode	Concentration 1	Concentration 2
Pa-233	β	$6.6 \cdot 10^{-13}$ mol/l	$6.6 \cdot 10^{-13}$ mol/l
Pa-231	α	-	$2.9 \cdot 10^{-10}$ mol/l

3. METHODS

3.1 Cation exchange capacities (CEC values)

The cation exchange capacities (CEC values) of the crushed rock samples were determined by AgTU method /1/, in which the exchangeable cations of the sample are replaced with silver-thiourea complex.

About one gramme of crushed rock and 25 ml of AgTU solution were placed in a 50-ml polypropylene centrifuge tube (Sorvall). Samples were shaken continuously for about 18 hours (overnight) and then centrifuged (6500 G, 30 min). Ag^+ and the exchangeable cations, Ca^{2+} , Mg^{2+} , K^+ and Na^+ , were measured from the liquid phase by atomic absorption spectrophotometry (AAS). Some experiments were made with a longer shaking time (about one week) and some with the pH of the AgTU solution adjusted to 1.0.

The CEC values were derived from Equation (1):

$$CEC = \frac{C * V * Z}{m * M} \quad (1)$$

where

- C = concentration of the exchangeable cation (g/ml)
- V = volume of the water (ml)
- Z = charge of the ion
- m = mass of the crushed rock (g)
- M = molecular weight of the cation (g/mol)

If the cation exchange capacity is calculated from more than one exchangeable cation, the CEC value is calculated with Equation (2):

$$CEC = \sum \frac{C_i * V_i * Z_i}{m_i * M_i} \quad (2)$$

The cation exchange capacities of the rocks were also measured by determination of sorption isotherm for Cs. In that case, 3.5 g of crushed rock and 35 ml of Cs solution were placed in a 50-ml polypropylene centrifuge tube. Samples were mixed for one week with 15 minutes of continuous turning end-over-end every hour. After equilibration the samples were centrifuged (6500 G, 30 min) and a 5-ml subsample was taken for Cs-134 gamma measurement. The detector was Wallac Ultrogamma 1280 (NaI(Tl)).

3.2 Sorption experiments

The sorption of the tracer nuclides was studied by batch method. Experiments were made at room temperature and under ambient (oxic) or nitrogen atmosphere (anoxic). The solid/water (m/V) ratio was 1/10 for both oxic and anoxic conditions.

3.2.1 Oxic conditions

Experiments were made in 50-ml polypropylene centrifuge tubes. The samples (three parallel) consisted of 3.5 g of crushed rock and 35 ml of groundwater (OIKR5 groundwater for O11, O12A and O12B rocks, RoKR5 groundwater for Ro1A, Ro1B and Ro1C rocks and KiKR4 groundwater for Ki4A and Ki4B rocks). The samples were mixed for 30 days (a 15-minute period every hour) to achieve steady state between the rock and the groundwater. The groundwater was refreshed three times during the equilibration. Spiking was done by immersing a teflon platelet containing the tracer into the solution. Mixing was continued for three weeks. After that the teflon platelet was removed and the teflon was leached with 1 ml of 8 M HCl + 1 M HF solution to collect the undissolved protactinium for measurement. The samples were centrifuged (6500 G, 30 min) and a 5-ml subsample was taken for measurement.

For desorption studies the whole sorption study solution was removed, the rock sample was weighed and 35 ml of fresh groundwater was added to the sample. The samples were mixed as before for four weeks, centrifuged and treated as after the sorption experiment. The procedure is schematically presented in Figure 1.

Both liquid subsamples were acidified with 100 μ l of 8 M HCl + 1 M HF solution to keep the protactinium in solution. The solutions were measured for Pa-233 by liquid scintillation counting (LSC Wallac 1217 Rackbeta or Wallac Quantulus 1220).

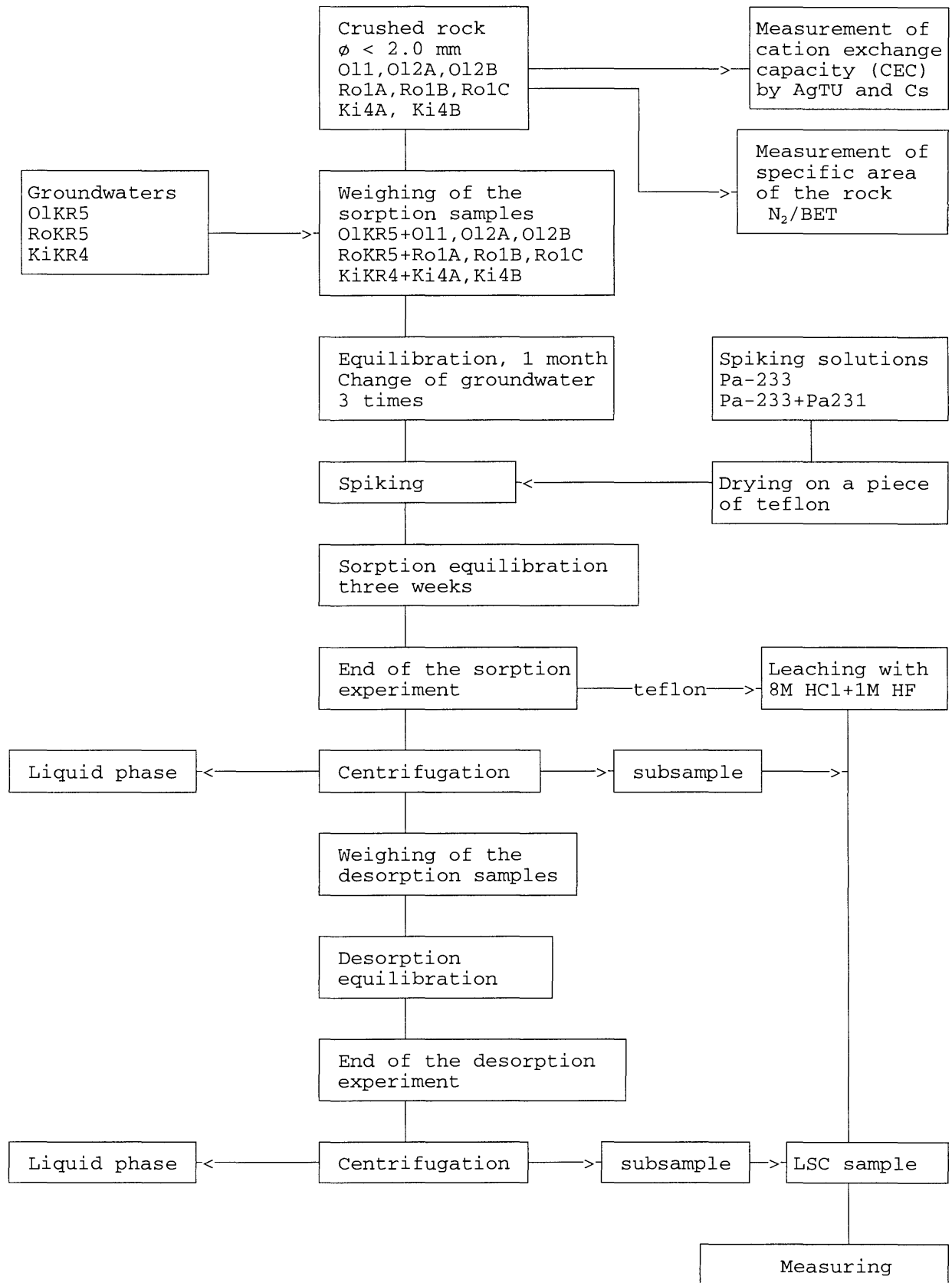


Figure 1. Experimental procedure for the studies on crushed rock under oxic conditions.

The sorption percentages, $S_s(\%)$, were calculated from the measurements with Equation (3):

$$S_s(\%) = \frac{\frac{A_{tracer} - A_{teflon}}{V_{total}} - \frac{A_{sample}}{V_{sample}}}{\frac{A_{tracer} - A_{teflon}}{V_{total}}} * 100\% \quad (3)$$

where A_{tracer} = activity dried on the teflon platelet
 A_{teflon} = activity left on the piece of teflon (undissolved)
 V_{total} = total volume of the solution
 A_{sample} = activity of the measured subsample
 V_{sample} = volume of the measured subsample

The sorption ratio, R_d , was calculated with Equation (4):

$$R_d = \frac{S_s(\%)}{100 - S_s(\%)} * \frac{V}{m} \quad (4)$$

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

$$R_d \text{ (ml/g)} = 10^3 * R_d \text{ (m}^3\text{/kg)}.$$

Surface sorption ratios for crushed rock samples were derived from Equation (5):

$$R_d(m^3/m^2) = \frac{10^{-6} * R_d(ml/g)}{A_s(m^2/g)} \quad (5)$$

where A_s = specific area of the crushed rock

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.

The sorption percentages in desorption experiments (S_{des}) are calculated by Equation (6):

$$S_{des}(\%) = \frac{A_{tracer} * \frac{S_s(\%)}{100\%} + \frac{V_r * A_{tracer}}{V_{s,total}} * (1 - \frac{S_s(\%)}{100\%}) - \frac{A_{de,sample}}{V_{de,sample}} * V_{de,total}}{A_{tracer} * \frac{S_s(\%)}{100\%} + \frac{V_r * A_{tracer}}{V_{s,total}} * (1 - \frac{S_s(\%)}{100\%})} * 100\% \quad (6)$$

where

A_{tracer}	= activity of the spiking solution
$S_s(\%)$	= sorption percentage
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

If the sorption percentage is high (> 90%) the activity of the spiking solution left in the crushed rock can be ignored.

3.2.2 Anoxic conditions

In anoxic experiments, 2.0 g of crushed rock was weighed into a glass vial and the teflon platelet containing the radionuclide was added to the vial. For Romuvaara and Kivetty samples the vials were filled with groundwater flowing from the groundwater sampling line (about 20 ml). All this was done under reduced oxygen (nitrogen) atmosphere in a portable glove bag. The amount of gas above the solution in the vials was 0.5–1.0 ml. The samples and the additional groundwater for the other studies were transported under nitrogen gas atmosphere (AGA 99.999% N_2) to the laboratory nitrogen atmosphere glove box. The Olkiluoto rock samples were prepared in the laboratory nitrogen atmosphere glove box. There was no equilibration of rock and groundwater before spiking the water because the equilibration probably would diminish the reduction capacity of the rock sample.

The sorption time was 30 days. After that the teflon piece was removed, a 5-ml subsample was taken and filtered through a 0.22 μm Millipore filter for LSC measurement, and the pH and Eh values of the solution were measured. The rest of the liquid phase was removed and the glass vial filled to the neck with anoxic groundwater to start the desorption experiment. The desorption time was also about 30 days. Sampling of the water and electrode measurements were done in the same way as in the first phase. The procedure is presented in Figure 2.

The sorption percentages and the R_d values are calculated with Equations 3–6.

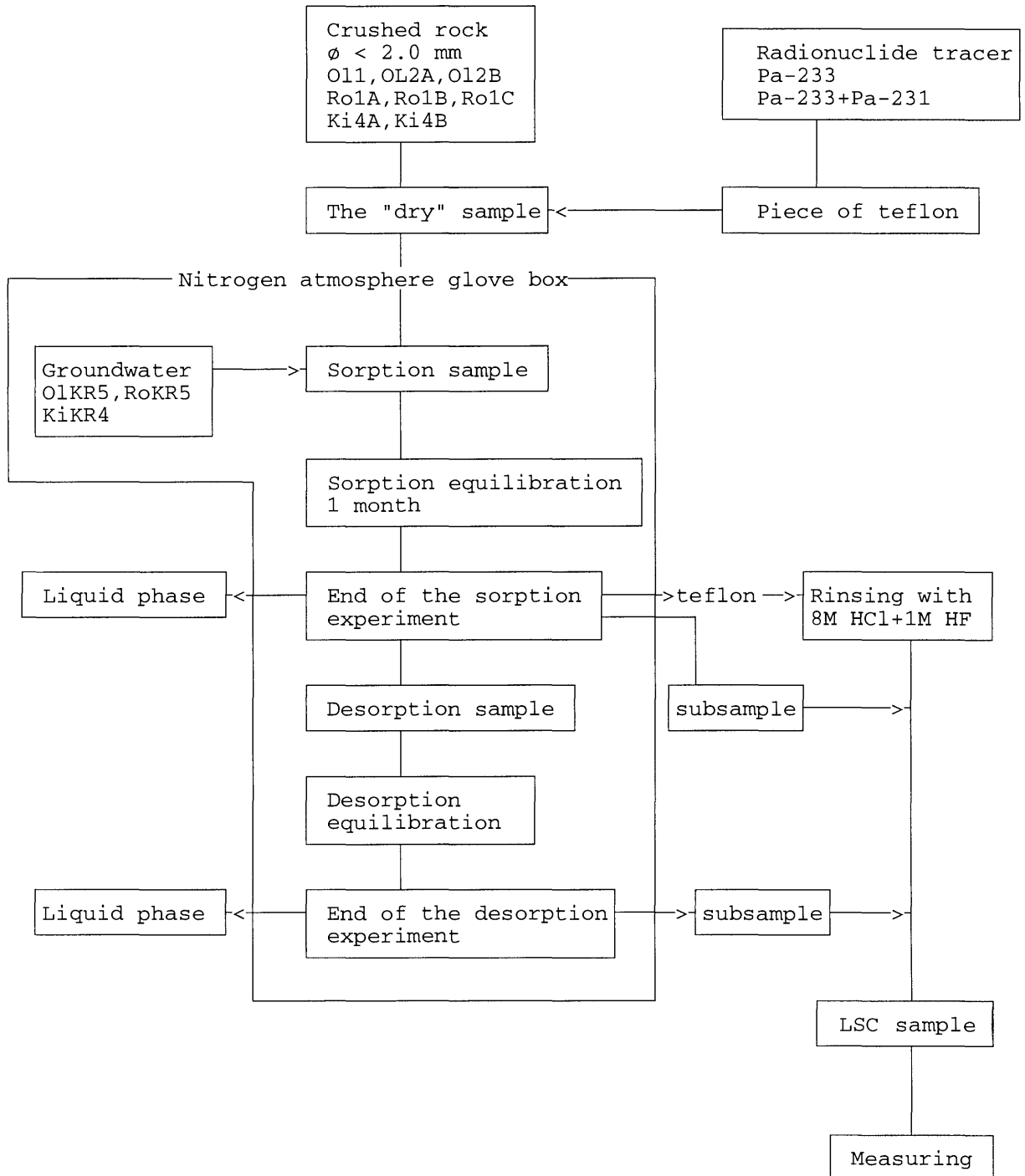


Figure 2. Experimental procedure for the studies on crushed rock under anoxic conditions.

3.3 Sorption on thin section samples

The sorption of the tracers on thin sections was studied by batch and autoradiographic methods /2/. The thin sections were first equilibrated with the groundwaters. The acid solution containing the radioactive Pa-233 tracer was evaporated gently to dryness and then dissolved in the appropriate groundwater. The equilibration water was removed and the thin section covered with 1.0 ml of the freshly spiked groundwater. The exposure time was about 18 hours (over night). After that the spiking solution was removed and the thin section rinsed with 1.0 ml of groundwater and left to dry. The rinsing solution was added to the removed sample solution. The Pa-233 in solution was measured by liquid scintillation counting.

Autoradiographs were made from the spiked thin sections. The film was Kodak X-OMAT MA X-ray film and exposure time was about one month. The autoradiographs were measured by scanner Ricoh FS2 connected to a PC with the Global Lab Image™ program.

The surface sorption ratio for thin sections, R_a , is calculated by Equation (7):

$$R_a = \frac{\frac{S(\%)}{A}}{\frac{L(\%)}{V}} = \frac{S(\%)}{100-S(\%)} * \frac{V}{A} \quad (7)$$

where	$S(\%)$	= amount of the radionuclide sorbed on the thin section as a percentage of the spiking solution
	$L(\%)=100-S(\%)$	= amount of the radionuclide left in the spiking solution
	A	= geometric surface area of the sample (m^2)
	V	= volume of the solution (m^3)

4. RESULTS

4.1 Cation exchange capacities, CEC values

The cation exchange capacities of the crushed rock are presented in Table 5. The table shows the total CEC values derived from changes in the Ag^+ concentration and the CEC (total) values calculated as a sum of the concentrations of the exchangeable cations Ca^{2+} , Mg^{2+} , K^+ and Na^+ . The CEC values from the experiments with longer shaking time, as well as the experiments with pH 1.0 AgTU solution, are included in Table 5.

The shaking time seems to have no effect on the CEC values: the difference in CEC values between 18 hours and one week was negligible. Reducing the pH to 1.0, which was done mainly to remove all free calcite minerals from the rock samples, on the other hand, increased the CEC values significantly, especially for the exchangeable cations K^+ and Mg^{2+} .

Table 6 summarizes the cation exchange capacities measured for caesium. The values are the mean values of three parallel samples. The results for individual samples are presented in Appendix B. The mass distribution ratios, R_d values, of caesium from CsCl solutions on Olkiluoto, Romuvaara and Kivetty rocks are displayed in Figures 3–5.

The CEC values measured for Cs were comparable to the CEC values measured for AgTU (Ag). With OI2A, OI2B and all Romuvaara rocks the CEC values for caesium were smaller than for Ag, which is probably due to too low Cs concentration to saturate all the cation exchange places. For Romuvaara rocks the CEC value measured for Ag differed from the CEC (total) value measured according to the exchangeable cations. This is caused by the release of Ca^{2+} , which is equivalent to about 90% of the total CEC. The high amount of Ca in water is probably due to both cation exchange as well as solution of some Ca containing mineral.

Table 5. Cation exchange capacities, CEC values, of the crushed rock samples ($\phi < 2.0$ mm). Results are the mean values of three parallel samples.

Rock	Method	Na ⁺ (meq/100g)	K ⁺ (meq/100g)	Ca ²⁺ (meq/100g)	Mg ²⁺ (meq/100g)	total Σ (meq/100g)	CEC/BET (meq/m ²)	Ag ⁺ (meq/100g)	CEC/BET (meq/m ²)
O11 mica gneiss	AgTU	0.80 ± 0.07	0.23 ± 0.02	2.2 ± 0.3	0.094 ± 0.002	3.3 ± 0.3	0.034	1.5 ± 0.4	0.016
	AgTU(long)	0.88 ± 0.08	0.29 ± 0.02	1.7 ± 0.4	0.17 ± 0.02	3.0 ± 0.4	0.032	0.98 ± 0.20	0.010
	AgTU(pH1)	0.71 ± 0.08	1.6 ± 0.1	1.6 ± 0.4	2.8 ± 0.1	6.7 ± 0.5	0.071	13 ± 1	0.13
O12A granodiorite (tonalite)	AgTU	0.20 ± 0.04	0.20 ± 0.05	1.0 ± 0.2	0.047 ± 0.005	1.4 ± 0.2	0.064	2.1 ± 0.4	0.095
	AgTU(long)	0.20 ± 0.02	0.14 ± 0.01	1.1 ± 0.1	0.088 ± 0.011	1.5 ± 0.1	0.068	0.82 ± 0.37	0.037
	AgTU(pH1)	0.28 ± 0.01	2.3 ± 0.2	6.6 ± 0.3	4.3 ± 0.1	13 ± 1	0.59	12 ± 2	0.55
O12B granodiorite (granite)	AgTU	0.28 ± 0.01	0.16 ± 0.03	1.5 ± 0.1	0.021 ± 0.001	2.0 ± 0.1	0.056	1.8 ± 0.4	0.050
	AgTU(long)	0.25 ± 0.01	0.15 ± 0.02	1.5 ± 0.2	0.032 ± 0.004	1.9 ± 0.2	0.053	0.82 ± 0.41	0.023
	AgTU(pH1)	0.32 ± 0.04	0.28 ± 0.23	3.0 ± 0.1	0.52 ± 0.32	4.1 ± 0.4	0.11	12 ± 1	0.33
Ro1A tonalite	AgTU	0.19 ± 0.01	0.12 ± 0.03	9.1 ± 0.4	0.11 ± 0.01	9.5 ± 0.4	0.34	1.8 ± 0.6	0.064
	AgTU(long)	0.14 ± 0.01	0.14 ± 0.01	8.4 ± 0.8	0.16 ± 0.01	8.8 ± 0.8	0.31	1.4 ± 0.5	0.050
	AgTU(pH1)	0.76 ± 0.16	2.3 ± 0.2	22 ± 1	4.0 ± 0.2	28 ± 1	1.0	12 ± 1	0.43
Ro1B granite (leukotonalite)	AgTU	0.24 ± 0.02	0.09 ± 0.04	4.7 ± 0.3	0.051 ± 0.002	5.1 ± 0.3	0.22	1.8 ± 0.3	0.078
	AgTU(long)	0.19 ± 0.02	0.10 ± 0.01	4.1 ± 0.1	0.076 ± 0.006	4.5 ± 0.1	0.20	1.1 ± 0.4	0.048
	AgTU(pH1)	0.68 ± 0.03	0.59 ± 0.02	8.5 ± 6.7	1.4 ± 0.1	11 ± 7	0.48	12 ± 1	0.52
Ro1C mica gneiss	AgTU	0.15 ± 0.02	0.11 ± 0.04	8.6 ± 0.5	0.11 ± 0.01	8.9 ± 0.5	0.49	1.3 ± 0.4	0.072
	AgTU(long)	0.14 ± 0.08	0.32 ± 0.26	7.6 ± 0.6	0.16 ± 0.01	8.2 ± 0.7	0.46	1.7 ± 1.5	0.094
	AgTU(pH1)	0.62 ± 0.23	2.6 ± 0.5	2.9 ± 1.1	2.3 ± 0.1	8.4 ± 1.3	0.47	12 ± 1	0.67
Ki4A porph. granite	AgTU	0.10 ± 0.01	0.10 ± 0.01	2.7 ± 0.1	0.057 ± 0.003	2.9 ± 0.1	0.14	1.2 ± 0.4	0.057
	AgTU(long)	0.06 ± 0.01	0.10 ± 0.04	2.6 ± 0.2	0.075 ± 0.011	2.8 ± 0.3	0.13	0.99 ± 0.24	0.047
	AgTU(pH1)	0.15 ± 0.02	1.3 ± 0.4	2.5 ± 0.3	3.1 ± 0.3	7.0 ± 0.6	0.33	13 ± 1	0.62
Ki4B porph. granite (granodiorite)	AgTU	0.15 ± 0.03	0.08 ± 0.02	2.8 ± 0.3	0.059 ± 0.007	2.8 ± 0.3	0.13	1.9 ± 0.2	0.086
	AgTU(long)	0.13 ± 0.04	0.10 ± 0.01	2.9 ± 0.3	0.096 ± 0.017	3.2 ± 0.3	0.15	1.7 ± 0.4	0.077
	AgTU(pH1)	0.25 ± 0.04	2.9 ± 0.5	4.2 ± 0.1	6.5 ± 0.3	13 ± 1	0.59	12 ± 1	0.55

rock names in parentheses are "field names"

AgTU(long) = shaking time 1 week

Table 6. Sorption of caesium on crushed rock.

Rock	[Cs] (mol/l)	Sorption%	R_d (m ³ /kg)	CEC (meq/100 g)
O11 mica gneiss	$1.11 \cdot 10^{-2}$	14.6 ± 0.3	0.0016–0.0018	1.61 ± 0.03
	$1.16 \cdot 10^{-4}$	95.6 ± 0.3	0.20–0.25	
	$1.22 \cdot 10^{-6}$	98.8 ± 0.2	0.73–0.96	
	$1.16 \cdot 10^{-7}$	99.6 ± 0.2	1.9–3.2	
O12A granodiorite (tonalite)	$1.11 \cdot 10^{-2}$	5.98 ± 0.58	$(0.58-0.72) \cdot 10^{-3}$	0.66 ± 0.07
	$1.16 \cdot 10^{-4}$	88.3 ± 0.57	0.034–0.15	
	$1.22 \cdot 10^{-6}$	97.6 ± 9.5	0.32–0.50	
	$1.16 \cdot 10^{-7}$	97.9 ± 1.8	0.24–1.1	
O12B granodiorite (granite)	$1.11 \cdot 10^{-2}$	4.88 ± 1.54	$(0.33-0.71) \cdot 10^{-3}$	0.54 ± 0.17
	$1.16 \cdot 10^{-4}$	38.1 ± 2.7	0.055–0.069	
	$1.22 \cdot 10^{-6}$	86.2 ± 4.9	0.042–0.086	
	$1.16 \cdot 10^{-7}$	97.2 ± 1.3	0.22–0.50	
Ro1A tonalite	$1.11 \cdot 10^{-2}$	6.91 ± 0.47	$(0.69-0.80) \cdot 10^{-3}$	0.76 ± 0.06
	$1.16 \cdot 10^{-4}$	64.9 ± 0.8	0.018–0.019	
	$1.22 \cdot 10^{-6}$	94.1 ± 0.4	0.15–0.17	
	$1.16 \cdot 10^{-7}$	95.6 ± 2.3	0.16–0.56	
Ro1B granite (leukotonalite)	$1.11 \cdot 10^{-2}$	4.61 ± 0.67	$(0.41-0.57) \cdot 10^{-3}$	0.51 ± 0.08
	$1.16 \cdot 10^{-4}$	53.2 ± 8.6	0.0078–0.015	
	$1.22 \cdot 10^{-6}$	87.8 ± 2.7	0.056–0.088	
	$1.16 \cdot 10^{-7}$	94.9 ± 2.3	0.14–0.39	
Ro1C mica gneiss	$1.11 \cdot 10^{-2}$	5.99 ± 1.5	$(0.45-0.80) \cdot 10^{-3}$	0.66 ± 0.17
	$1.16 \cdot 10^{-4}$	80.7 ± 12.6	0.020–0.083	
	$1.22 \cdot 10^{-6}$	90.7 ± 3.1	0.069–0.13	
	$1.16 \cdot 10^{-7}$	96.2 ± 2.2	0.18–0.73	
Ki4A porph. granite	$1.11 \cdot 10^{-2}$	9.62 ± 0.55	$(0.96-1.1) \cdot 10^{-3}$	1.06 ± 0.06
	$1.16 \cdot 10^{-4}$	43.9 ± 0.8	$(7.5-7.8) \cdot 10^{-3}$	
	$1.22 \cdot 10^{-6}$	87.5 ± 4.4	0.046–0.092	
	$1.16 \cdot 10^{-7}$	93.4 ± 1.9	0.10–0.17	
Ki4B porph. granite (granodiorite)	$1.11 \cdot 10^{-2}$	11.4 ± 1.1	$(1.1-1.3) \cdot 10^{-3}$	1.26 ± 0.11
	$1.16 \cdot 10^{-4}$	72.2 ± 14.4	0.015–0.075	
	$1.22 \cdot 10^{-6}$	89.5 ± 4.9	0.051–0.13	
	$1.16 \cdot 10^{-7}$	96.0 ± 0.8	0.19–0.26	

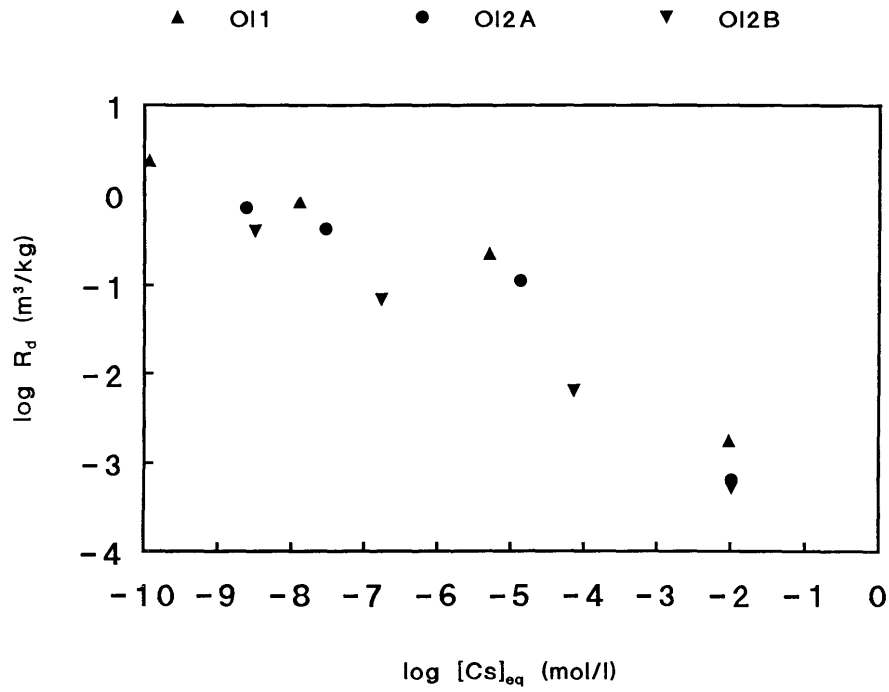


Figure 3. Sorption of caesium, R_d values, on Olkiluoto rock samples from CsCl solution. ($[Cs]_{eq}$ = conc. of Cs in equilibrium)

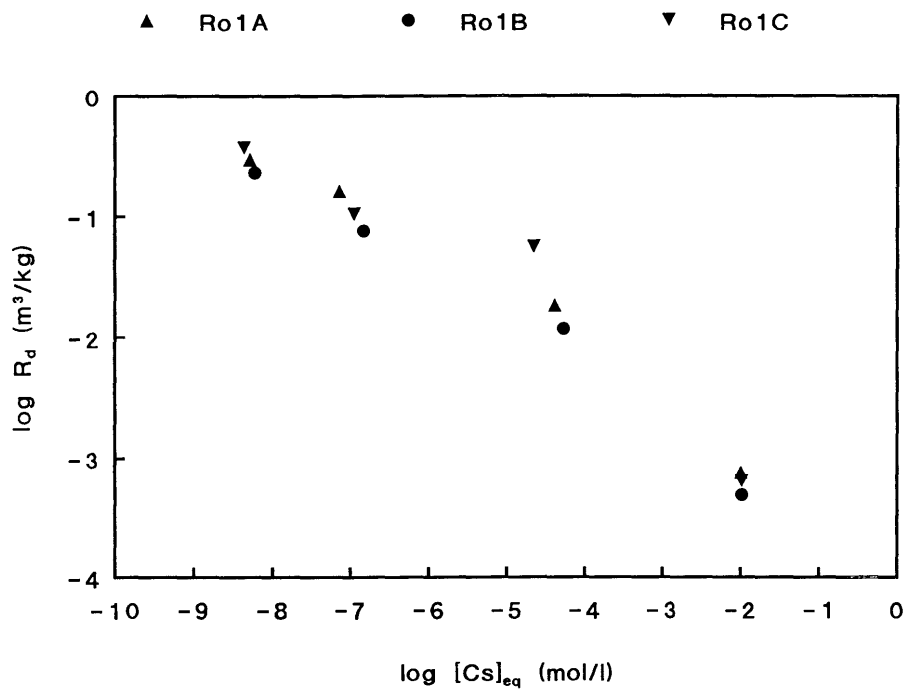


Figure 4. Sorption of caesium, R_d values, on Romuvaara rock samples from CsCl solution. ($[Cs]_{eq}$ = conc. of Cs on equilibrium)

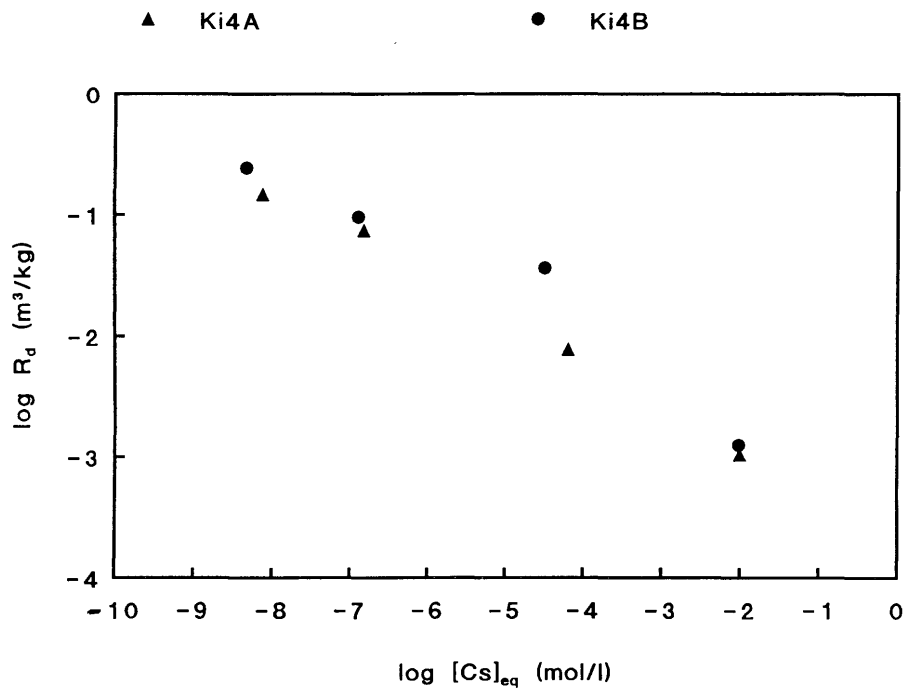


Figure 5. Sorption of caesium, R_d values, on Kivetty rock samples from CsCl solution. ($[Cs]_{eq}$ = conc. of Cs on equilibrium)

4.2 Experiments with crushed rock, R_d values

Protactinium is an actinoid element which mainly exists in oxidation state +5. Although Pa(IV) is possible as well, in a non-tightly closed vessel it is oxidized quantitatively to Pa(V) after only 72 and 144 hours in 5.3 M and 11.5 M HCl, respectively [3]. The sorption studies were made under both oxidic and anoxic conditions.

4.2.1 Oxidic conditions

The sorption percentages and the R_d values for sorption and desorption are presented in Table 7. The sorption percentages are the mean values of three parallel samples, but the R_d values are presented as variation between parallel samples ($R_{d,min}$ – $R_{d,max}$). The detailed results, i.e. the sorption percentages of all parallel samples, are presented in Appendix B.

Table 7. Sorption and desorption (sorption percentages and R_d values) of protactinium under oxic conditions in site-specific groundwaters.

Rock	$\log C_{Pa}$ C_{Pa} mol/l	Undissolved fraction (%)	S_s (%)	R_d (m^3/kg)	S_{des} (%)	$R_{d,des}$ (m^3/kg)
Ol1 mica gneiss	-13	0.48±0.12	99.4±0.2	1.4–3.1	99.0±0.3	0.63–0.76
	-10	0.38±0.14	99.3±0.2	1.3–2.1	98.9±0.3	0.62–0.80
Ol2A granodiorite	-13	0.70±0.27	99.0±0.2	0.94–1.3	98.3±0.5	0.38–0.46
	-10	0.58±0.17	98.8±0.3	0.79–1.1	98.1±0.5	0.35–0.44
Ol2B granodiorite	-13	0.63±0.28	98.9±0.0	0.78–1.1	97.8±0.6	0.28–0.35
	-10	0.82±0.22	98.7±0.3	0.71–1.1	98.4±0.4	0.41–0.51
Ro1A tonalite	-13	0.58±0.19	98.4±0.3	0.53–0.74	98.3±1.2	0.31–1.9
	-10	0.38±0.11	98.4±0.9	0.43–1.8	97.8±0.7	0.24–0.45
Ro1B granite	-13	1.0±0.4	97.1±0.8	0.27–0.49	96.6±1.1	0.14–0.26
	-10	0.59±0.13	95.2±1.3	0.16–0.29	96.0±1.7	0.11–0.30
Ro1C mica gneiss	-13	1.0±0.2	98.3±0.3	0.52–0.73	98.0±0.5	0.31–0.51
	-10	0.89±0.10	97.0±1.5	0.20–0.50	96.8±1.5	0.14–0.36
Ki4A porphyritic granite	-13	0.57±0.30	99.4±0.2	1.8–2.4	99.4±0.4	0.79–3.2
	-10	0.69±0.19	99.0±0.2	0.96–1.4	99.2±0.2	0.75–1.1
Ki4B porphyritic granite	-13	0.46±0.04	98.9±0.8	0.51–1.7	98.9±0.7	0.39–1.1
	-10	0.41±0.09	99.3±0.2	1.4–1.7	99.3±0.2	1.0–1.6

The sorption of protactinium was high for all samples, but lowest for Romuvaara rocks from RoKR5 groundwater, where the R_d values were 0.2–1.8 m^3/kg . Sorption on Olkiluoto and on Kivetty rocks was almost the same, the R_d values being mostly about 1.0 m^3/kg . The concentration of protactinium had no effect on the sorption.

4.2.2 Anoxic conditions

The sorption percentages and the R_d values for different water-rock combinations are presented in Tables 8–10. The sorption percentages are the mean values of three parallel samples and the R_d values are presented as variation between samples. The detailed results are presented in Appendix B.

Table 8. Sorption of protactinium, sorption percentages and R_d values, on Romuvaara rock samples from RoKR5 groundwater under anoxic conditions.

Rock	$\log C_{Pa}$	Undissolved fraction (%)	S_s (%)	R_d (m ³ /kg)	S_{des} (%)	$R_{d,des}$ (m ³ /kg)
Ro1A tonalite	-13	10.1±3.6	93.0±5.7	0.07–0.73	86.3±4.3	0.05–0.10
	-10	4.70±2.86	97.1±1.3	0.25–0.38	90.8±8.2	0.05–0.62
Ro1B granite	-13	5.36±2.26	94.8±6.0	0.08–1.0	87.0±9.4	0.04–0.41
	-10	8.75±0.35	93.0±5.9	0.08–1.1	86.0±7.1	0.05–0.17
Ro1C mica gneiss	-13	4.78±2.13	98.7±0.4	0.66–1.3	96.5±1.7	0.21–0.61
	-10	5.82±2.59	97.4±1.9	0.25–2.3	89.5±7.6	0.04–0.22

The RoKR5 groundwater contained some dissolved oxygen at collection, so the experiments with Romuvaara crushed rock samples were also made with KiKR4 and OIKR5 groundwaters.

Because of expected difficulties in Eh measurements several Tc-99 samples were spiked with Tc(VII)-99 to check the redox conditions of the rock/water system. The concentrations of technetium in the liquid phase are presented in Table 11 and the detailed results in Appendix B.

Table 9. Sorption and desorption of protactinium, sorption percentages and R_d values, on Kivetty and Romuvaara rock samples from KiKR4 groundwater under anoxic conditions.

Rock	$\log C_{Pa}$	Undissolved fraction (%)	S_s (%)	R_d (m ³ /kg)	S_{des} (%)	$R_{d,des}$ (m ³ /kg)
Ki4A porph. granite	-13	6.41±1.83	99.5±0.2	1.7–4.2	99.2±0.6	>0.92
	-10	9.09±1.57	99.5±0.1	2.3–3.4	98.6±0.9	0.52–3.2
Ki4B porph. granite (granodiorite)	-13	10.1±1.12	99.6±0.1	2.8–4.3	99.1±0.7	>0.73
	-10	9.16±4.28	99.7±0.2	>3.2	99.0±0.6	0.71–2.9
Ro1A tonalite	-13	8.61±1.5	99.2±0.5	>1.3	97.2±1.8	0.25–0.70
	-10	11.6±2.7	99.8±0.2	>4.0	99.1±0.5	0.88–2.4
Ro1B granite	-13	10.9±1.5	99.3±0.2	1.4–1.9	97.9±1.2	0.35–0.85
	-10	10.2±0.7	99.0±0.9	0.70–3.1	98.9±0.6	0.9
Ro1C mica gneiss	-13	16.7±1.0	99.2±0.8	0.87–4.4	97.4±2.4	0.25–1.3
	-10	13.2±6.5	99.5±0.4	>1.6	98.2±0.3	0.56–0.72

Table 10. Sorption and desorption of protactinium, sorption percentages and R_d values, on Olkiluoto, Romuvaara and Kivetty rock samples from OIKR5 groundwater under anoxic conditions.

Rock	$\log C_{Pa}$	Undissolved fraction (%)	S_s (%)	R_d (m ³ /kg)	S_{des} (%)	$R_{d,des}$ (m ³ /kg)
Ol1 mica gneiss	-13	12.2±3.7	98.9±0.3	0.78–1.3	99.5±0.2	1.9–4.1
	-10	6.23±1.32	98.3±1.2	0.31–1.1	99.5±0.4	>1.2
Ol2A granodiorite	-13	8.09±6.93	99.0±0.4	0.73–1.6	99.0±0.2	0.96–1.4
	-10	6.21±1.88	99.2±0.2	1.1–2.0	99.4±0.2	1.4–2.7
Ol2B granodiorite	-13	9.80±4.96	99.1±0.2	1.0–1.5	96.2±2.7	0.18–1.5
	-10	5.93±0.95	98.9±1.0	0.46–2.4	99.2±0.2	1.1–2.0
Ro1A tonalite	-13	13.2±4.1	98.3±1.2	0.35–1.2	99.2±0.1	1.1–1.7
Ro1B granite	-13	15.8±3.4	98.2±0.8	0.38–0.84	99.0±0.4	0.95–1.9
Ro1C mica gneiss	-13	11.6±5.6	99.0±0.4	0.82–1.7	99.0±0.3	0.97–1.7
Ki4A porph. granite	-13	14.9±8.1	98.6±1.0	0.41–1.4	99.1±0.1	1.1–1.4
Ki4B porph. granite	-13	16.0±4.3	99.2±0.1	1.2–1.6	98.9±0.1	0.98–1.2

Table 11. Fraction of Tc-99 spike in groundwater/crushed rock samples under anoxic conditions (spiking time two weeks).

Groundwater	Rock	Tc-99 in water
RoKR5	no rock	82.8%
	Ro1A	27.3%
KiKR4	no rock	52.0%
	Ki4A	0.30%
OIKR5	no rock	87.3%
	Ol1	14.3%
	Ol2A	6.3%
	Ol2B	8.0%

The partition of technetium in the samples was such as to imply that conditions were reducing for Tc in groundwater/rock systems /4/. Although the concentration of Tc-99 was greater in the RoKR5 groundwater than the other groundwaters, the sorption percentage of over 70% indicated the conditions to be at least mildly reducing for technetium.

The pH and Eh(Pt) values of the samples were also measured, but because of the lack of a suitable redox pair, the Eh values of the RoKR5 and KiKR4 groundwaters differed between parallel samples. The readings were also associated with uncertainties. The measured Eh values for RoKR5 groundwater were -121– +464 mV and for KiKR4 groundwater +54– +364 mV and -266– +300 mV for Kivetty and Romuvaara rock samples, respectively. For OIKR5 and Palmottu groundwaters there was a suitable redox pair, so the Eh values did not differ so much between parallel samples. The Eh values for OIKR5 were -9– -106 mV, -13– -161 mV and -36– -89 mV for Olkiluoto, Romuvaara and Kivetty rock samples, respectively. The pH values were 8.0–8.6 (mainly 8.3), 7.7–9.1 (mainly 8.0) and 8.1–8.7 (mainly 8.4) for OIKR5, RoKR5 and KiKR4 groundwaters, respectively. The results of the pH and of the Eh measurements are presented in Appendix B, along with the sorption percentages and R_d values for each parallel sample.

4.2.3 Palmottu crushed rock samples

The sorption experiments for Palmottu samples were made under both oxic and anoxic conditions. The results are presented in Table 12.

Table 12. Sorption and desorption of protactinium, sorption percentages and R_d values, on Palmottu crushed rock from Palmottu groundwater (R385/403*) ($\phi < 2.0$ mm, [Pa] = 10^{-13} mol/l).

Conditions	Rock	Undissolved fraction (%)	Sorption (%)	R_d (m^3/kg)	Desorption (%)	$R_{d,des}$ (m^3/kg)
Oxic	R373/139.7	0.26±0.23	99.8±0.1	> 1.02		
	R373/146.7	0.22±0.09	99.9±0.1	> 2.03		
Anoxic	R373/139.7	6.48±1.76	99.6±0.2	2.0–4.4	99.6±0.2	2.1–3.7
	R373/146.7	7.20±7.10	99.4±0.5	1.0–4.8	99.3±0.4	0.95–3.6

* no chemical composition

4.3 Rock thin sections, R_a values

The sorption percentages and R_a values for the rock thin section studies are presented in Tables 13–15.

Table 13. Sorption (sorption percentages and R_a values) of protactinium on Olkiluoto rock thin sections.

OIKR5 groundwater			KiKR4 groundwater		
Rock	Sorption (%)	$R_a \cdot 10^{-5}$ (m^3/m^2)	Rock	Sorption (%)	$R_a \cdot 10^{-5}$ (m^3/m^2)
O11	69.8 ± 2.5	350–420	O11	27.5 ± 1.5	58–63
O12A	74.9 ± 1.4	420–540	O12A	28.0 ± 0.8	60–66
O12B	72.9 ± 1.1	440–480	O12B	67.7 ± 1.8	320–390

Table 14. Sorption (sorption percentages and R_a values) of protactinium on Romuvaara rock thin sections.

RoKR5 groundwater			OIKR5 groundwater		
Rock	Sorption (%)	$R_a \cdot 10^{-5}$ (m^3/m^2)	Rock	Sorption (%)	$R_a \cdot 10^{-5}$ (m^3/m^2)
Ro1A	5.82 ± 3.07	4.4–16	Ro1A	66.4 ± 4.5	270–400
Ro1B	7.41 ± 2.63	8.5–17	Ro1B	73.2 ± 0.7	190–460
Ro1C	6.60 ± 1.23	9.3–14	Ro1C	76.2 ± 1.4	470–570

Table 15. Sorption (sorption percentages and R_a values) of protactinium on Kivetty rock thin sections.

KiKR4 groundwater			OIKR5 groundwater		
Rock	Sorption (%)	$R_a \cdot 10^{-5}$ (m^3/m^2)	Rock	Sorption (%)	$R_a \cdot 10^{-5}$ (m^3/m^2)
Ki4A	4.40 ± 0.94	5.9–7.7	Ki4A	68.6 ± 5.7	260–410
Ki4B	5.06 ± 1.74	4.5–9.1	Ki4B	13.6 ± 13.6	23

The sorption experiments were made under oxic conditions. The R_a values varied widely with the groundwater. The results for the parallel samples were very different, moreover probably because of the different method of rinsing the samples after removing the spiking solution. With fresh groundwater the thin sections were rinsed three times and with saline groundwater only once. The rinsing method may be one cause of the higher R_a values for saline, OIKR5, groundwater. Another cause could be a slight precipitation of Pa, although the autoradiographs showed no such marks. The precipitated Pa could anyhow be removed by rinsing water. The autoradiographs of the rock thin sections were weak and not very sharp (β decay). No mineral specific sorption was observed which indicates to precipitation.

5. CONCLUSIONS

The sorption of protactinium was high on all rocks under both anoxic and oxic conditions. The R_d values varied from 0.07 m³/kg to 6.8 m³/kg under anoxic conditions and from 0.2 m³/kg to 2.4 m³/kg under oxic conditions. The main difference between the anoxic and oxic conditions was the solubility of the spiking solution: under anoxic conditions about ten percent of the dried spike was left on the teflon, whereas under oxic conditions all spiking solution was removed from the teflon platelet into the liquid phase.

Under anoxic conditions there was no difference between the groundwaters in the amount of undissolved fraction of the spiking solution. The undissolved fraction was 5.9–16%, 6.4–17% and 4.7–10% for OIKR5, KiKR4 and RoKR5 groundwaters, respectively. The fraction of undissolved Pa was slightly smaller with RoKR5 groundwater, probably because this groundwater was initially oxidizing (Table 3).

The sorption of protactinium was lowest on Romuvaara rocks from RoKR5 groundwater. The R_d values were 0.07–2.3 m³/kg under anoxic conditions and 0.27–1.8 m³/kg under oxic conditions. A better sorption value (R_d) was obtained for KiKR4 groundwater under anoxic conditions. The R_d values from OIKR5 groundwater under anoxic conditions were about the same as from RoKR5 groundwater. The R_d values were 0.70–8.6 m³/kg and 0.35–1.7 m³/kg for KiKR4 and OIKR5 groundwater, respectively. Eh (both field and laboratory) values were lower for the KiKR4 and OIKR5 groundwaters than the RoKR5 groundwater (Table 3).

The sorption was slightly higher on Kivetty rocks than Romuvaara rocks: the R_d values were > 1.7 m³/kg and 0.51–2.4 m³/kg for KiKR4 groundwater under anoxic and oxic conditions, respectively. The sorption on Kivetty rocks under anoxic conditions was also studied in OIKR5 saline groundwater. The R_d values were slightly lower, 0.41–1.6 m³/kg, presumably due to the higher salinity of the OIKR5 groundwater.

The sorption of protactinium on Olkiluoto rock samples from OIKR5 groundwater was about the same as the sorption on Kivetty rock samples from KiKR4 groundwater. The R_d values were 0.31–2.4 m³/kg under anoxic conditions and 0.71–3.1 m³/kg under oxic conditions. Under anoxic conditions the sorption was slightly higher on Kivetty rock samples from KiKR4 groundwater ($R_d > 1.7$ m³/kg).

Protactinium is known to hydrolyse strongly. The mechanism of the sorption of protactinium is not known, but in this study its disappearance from the water phase is not likely to have been due to precipitation. In their study Saarinen et al. found about $1.2 \cdot 10^{-13}$ mol/kg of protactinium in Palmottu groundwaters /5/, which is a higher concentration than the theoretical maximum for the lower amount of spike in this study. If the controlling process were precipitation, the "sorption" should be higher for higher Pa concentration. According to our study the concentration of Pa had no effect on the sorption.

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APPENDIX A.

Table 1. Mineral composition of thin sections (Vol.-%) calculated by point counting method (500 points/sample).

Table 2. Chemical composition of the OlKR5, RoKR5 and KiKR4 groundwaters (IVO Chem.Lab.)

Table 1. Mineral composition of thin sections (Vol.-%) calculated by point counting method (500 points/sample).

Minerals	O11/11	O12A/9	O12B/9	Ro1A/9	Ro1B/10	Ro1C/11	Ki4A/6	Ki4B/4
K-feldspar	19.2	16.6	15.2	-	30.2	0.2	26.4	36.6
Plagioclase	14.8	38.6	38.4	25.4	24.8	27.0	23.6	25.4
Quartz	16.6	26.0	35.8	44.4	36.4	38.8	39.0	20.0
Biotite	32.4	6.6	0.2	16.4	2.6	31.2	5.4	12.0
Muscovite	2.4	3.6	9.2	12.2	3.8	0.2	0.8	0.8
Chlorite	-	6.4	0.2	+	0.6	0.2	-	-
Hornblende	-	-	-	-	+?	-	4.0	4.4
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	-	-	-
Opakes	1.6	0.2	-	0.4	+	1.0	0.6	0.2

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.

O12B/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.

Table 2. Chemical composition of the OIKR5, RoKR5 and KiKR4 groundwaters (IVO Chem. Lab.)

	RO-KR5/T1 13.11.1995	KI-KR4/T1 20.11.1995	OL-KR5/T1 30.10.1995
Eh(Pt)F (mV)	300	-75	-270
O ₂ F (mg/l)	2.70	0.00	0.00
CondF (mS/m)	16.0	14.0	1981
CondL (mS/m)	18.0	17.0	2330
pHF	10.1	8.4	8.3
pHL	9.3	8.2	8.2
AlkF(m) (meq/l)	1.52	1.40	0.30
AlkF(p) (meq/l)	0.28	0.24	0.10
CO _{2, tot} (mg/l)	53.8	58.20	4.20
DOC (mg/l)	2.0	7.2	1.2
Fe _{tot} Fz (mg/l)	0.050	0.600	0.084
Fe _{tot} AAS (mg/l)	0.059	0.520	0.230
Fe ₂ +Fz (mg/l)	<0.01	0.150	0.076
SiO ₂ (mg/l)	13.0	13.0	6.3
Al (mg/l)	0.071	0.030	0.032
Na (mg/l)	39.0	18.0	3330
K (mg/l)	0.69	0.87	15.0
Ca (mg/l)	2.1	13.0	2100
Mg (mg/l)	0.08	3.0	67.0
Mn (mg/l)	0.001	0.210	0.360
Cs (mg/l)	<0.005	<0.005	0.008
S _{tot} (mg/l)	1.44	0.73	1.50
S(-II)F (mg/l)	0.066	0.025	0.160
S(-II)L (mg/l)	0.04	0.04	0.30
SO ₄ (mg/l)	3.9	2.00	3.30
P _{tot} (mg/l)	0.15	<0.01	<0.01
PO ₄ (mg/l)	0.44	0.02	<0.01
Cl (mg/l)	4.3	3.20	9200
F (mg/l)		3.00	1.30
Br (mg/l)	0.10	0.03	58.0
Uranine FL(μg/l)	4.0	4.0	<1
H-3 (TU)	0.9	1.9	2.0
U-238(H ₂ O) (mBq/l)	5.4	39.0	0.32
U-238(H ₂ O) (ppb)	0.439	3.190	0.026
U-234/U-238 (Bq/Bq)	3.83	3.44	2.07
U-238(part.) (mBq/l)	<0.2	<0.07	<0.11
U238(part.) (ppb)	<0.02	<0.01	<0.01
Ionic strength	1.5·10 ⁻³	1.9·10 ⁻³	3.1·10 ⁻¹

F = field measurement or analysis in the field laboratory

L = laboratory measurement or analysis

FL = fluorimetry

Fz = Ferrozine analytical method

APPENDIX B

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Table 1. Sorption of caesium on Olkiluoto rock samples from CsCl solution under oxic conditions.

Sample	[Cs] _{initial} (mol/l)	Sorption%	R _d (m ³ /kg)
Ol1Cs1-1	1.11·10 ⁻²	14.3	1.69·10 ⁻³
Ol1Cs1-2		14.7	1.75·10 ⁻³
Ol1Cs1-3		14.8	1.76·10 ⁻³
Ol1Cs2-1	1.16·10 ⁻⁴	95.3	0.209
Ol1Cs2-2		95.5	0.219
Ol1Cs2-3		95.9	0.245
Ol1Cs3-1	1.22·10 ⁻⁶	98.6	0.737
Ol1Cs3-2		98.8	0.869
Ol1Cs3-3		98.9	0.958
Ol1Cs4-1	1.16·10 ⁻⁷	99.6	2.12
Ol1Cs4-2		99.7	3.22
Ol1Cs4-3		99.5	1.94
Ol2ACs1-1	1.11·10 ⁻²	6.59	7.19·10 ⁻⁴
Ol2ACs1-2		5.90	6.36·10 ⁻⁴
Ol2ACs1-3		5.46	5.87·10 ⁻⁴
Ol2ACs2-1	1.16·10 ⁻⁴	93.7	0.151
Ol2ACs2-2		93.7	0.151
Ol2ACs2-3		77.2	0.0348
Ol2ACs3-1	1.22·10 ⁻⁶	98.0	0.495
Ol2ACs3-2		97.8	0.450
Ol2ACs3-3		96.9	0.324
Ol2ACs4-1	1.16·10 ⁻⁷	99.0	1.07
Ol2ACs4-2		98.8	0.868
Ol2ACs4-3		96.0	0.243
Ol2BCs1-1	1.11·10 ⁻²	6.48	7.12·10 ⁻⁴
Ol2BCs1-2		3.15	3.31·10 ⁻⁴
Ol2BCs1-3		4.74	5.10·10 ⁻⁴
Ol2BCs2-1	1.16·10 ⁻⁴	40.2	6.93·10 ⁻³
Ol2BCs2-2		38.9	6.54·10 ⁻³
Ol2BCs2-3		35.0	5.54·10 ⁻³
Ol2BCs3-1	1.22·10 ⁻⁶	89.4	0.0856
Ol2BCs3-2		88.7	0.0807
Ol2BCs3-3		80.6	0.0425
Ol2BCs4-1	1.16·10 ⁻⁷	97.9	0.470
Ol2BCs4-2		98.0	0.497
Ol2BCs4-3		95.8	0.226

Table 2. Sorption of caesium on Romuvaara rock samples from CsCl solution under oxic conditions.

Sample	[Cs] _{initial} (mol/l)	Sorption%	R _d (m ³ /kg)
Ro1ACs1-1	1.11·10 ⁻²	6.38	6.91·10 ⁻⁴
Ro1ACs1-2		7.08	7.72·10 ⁻⁴
Ro1ACs1-3		7.26	7.94·10 ⁻⁴
Ro1ACs2-1	1.16·10 ⁻⁴	64.1	0.0180
Ro1ACs2-2		64.9	0.0187
Ro1ACs2-3		65.6	0.0194
Ro1ACs3-1	1.22·10 ⁻⁶	93.6	0.149
Ro1ACs3-2		94.3	0.169
Ro1ACs3-3		94.2	0.166
Ro1ACs4-1	1.16·10 ⁻⁷	94.2	0.163
Ro1ACs4-2		94.3	0.168
Ro1ACs4-3		98.2	0.560
Ro1BCs1-1	1.11·10 ⁻²	5.27	5.67·10 ⁻⁴
Ro1BCs1-2		3.93	4.14·10 ⁻⁴
Ro1BCs1-3		4.61	4.88·10 ⁻⁴
Ro1BCs2-1	1.16·10 ⁻⁴	59.7	0.0150
Ro1BCs2-2		56.2	0.0130
Ro1BCs2-3		43.5	0.00786
Ro1BCs3-1	1.22·10 ⁻⁶	89.0	0.0818
Ro1BCs3-2		89.7	0.0874
Ro1BCs3-3		84.7	0.0562
Ro1BCs4-1	1.16·10 ⁻⁷	93.2	0.139
Ro1BCs4-2		94.1	0.163
Ro1BCs4-3		97.4	0.391
Ro1CCs1-1	1.11·10 ⁻²	6.20	6.68·10 ⁻⁴
Ro1CCs1-2		7.35	8.01·10 ⁻⁴
Ro1CCs1-3		4.43	4.70·10 ⁻⁴
Ro1CCs2-1	1.16·10 ⁻⁴	89.1	0.0832
Ro1CCs2-2		86.7	0.0661
Ro1CCs2-3		66.3	0.0201
Ro1CCs3-1	1.22·10 ⁻⁶	92.9	0.132
Ro1CCs3-2		91.9	0.114
Ro1CCs3-3		87.2	0.0689
Ro1CCs4-1	1.16·10 ⁻⁷	95.4	0.210
Ro1CCs4-2		94.6	0.179
Ro1CCs4-3		98.6	0.733

Table 3. Sorption of caesium on Kivetty rock samples from CsCl solution under oxic conditions.

Sample	[Cs] _{initial} (mol/l)	Sorption%	R _d (m ³ /kg)
Ki4ACs1-1	1.11·10 ⁻²	10.0	1.10·10 ⁻³
Ki4ACs1-2		9.00	9.64·10 ⁻⁴
Ki4ACs1-3		9.88	1.07·10 ⁻³
Ki4ACs2-1	1.16·10 ⁻⁴	44.5	7.82·10 ⁻³
Ki4ACs2-2		44.0	7.67·10 ⁻³
Ki4ACs2-3		43.7	7.56·10 ⁻³
Ki4ACs3-1	1.22·10 ⁻⁶	89.5	0.0829
Ki4ACs3-2		90.3	0.0921
Ki4ACs3-3		82.6	0.0465
Ki4ACs4-1	1.16·10 ⁻⁷	94.6	0.170
Ki4ACs4-2		94.5	0.167
Ki4ACs4-3		91.3	0.103
Ki4BCs1-1	1.11·10 ⁻²	11.8	1.31·10 ⁻³
Ki4BCs1-2		12.1	1.36·10 ⁻³
Ki4BCs1-3		10.2	1.11·10 ⁻³
Ki4BCs2-1	1.16·10 ⁻⁴	88.5	0.0747
Ki4BCs2-2		67.0	0.0198
Ki4BCs2-3		61.2	0.0154
Ki4BCs3-1	1.22·10 ⁻⁶	92.9	0.128
Ki4BCs3-2		91.8	0.109
Ki4BCs3-3		83.9	0.0509
Ki4BCs4-1	1.16·10 ⁻⁷	96.4	0.262
Ki4BCs4-2		96.4	0.261
Ki4BCs4-3		95.2	0.194

Table 4. Sorption and desorption of protactinium, sorption percentages and R_d values, on Olkiluoto rock samples from OIKR5 groundwater under oxic conditions.

Sample	Undissolved fraction (%)	Sorption S_s (%)	R_d (m ³ /kg)	S_{des} (%)	$R_{d,des}$ (m ³ /kg)
Ol1Pa1-1	0.358 ± 0.052	99.6 ± 0.1%	3.11	98.9 ± 0.2%	0.634
Ol1Pa1-2	0.584 ± 0.078	99.5 ± 0.1%	2.22	99.0 ± 0.2%	0.751
Ol1Pa1-3	0.493 ± 0.069	99.3 ± 0.2%	1.45	99.1 ± 0.2%	0.805
Ol1Pa2-1	0.487 ± 0.074	99.3 ± 0.2%	1.39	98.9 ± 0.2%	0.625
Ol1Pa2-2	0.219 ± 0.035	99.4 ± 0.2%	1.96	99.1 ± 0.2%	0.798
Ol1Pa2-3	0.418 ± 0.059	99.5 ± 0.1%	2.03	98.9 ± 0.2%	0.665
Ol2APa1-1	0.736 ± 0.094	98.9 ± 0.2%	0.944	98.1 ± 0.4%	0.384
Ol2APa1-2	0.412 ± 0.059	99.1 ± 0.2%	1.21	98.4 ± 0.3%	0.434
Ol2APa1-3	0.949 ± 0.118	99.0 ± 0.2%	1.07	98.4 ± 0.3%	0.452
Ol2APa2-1	0.419 ± 0.059	98.7 ± 0.3%	0.794	98.1 ± 0.4%	0.358
Ol2APa2-2	0.754 ± 0.100	98.9 ± 0.3%	0.977	98.0 ± 0.4%	0.355
Ol2APa2-3	0.585 ± 0.082	99.0 ± 0.2%	1.09	98.4 ± 0.3%	0.440
Ol2BPa1-1	0.435 ± 0.061	99.0 ± 0.2%	1.01	98.0 ± 0.4%	0.347
Ol2BPa1-2	0.944 ± 0.116	98.7 ± 0.3%	0.786	97.5 ± 0.5%	0.282
Ol2BPa1-3	0.525 ± 0.071	99.1 ± 0.2%	1.10	98.0 ± 0.4%	0.343
Ol2BPa2-1	1.03 ± 0.14	98.6 ± 0.3%	0.711	98.6 ± 0.3%	0.510
Ol2BPa2-2	0.825 ± 0.107	98.7 ± 0.3%	0.781	98.3 ± 0.3%	0.416
Ol2BPa2-3	0.606 ± 0.087	99.0 ± 0.2%	1.06	98.4 ± 0.3%	0.447

Table 5. Sorption and desorption of protactinium, sorption percentages and R_d values, on Romuvaara rock samples from RoKR5 groundwater under oxic conditions.

Sample	Undissolved fraction (%)	S_s (%)	R_d (m ³ /kg)	S_{des} (%)	$R_{d,des}$ (m ³ /kg)
Ro1APa1-1	0.392 ± 0.115	98.1 ± 0.8%	0.538	97.7 ± 0.7%	0.315
Ro1APa1-2	0.764 ± 0.218	98.6 ± 0.6%	0.740	97.6 ± 0.7%	0.298
Ro1APa1-3	0.603 ± 0.174	98.5 ± 0.7%	0.699	99.6 ± 0.2%	1.88
Ro1APa2-1	0.414 ± 0.122	99.4 ± 0.3%	1.82	97.9 ± 0.6%	0.345
Ro1APa2-2	0.259 ± 0.077	97.7 ± 1.0%	0.433	97.1 ± 0.9%	0.245
Ro1APa2-3	0.469 ± 0.136	98.3 ± 0.7%	0.609	98.4 ± 0.5%	0.444
Ro1BPa1-1	0.588 ± 0.170	97.9 ± 0.9%	0.489	97.2 ± 0.9%	0.254
Ro1BPa1-2	1.36 ± 0.39	97.2 ± 1.2%	0.358	97.2 ± 0.9%	0.249
Ro1BPa1-3	1.14 ± 0.33	96.4 ± 1.5%	0.274	95.4 ± 1.4%	0.149
Ro1BPa2-1	0.696 ± 0.200	94.2 ± 2.5%	0.163	94.3 ± 1.7%	0.119
Ro1BPa2-2	0.456 ± 0.133	95.0 ± 2.2%	0.193	96.1 ± 1.2%	0.179
Ro1BPa2-3	0.615 ± 0.178	96.6 ± 1.5%	0.287	97.6 ± 0.7%	0.295
Ro1CPa1-1	1.17 ± 0.33	98.6 ± 0.6%	0.728	97.7 ± 0.7%	0.317
Ro1CPa1-2	0.933 ± 0.264	98.2 ± 0.8%	0.566	98.5 ± 0.5%	0.505
Ro1CPa1-3	0.941 ± 0.267	98.1 ± 0.8%	0.520	97.8 ± 0.7%	0.325
Ro1CPa2-1	0.789 ± 0.225	97.9 ± 0.9%	0.498	98.0 ± 0.6%	0.359
Ro1CPa2-2	0.936 ± 0.268	95.3 ± 2.0%	0.207	95.2 ± 1.5%	0.145
Ro1CPa2-3	0.966 ± 0.274	97.8 ± 0.9%	0.462	97.2 ± 0.9%	0.258

Table 6. Sorption and desorption of protactinium, sorption percentages and R_d values, on Kivetty rock samples from KiKR4 groundwater under oxic conditions.

Sample	Undissolved fraction (%)	S_s (%)	R_d (m ³ /kg)	S_{des} (%)	$R_{d,des}$ (m ³ /kg)
Ki4APa1-1	0.227 ± 0.047	99.4 ± 0.2%	1.94	99.6 ± 0.1%	1.90
Ki4APa1-2	0.753 ± 0.140	99.5 ± 0.2%	2.35	99.1 ± 0.2%	0.790
Ki4APa1-3	0.727 ± 0.135	99.4 ± 0.2%	1.82	99.7 ± 0.1%	3.15
Ki4APa2-1	0.906 ± 0.169	99.1 ± 0.3%	1.19	99.3 ± 0.2%	1.09
Ki4APa2-2	0.533 ± 0.104	99.2 ± 0.3%	1.31	99.3 ± 0.2%	1.06
Ki4APa2-3	0.638 ± 0.120	98.9 ± 0.3%	0.960	99.0 ± 0.2%	0.755
Ki4BPa1-1	0.430 ± 0.084	99.4 ± 0.2%	1.70	99.3 ± 0.2%	1.07
Ki4BPa1-2	0.460 ± 0.089	98.0 ± 0.6%	0.512	98.2 ± 0.4%	0.395
Ki4BPa1-3	0.513 ± 0.098	99.3 ± 0.2%	1.50	99.3 ± 0.2%	1.07
Ki4BPa2-1	0.324 ± 0.065	99.3 ± 0.2%	1.56	99.5 ± 0.1%	1.60
Ki4BPa2-2	0.421 ± 0.083	99.4 ± 0.2%	1.67	99.3 ± 0.2%	1.09
Ki4BPa2-3	0.491 ± 0.096	99.3 ± 0.2%	1.43	99.3 ± 0.2%	1.03

Table 7. Sorption of protactinium, sorption percentages and R_d values, on Olkiluoto, Romuvaara and Kivetty rock samples from OIKR5 groundwater under anoxic conditions.

Sample	Undissolved fraction (%)	S_s (%)	R_d (m ³ /kg)	pH	Eh(Pt) (mV)
Ol1Pa1-1	14.7 ± 1.46	98.7 ± 0.3%	0.785	8.05	-24
Ol1Pa1-2	7.98 ± 0.81	99.3 ± 0.2%	1.32	7.95	-68
Ol1Pa1-3	13.8 ± 1.4	99.0 ± 0.2%	1.01	8.23	-20
Ol1Pa2-1	7.56 ± 0.79	96.9 ± 0.6%	0.313	8.15	-9
Ol1Pa2-2	4.94 ± 0.58	99.1 ± 0.2%	1.09	8.05	-88
Ol1Pa2-3	6.20 ± 0.63	99.0 ± 0.2%	0.978	8.32	-49
Ol2APa1-1	5.15 ± 0.54	99.1 ± 0.2%	1.18	8.19	-19
Ol2APa1-2	16.0 ± 1.6	98.6 ± 0.3%	0.735	8.55	-24
Ol2APa1-3	3.11 ± 0.33	99.3 ± 0.2%	1.56	8.14	-106
Ol2APa2-1	8.30 ± 0.85	99.5 ± 0.2%	1.99	8.27	-49
Ol2APa2-2	5.63 ± 0.57	99.1 ± 0.2%	1.16	8.43	-46
Ol2APa2-3	4.69 ± 0.52	99.1 ± 0.2%	1.17	8.42	-36
Ol2BPa1-1	6.00 ± 0.66	99.3 ± 0.2%	1.49	8.45	-24
Ol2BPa1-2	8.00 ± 0.80	99.1 ± 0.2%	1.22	8.00	-71
Ol2BPa1-3	15.4 ± 1.5	99.1 ± 0.2%	1.02	8.40	-41
Ol2BPa2-1	6.62 ± 0.68	99.3 ± 0.2%	1.55	8.39	-21
Ol2BPa2-2	6.32 ± 0.65	99.6 ± 0.1%	2.41	8.39	-16
Ol2BPa2-3	5.83 ± 0.50	97.9 ± 0.4%	0.459	8.36	-18
Ro1APa1-7	14.1 ± 1.4	97.1 ± 0.5%	0.350	8.36	-13
Ro1APa1-8	8.88 ± 1.0	98.8 ± 0.3%	0.905	8.07	-117
Ro1APa1-9	19.2 ± 1.7	99.2 ± 0.2%	1.18	8.39	-73
Ro1BPa1-7	14.0 ± 1.5	98.8 ± 0.3%	0.834	8.31	-70
Ro1BPa1-8	19.9 ± 2.0	98.7 ± 0.3%	0.779	8.38	-62
Ro1BPa1-9	13.8 ± 1.4	97.4 ± 0.5%	0.380	8.35	-49
Ro1CPa1-7	17.8 ± 1.8	99.3 ± 0.2%	1.39	8.42	-48
Ro1CPa1-8	7.26 ± 0.75	98.8 ± 0.3%	0.825	8.18	-161
Ro1CPa1-9	9.74 ± 0.98	99.4 ± 0.2%	1.67	8.31	-106
Ki4APa1-7	6.16 ± 0.66	97.6 ± 0.5%	0.414	8.33	-86
Ki4APa1-8	21.9 ± 2.2	99.2 ± 0.2%	1.36	8.41	-89
Ki4APa1-9	16.7 ± 1.7	99.3 ± 0.2%	1.39	8.42	-80
Ki4BPa1-7	21.0 ± 2.2	99.2 ± 0.2%	1.27	8.40	-62
Ki4BPa1-8	13.6 ± 1.4	99.3 ± 0.2%	0.51	8.38	-42
Ki4BPa1-9	13.5 ± 1.4	99.2 ± 0.2%	1.33	8.44	-36

Table 8. Desorption of protactinium, sorption percentages and $R_{d,des}$ values, on Olkiluoto, Romuvaara and Kivetty rock samples from OIKR5 groundwater under anoxic conditions.

Sample	S_{des} (%)	$R_{d,des}$ (m ³ /kg)	pH	Eh(Pt) (mV)
Ol1Pa1-1	99.7 ± 0.2%	4.05	8.20	-148
OL1Pa1-2	99.6 ± 0.1%	2.39	8.01	-171
Ol1Pa1-3	99.5 ± 0.1%	1.99	8.20	-160
Ol1Pa2-1	99.8 ± 0.1%	4.96	8.06	-158
Ol1Pa2-2	99.8 ± 0.1%	6.68	8.00	-169
Ol1Pa2-3	99.1 ± 0.2%	1.24	8.17	-168
Ol2APa1-1	99.2 ± 0.2%	1.44	8.11	-169
Ol2APa1-2	98.9 ± 0.2%	0.963	8.12	-148
Ol2APa1-3	99.1 ± 0.2%	1.29	8.10	-192
Ol2APa2-1	99.4 ± 0.2%	1.82	8.16	-190
Ol2APa2-2	99.3 ± 0.2%	1.46	8.26	-178
Ol2APa2-3	99.6 ± 0.1%	2.65	8.20	-152
Ol2BPa1-1	99.3 ± 0.2%	1.45	8.23	-174
Ol2BPa1-2	94.6 ± 0.8%	0.182	8.21	-187
Ol2BPa1-3	94.9 ± 1.0%	0.189	8.28	-181
Ol2BPa2-1	99.3 ± 0.2%	1.55	8.17	-195
Ol2BPa2-2	99.1 ± 0.2%	1.15	8.35	-189
Ol2BPa2-3	99.5 ± 0.2%	1.98	8.16	-204
Ro1APa1-7	99.2 ± 0.2%	1.39	8.27	-187
Ro1APa1-8	99.4 ± 0.2%	1.70	8.18	-189
Ro1APa1-9	99.1 ± 0.2%	1.16	8.26	-177
Ro1BPa1-7	98.9 ± 0.3%	0.959	8.28	-194
Ro1BPa1-8	99.4 ± 0.2%	1.85	8.30	-179
Ro1BPA1-9	98.9 ± 0.2%	0.969	8.28	-189
Ro1CPa1-7	98.9 ± 0.2%	0.984	8.43	-206
Ro1CPa1-8	99.3 ± 0.2%	1.71	8.30	-221
Ro1CPa1-9	98.9 ± 0.2%	0.973	8.34	-201
Ki4APa1-7	99.2 ± 0.2%	1.35	8.29	-199
Ki4APa1-8	99.1 ± 0.2%	1.21	8.29	-190
Ki4APa1-9	99.1 ± 0.2%	1.13	8.31	-202
Ki4BPa1-7	98.9 ± 0.2%	0.985	8.38	-200
Ki4BPa1-8	99.1 ± 0.2%	1.15	8.29	-203
Ki4BPa1-9	99.1 ± 0.2%	1.16	*	-212

* no pH value, glass electrode broke.

Table 9. Sorption of protactinium, sorption percentages and R_d values, on Romuvaara rock samples from RoKR5 groundwater under anoxic conditions.

Sample	Undissolved fraction (%)	S_s (%)	R_d (m ³ /kg)	pH	Eh(Pt) (mV)
Ro1APa1-1	14.1 ± 0.70	98.7 ± 0.1%	0.731	8.34	54
Ro1APa1-2	7.23 ± 0.31	94.2 ± 0.5%	0.169	8.11	129
Ro1APa1-3	9.22 ± 0.39	87.0 ± 1.0%	0.0731	8.47	79
Ro1APa2-1	2.57 ± 0.13	97.4 ± 0.2%	0.379	8.00	377
Ro1APa2-2	7.88 ± 0.33	95.7 ± 0.4%	0.253	8.82	184
Ro1APa2-3	3.88 ± 0.17	98.3 ± 0.2%	0.593	7.65	364
Ro1BPa1-1	3.43 ± 0.18	88.0 ± 0.2%	0.0795	7.75	-121
Ro1BPa1-2	7.84 ± 0.33	97.6 ± 0.2%	0.404	7.88	194
Ro1BPa1-3	4.83 ± 0.21	99.0 ± 0.1%	1.05	8.63	232
Ro1BPa2-1	8.63 ± 0.39	99.1 ± 0.1%	1.12	7.84	414
Ro1BPa2-2	9.16 ± 0.38	92.5 ± 0.6%	0.138	7.84	314
Ro1BPa2-3	8.50 ± 0.36	87.6 ± 1.0%	0.0793	8.84	214
Ro1CPa1-1	6.99 ± 0.33	99.2 ± 0.1%	1.25	8.22	348
Ro1CPa1-2	4.60 ± 0.20	98.5 ± 0.2%	0.660	8.57	144
Ro1CPa1-3	2.76 ± 0.13	98.7 ± 0.2%	0.801	8.13	192
Ro1CPa2-1	7.85 ± 0.37	96.0 ± 0.3%	0.256	7.89	464
Ro1CPa2-2	2.92 ± 0.13	97.0 ± 0.3%	0.320	8.79	131
Ro1CPa2-3	6.71 ± 0.29	99.6 ± 0.1%	2.33	9.04	181

Table 10. Desorption of protactinium, sorption percentages and $R_{d,des}$ values, on Romuvaara rock samples from RoKR5 groundwater under anoxic conditions.

Sample	S_{des} (%)	$R_{d,des}$ (m ³ /kg)	pH	Eh(Pt) (mV)
Ro1APa1-1	90.0 ± 1.1%	0.100	9.30	49
Ro1APa1-2	87.4 ± 1.4%	0.0766	9.45	69
Ro1APa1-3	81.6 ± 2.2%	0.0472	9.41	60
Ro1APa2-1	92.1 ± 0.9%	0.122	9.11	116
Ro1APa2-2	82.1 ± 2.1%	0.0479	9.49	125
Ro1APa2-3	98.4 ± 0.2%	0.614	9.25	150
Ro1BPa1-1	84.1 ± 1.8%	0.0531	9.22	142
Ro1BPa1-2	79.5 ± 2.5%	0.0405	9.45	210
Ro1BPa1-3	97.5 ± 0.3%	0.413	9.20	117
Ro1BPa2-1	94.1 ± 0.7%	0.162	9.10	144
Ro1BPa2-2	82.8 ± 2.0%	0.0494	9.55	13
Ro1BPa2-3	81.2 ± 2.3%	0.0467	9.57	54
Ro1CPa1-1	96.3 ± 0.4%	0.279	9.66	29
Ro1CPa1-2	98.3 ± 0.2%	0.606	9.23	29
Ro1CPa1-3	95.1 ± 0.6%	0.208	9.45	3
Ro1CPa2-1	81.1 ± 2.3%	0.0427	8.83	327
Ro1CPa2-2	92.6 ± 0.8%	0.133	9.38	306
Ro1CPa2-3	95.2 ± 0.5%	0.215	9.36	288

Table 11. Sorption of protactinium, sorption percentages and R_d values, on Kivetty and Romuvaara rock samples from KiKR4 groundwater under anoxic conditions.

Sample	Undissolved fraction (%)	S_s (%)	R_d (m ³ /kg)	pH	Eh(Pt) (mV)
Ki4APa1-1	8.24 ± 0.37	99.6 ± 0.1%	3.05	8.10	324
Ki4APa1-2	4.59 ± 0.20	99.4 ± 0.1%	1.74	8.61	119
Ki4APa1-3	6.42 ± 0.27	99.7 ± 0.1%	4.15	8.37	99
Ki4BPa2-1	10.9 ± 0.5	99.5 ± 0.1%	2.39	8.04	364
Ki4APa2-2	8.20 ± 0.34	99.5 ± 0.1%	2.37	8.41	124
Ki4APa2-3	8.18 ± 0.34	99.6 ± 0.1%	3.38	8.65	154
Ki4BPa1-1	8.74 ± 0.39	99.6 ± 0.1%	3.04	8.18	311
Ki4BPa1-2	10.8 ± 0.5	99.6 ± 0.1%	2.88	6.96	209
Ki4BPa1-3	10.5 ± 0.5	99.7 ± 0.1%	4.25	8.66	171
Ki4BPa2-1	10.9 ± 0.5	99.7 ± 0.1%	3.67	8.20	324
Ki4BPa2-2	4.29 ± 0.18	99.6 ± 0.1%	3.21	8.62	54
Ki4BPa2-3	12.3 ± 0.6	99.9 ± 0.1%	12.3	8.25	63
Ro1APa1-4	7.60 ± 0.27	99.2 ± 0.1%	1.38	8.45	6
Ro1APa1-5	9.62 ± 0.33	99.2 ± 0.4%	8.61	8.10	-266
Ro1APa2-4	13.5 ± 0.5	100 ± 1%	> 4.0	8.06	-106
Ro1APa2-5	9.74 ± 0.34	99.7 ± 0.1%	4.03	8.45	-46
Ro1BPa1-4	9.95 ± 0.5	99.3 ± 0.1%	1.49	8.47	3
Ro1BPa1-5	12.0 ± 0.5	99.4 ± 0.1%	1.86	8.52	24
Ro1BPa2-4	10.7 ± 0.4	99.6 ± 0.1%	3.03	7.91	-230
Ro1BPa2-5	9.78 ± 0.34	98.4 ± 0.2%	0.695	7.75	300
Ro1CPa1-4	17.4 ± 0.6	98.7 ± 0.1%	0.872	8.38	144
Ro1CPa1-5	16.0 ± 0.6	99.7 ± 0.1%	4.38	8.45	129
Ro1CPa2-4	17.8 ± 0.6	99.8 ± 0.1%	6.74	8.84	49
Ro1CPa2-5	8.61 ± 0.30	99.3 ± 0.1%	1.61	7.92	-36

Table 12. Desorption of protactinium, sorption percentages and $R_{d,des}$ values, on Kivetty and Romuvaara rock samples from KiKR4 groundwater under anoxic conditions.

Sample	S_{des} (%)	$R_{d,des}$ (m ³ /kg)	pH	Eh(Pt) (mV)
Ki4APa1-1	99.9 ± 0.1%	35.1	9.00	302
Ki4APa1-2	98.9 ± 0.3%	0.949	8.97	314
Ki4APa1-3	98.8 ± 0.3%	0.929	9.12	321
Ki4BPa2-1	99.6 ± 0.1%	3.17	9.15	224
Ki4APa2-2	98.3 ± 0.4%	0.597	9.27	238
Ki4APa2-3	98.1 ± 0.4%	0.527	8.72	261
Ki4BPa1-1	98.9 ± 0.2%	0.994	9.21	162
Ki4BPa1-2	98.6 ± 0.3%	0.731	9.30	187
Ki4BPa1-3	99.9 ± 0.1%	34.5	9.11	216
Ki4BPa2-1	99.6 ± 0.1%	2.93	9.35	235
Ki4BPa2-2	98.5 ± 0.3%	0.711	8.96	272
Ki4BPa2-3	99.0 ± 0.2%	1.07	9.05	241
Ro1APa1-4	98.5 ± 0.3%	0.691	9.15	134
Ro1APa1-5	96.0 ± 0.7%	0.254	9.12	159
Ro1APa2-4	99.5 ± 0.1%	2.34	9.13	182
Ro1APa2-5	98.8 ± 0.3%	0.881	9.15	346
Ro1BPa1-4	97.1 ± 0.6%	0.355	9.17	148
Ro1BPa1-5	98.7 ± 0.3%	0.847	9.00	144
Ro1BPa2-4	98.9 ± 0.3%	0.921	9.20	138
Ro1BPa2-5	-	-	9.21	134
Ro1CPa1-4	95.8 ± 0.5%	0.255	8.97	162
Ro1CPa1-5	99.1 ± 0.1%	1.33	8.97	174
Ro1CPa2-4	98.4 ± 0.2%	0.720	9.20	187
Ro1CPa2-5	98.0 ± 0.2%	0.560	9.00	202

Table 13. Sorption of protactinium, sorption percentages and R_d values, on Palmottu rock samples from Palmottu groundwater under anoxic conditions.

Sample	Undissolved fraction (%)	S_s (%)	R_d (m ³ /kg)	pH	Eh(Pt) (mV)
Pa1Pa1-1	4.45 ± 0.51	99.7 ± 0.1%	4.70	8.54	-136
Pa1Pa1-2	15.3 ± 1.8	99.6 ± 0.1%	2.82	8.34	-161
Pa1Pa1-3	1.79 ± 0.21	99.0 ± 0.2%	1.02	8.32	-155
Pa2Pa1-1	4.67 ± 0.54	99.5 ± 0.1%	2.03	8.64	-356
Pa2Pa1-2	6.60 ± 0.76	99.7 ± 0.1%	4.27	8.71	-337
Pa2Pa1-3	8.18 ± 0.94	99.6 ± 0.1%	3.05	8.71	-375

Table 14. Desorption of protactinium, sorption percentages and $R_{d,des}$ values, on Palmottu rock samples from Palmottu groundwater under anoxic conditions.

Sample	S_{des} (%)	$R_{d,des}$ (m ³ /kg)	pH	Eh(Pt) (mV)
Pa1Pa1-1	99.7 ± 0.1%	3.48	8.53	-149
Pa1Pa1-2	99.4 ± 0.2%	1.94	8.73	-163
Pa1Pa1-3	98.9 ± 0.3%	0.942	8.80	-173
Pa2Pa1-1	99.7 ± 0.1%	3.67	8.80	-186
Pa2Pa1-2	99.5 ± 0.2%	2.07	8.90	-194
Pa2Pa1-3	99.6 ± 0.2%	2.92	8.84	-191

Table 15. Sorption of protactinium, sorption percentages and R_d values, on Palmottu rock samples from Palmottu groundwater under oxic conditions.

Sample	Undissolved fraction (%)	S_s (%)	R_d (m ³ /kg)
Pa1Pa1-1	0.200 ± 0.028	99.9 ± 0.1%	33.5
Pa1Pa1-2	0.321 ± 0.043	99.8 ± 0.1%	5.63
Pa1Pa1-3	0.151 ± 0.022	99.9 ± 0.1%	14.3
Pa2Pa1-1	0.101 ± 0.015	99.8 ± 0.1%	5.58
Pa2Pa1-2	0.522 ± 0.067	99.7 ± 0.1%	4.71
Pa2Pa1-3	0.159 ± 0.023	99.9 ± 0.1%	19.9

Table 16. Concentration of Tc-99 in groundwater/crushed rock samples under anoxic conditions.

Groundwater	Rock	Tc-99 undissolved (%)	Tc-99 in water (%)	pH	Eh(Pt) (mV)
RoKR5	no rock	0.50	74.3	8.34	106
		0.38	84.6	9.10	97
		0.32	88.7	7.75	201
		1.05	83.9	8.33	94
KiKR4	no rock	1.55	40.3	7.14	295
		1.04	63.6	7.35	224
	Ro1A	2.56	52.5	8.72	122
		1.85	2.13	9.36	74
	Ki4A	3.21	0.06	8.57	100
		0.88	0.53	8.59	42
OIKR5	no rock	2.63	86.5	8.15	-213
		2.06	87.7	8.13	-218
		5.13	87.6	8.07	-220
	O11	2.31	10.6	8.12	-168
		0.655	11.6	8.26	-123
		1.32	20.4	8.38	-101
	O12A	3.12	0.10	8.19	-175
		0.612	6.10	8.54	-99
		1.04	12.5	8.40	-83
	O12B	2.47	4.37	8.14	-172
		0.947	14.0	8.34	-131
		1.11	5.61	8.43	-165

Table 17. Sorption of protactinium, sorption percentages and R_a values, on Romuvaara rock thin sections under oxic conditions.

from RoKR5 groundwater			from OIKR5 groundwater		
Sample	S_s (%)	$R_a \cdot 10^{-5}$ (m ² /m ³)	Sample	S_s (%)	$R_a \cdot 10^{-5}$ (m ² /m ³)
Ro1A-6	5,92	11,3	Ro1A-9	61,33	269
Ro1A-7	2,70	4,43	Ro1A-10	68,25	365
Ro1A-8	8,84	16,2	Ro1A-11	69,71	403
Ro1B-6	9,15	17,4	Ro1B-9	73,36	426
Ro1B-7	4,39	8,51	Ro1B-10	72,56	463
Ro1B-8	8,69	15,4	Ro1B-11	73,95	192
Ro1C-6	7,89	13,7	Ro1C-9	74,75	470
Ro1C-7	5,43	9,27	Ro1C-10	77,43	571
Ro1C-8	6,49	11,1	Ro1C-11	76,65	547

Table 18. Sorption of protactinium, sorption percentages and R_a values, on Kivetty rock thin sections under oxic conditions.

from KiKR4 groundwater			from OIKR5 groundwater		
Sample	S_s (%)	$R_a \cdot 10^{-5}$ (m ² /m ³)	Sample	S_s (%)	$R_a \cdot 10^{-5}$ (m ² /m ³)
Ki4A-4	3,58	5,91	Ki4A-7	62,12	260
Ki4A-5	4,21	6,06	Ki4A-8	71,24	399
Ki4A-6	5,42	7,66	Ki4A-9	72,45	411
Ki4B-4	3,12	4,45	Ki4B-7	-	-
Ki4B-5	6,47	9,12	Ki4B-8	13,61	23
Ki4B-6	5,60	8,57	Ki4B-9	-	-

Table 19. Sorption of protactinium, sorption percentages and R_a values, on Olkiluoto rock thin sections under oxic conditions.

from OIKR5 groundwater			from KiKR4 groundwater		
Sample	S_s (%)	$R_a \cdot 10^{-5}$ (m ² /m ³)	Sample	S_s (%)	$R_a \cdot 10^{-5}$ (m ² /m ³)
Ol1-6	71,53	386	Ol1-9	25,87	62,6
Ol1-7	66,95	349	Ol1-10	28,02	62,8
Ol1-8	71,06	417	Ol1-11	28,73	58,2
Ol2A-6	74,60	498	Ol2A-9	28,60	65,9
Ol2A-7	76,51	535	Ol2A-10	27,48	60,1
Ol2A-8	73,76	418	Ol2A-11	73,34	296
Ol2B-6	72,89	436	Ol2B-9	67,58	315
Ol2B-7	71,89	473	Ol2B-10	69,61	389
Ol2B-8	74,10	481	Ol2B-11	66,15	321

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