
KBS TEKNISK RAPPORT

120

**Some notes in connection with the
KBS studies of final disposal of spent
fuel**

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Kungl Tekniska Högskolan september 1978

SOME NOTES IN CONNECTION WITH THE KBS
STUDIES OF FINAL DISPOSAL OF SPENT FUEL

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I slutet av rapporten har bifogats en förteckning över av KBS hittills publicerade tekniska rapporter i denna serie.

Some notes in connection with the KBS studies of final disposal of spent fuel

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Abstract

This report contains four short notes on problems briefly studied in connection with the KBS investigations on final disposal of spent unprocessed fuel.

The first note discusses the possibility for thermal convection flow of water in the buffer material. It is concluded that thermal convection will not have a noticeable effect on the transport of water dissolved species.

The second note reports some experimental data on the sorption of strontium and cesium on granite.

The third note discusses the time required to accumulate a critical mass of uranium in the buffer material. It is concluded that it would take millions of years even if very unlikely series of events are postulated.

The fourth note discusses the consequences of hydrogen production in the repository. It is concluded that no grave consequences are expected if escaping hydrogen opens a channel in the clay and in the rock.

Some notes in connection with the KBS project

- 1 A note on thermal convection.771229
- 2 A note on sorption of Sr^{2+} and Cs^+ on granite - some experimental results.780125
- 3 A note on the time to accumulate a critical mass of uranium.780405
- 4 A note on the consequence of hydrogen production in the repository. 780411

Ivars Neretnieks

A NOTE ON THERMAL CONVECTION

- Contents:
- 1 Background
 - 2 Estimation of the magnitude
of the termal convection
 - 3 Conclusions

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1 Background

The buried capsules evolve heat. This heat will induce thermal convection due to the resulting thermal gradient. It is of some interest to determine if the thermal convection will induce a velocity in the clay barrier which will transport the nuclides and other components faster than other mechanisms.

2 Estimation of the magnitude of the thermal convection

Häggbloom (1) investigated this problem. The thermally induced hydraulic gradient i_{th} may be determined from

$$i_{th} = \frac{\alpha \Delta \theta}{k_1(k_2+1)} \quad 1$$

where α is the thermal expansion coefficient for water. α is between $2 \cdot 10^{-4}$ and $10 \cdot 10^{-4}$ for water in the temperature range $20^\circ\text{C} - 140^\circ\text{C}$. $\Delta\theta$ is the temperature difference over the clay barrier. k_1 is a constant between 1 and 2 depending on the geometry of the induced water movement. k_2 is another constant dependent on the geometry. k_2 can vary from 0 to very large values. Häggbloom used $k_1 = 2$ and $k_2 = 2$. Using these values and $\Delta\theta = 20^\circ\text{C}$ and the temperature at the capsule wall = 80°C we get $\alpha = 5.8 \cdot 10^{-4}$ and $i_{th} = 1.5 \cdot 10^{-3}$ m/m. This is of the same magnitude as the figure often used for the hydraulic gradient.

With this thermally induced hydraulic gradient, a permeability of $K_p = 10^{-10}$ m/s and with a porosity $\epsilon = 0.25$ for the compacted clay it takes 20 000 years for the water to travel a distance equal to the thickness of the clay barrier ($z = 0.38$ m).

This figure should be compared with the time it takes for a species to diffuse the same distance. This may be computed from

$$\frac{Dt}{z^2} = \text{Const} \quad 2$$

for the instationary case, z is the barrier thickness, D the diffusivity of the species in the compacted clay. For $\text{Const} = 0.1$ the concentration on the other side of the barrier has increased to $0.05 c_0$. c_0 is the sudden concentration difference at time $t = 0$. For $\text{Const} = 1$ the concentration has increased to $0.95 c_0$.

It takes 13 years to reach $0.05 c_0$ and 130 years to reach $0.95 c_0$ for methane. D for methane has been measured in our laboratories. $D = 3.9 \cdot 10^{-11} \text{ m}^2/\text{s}$. Most other species will diffuse slower.

3 Conclusions

Although many of the data used are approximate, it may safely be concluded that thermal convection will not have a serious or even noticeable effect on the transport of water solved species to and from the capsule.

References:

- 1 Häggblom, H. Calculation of nuclide migration in rock and porous media, penetrated by water. KBS teknisk rapport nr 52, 1977-09-14.

KEMISK APPARATTEKNIK

KTH

A note on sorption of Sr^{2+} and Cs^+ on granite - some experimental results.

Contents:	1	Summary
	2	Background
	3	Experimental
	4	Results
	5	Discussion and conclusions
	6	Literature

1978-01-25

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1 Summary

Sorption experiments were made to determine the equilibrium of Cs^+ and Sr^{2+} in water solutions with granite.

Particle sizes between 0.044 and 1 mm were used. The results indicate that the whole volume of the particles is utilized - not only the outer surface. The K_d -values were

$$\begin{aligned} \text{Sr}^{2+} &: K_d = 10 \text{ g/g} \\ \text{Cs}^+ &: K_a = 15 \text{ g/g} \end{aligned}$$

at 25°C in a synthetic groundwater.

The results are in good agreement with previous investigations.

The results should however not be used for larger particles than 1 mm without further investigations.

2 Background

It is important to have good equilibrium data on ion exchange and adsorption reactions for radionuclides on rock, as ion-exchange and adsorption reactions will retard escaping nuclides from a final repository. It is also important to know if it is a surface reaction or a reaction in depth.

If only the surface of the fissures reacts, there is a limited amount of rock surface which may be utilized. If it is a reaction in depth there is a considerable amount of rock which may be utilized.

In the latter case, however, the time for the ion to diffuse into the rock proper may be considerable.

The literature does not give a clear picture of these phenomena. See 4.2.2 in reference (2).

3 Experimental

Granite was crushed and sieved to various fractions ranging in sizes from less than 0.044 to 1 mm in diameter.

Between 5 and 20 g of granite were contacted in a stirred vessel with 100, 200 and 400 ml of water.

The water had the following composition

Na	288	mg/l	Cl	500	mg/l
K	10	"-	F	1.5	"-
Mg	15	"-	SO ₄	15	"-
Ca	75	"-	CO ₃	200	"-

Cs⁺ and Sr²⁺ concentrations were near 5 mg/l. The temperature in all experiments was 25°C.

The contact time was up to 15 days. Frequent sampling was made during the first day. Thereafter the sampling was done every 2 to 3 days.

4 Results

During the first hour concentration fell rapidly. Thereafter the concentration fell vary slowly. Evaluated K_d -values increased with time, indicating a slow diffusion into the particle. The time used (7 - 14 days) was probably not sufficient to reach a real equilibrium even for the rather small particles $d_p = 0.1 - 0.12$ mm. Table 1 and 2 give the experimental conditions for the runs.

In diagram 1 K_d and K_a values are plotted versus the particle diameter for Sr^{2+} and Cs^+ equilibria. There is a considerable scatter in the results. The results indicate that the equilibrium cannot be described by a surface equilibrium constant

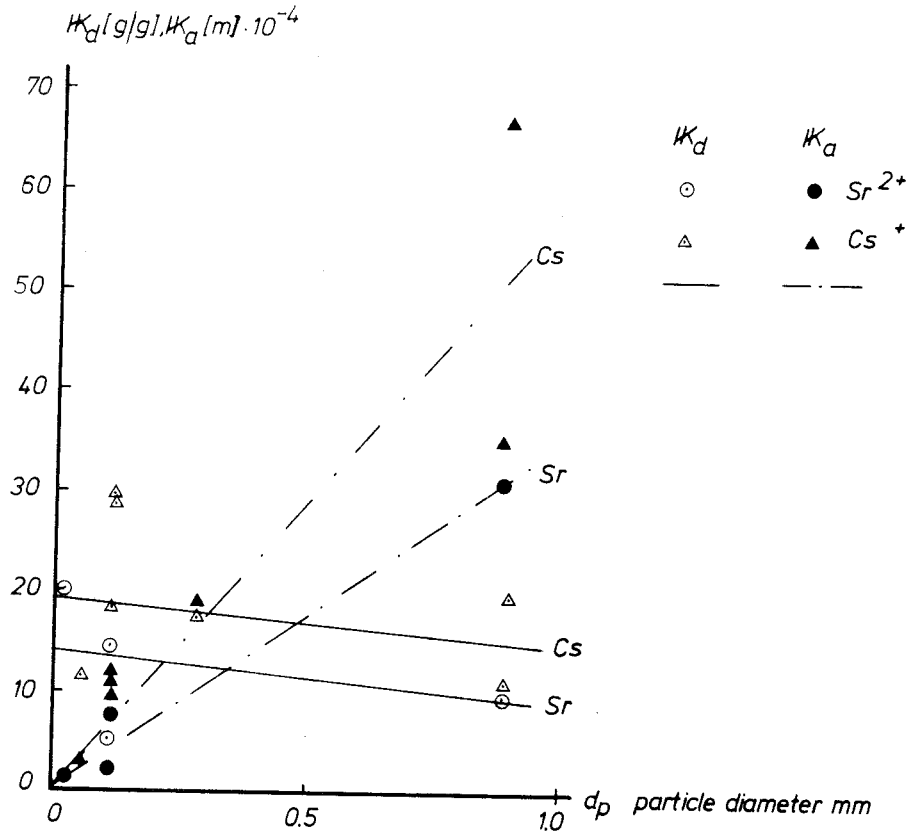
$$K_a = \frac{g/m^2 \text{ solid}}{g/m^3 \text{ liquid}}$$

as K_a is so strongly dependent on the particle size.

There is however some indication that the mass equilibrium constant K_d decreases somewhat with increasing particle size. This would mean that the interior parts of the particles are not as active as the outer or that they are less accessible and that equilibrium was not reached.

These results compare well with those of Allard (1).

Diagram 1



5 Conclusions

The ion exchange equilibrium of Cs and Sr with granite is better described by a mass equilibrium constant than a surface equilibrium constant.

Due to the rather large scatter in the results and the very small particles used this result should not be used to extrapolate to large particles such as the rock itself.

A further investigation must be made on larger particles.

6 Literature

- 1 Allard, B., Kipatsi, H., Rydberg, J.,
Sorption av långlivade radionuklider i lera och berg.
KBS teknisk rapport 55 1977-10-10

- 2 Neretnieks, I., Retardation of escaping nuclides from
a final depository.
KBS teknisk rapport 30 1977-09-14

Table 1. Experimental conditions and results for Sr^{2+}

Experiment	Initial concentration C_o (ppm)	Final concentration C_s (ppm)	Particle diameter d_p (mm)	Mass of granite L (g)	Volume of liquid V (ml)	Temperature (°C)	Mass equilibrium constant K_d (g/g)	Surface equilibrium constant K_{am}
1	5.025	3.5	0.0 - 0.044	5.76	200	25	20	$1.7 \cdot 10^{-4}$
2	5.025	3.9	0.105 - 0.149	5.76	200	25	14.6	$7.3 \cdot 10^{-4}$
5	4.66	4.2	0.1 - 0.120	11.40	400	25	4.7	$2.0 \cdot 10^{-4}$
6	4.66	3.78	0.75 - 1.00	11.40	400	25	9.4	$31.5 \cdot 10^{-4}$

Table 2. Experimental conditions and results for Cs⁺

Experiment	Initial concentration C _o (ppm)	Final concentration C _s (ppm)	Particle diameter d _p (mm)	Mass of granite L (g)	Volume of liquid V (ml)	Temperature (°C)	Mass equilibrium constant K _d (g/g)	Surface equilibrium constant K _a m
3	5.11	4.17	0.105-0.149	5.74	400	25	18	9.10 ⁻⁴
4	5.11	4.44	0.044-0.063	5.79	400	25	13	2.7.10 ⁻⁴
7	5.14	3.28	0.1 -0.12	11.40	400	25	28.2	11.9.10 ⁻⁴
8	5.14	3.28	0.75 -1.00	11.40	400	25	10.5	35.2.10 ⁻⁴
9	10.91	1.553	0.1 -0.120	20.93	100	25	29	12.8.10 ⁻⁴
10	10.91	2.193	0.75 -1.00	20.93	100	25	20.1	67.4.10 ⁻⁴
11	10.91	2.45	0.25 -0.3	20.93	100	25	17.3	18.2.10 ⁻⁴

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A note on the time to accumulate a critical mass of
uranium

19780405

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Time to accumulate a critical mass of uranium

The minimum critical mass is some 4-7 tons if it is arranged in a sphere with a radius of 1 m. Other geometries are also possible but then considerably more uranium is needed. If the uranium from all capsules is distributed evenly over the tunnel no criticality is achieved. The uranium from at least 3 capsules is needed to get one critical mass. There are various ways to achieve this mass. Three capsules in a row may give up their uranium content simultaneously, in which case the transport distances are short, or many capsules (there are 160 in a tunnel at 6 m intervals) contribute to achieve the necessary mass. In the latter case the transport distances are very large for the capsules furthest away from the point where the mass is accumulating.

The transport velocity for uranium from one capsule is determined by a series of resistances. The first (after the penetration of the copper) is the gradual dissolution and diffusion of uranium in the penetrated capsule. The second is the transport in the clay in the hole up to the tunnel bottom and the third is the transport in the clay/sand mixture in the tunnel.

The first and second mechanisms have been treated in (1,2). If the capsule is chopped off at the top and the transport resistance in the tunnel is neglected, the time to transport a given amount of uranium from the capsule to the tunnel bottom may be computed. The results are given in table 1 for oxidizing conditions when the uranium oxide solubility is high.

It will take 35 million years for one capsule to give up its entire uranium content by the very improbable mechanisms assumed here. That will also be the minimum time for three capsules

to simultaneously contribute to the necessary minimum critical mass. The other possibility when many capsules contribute may be somewhat faster, provided the transport resistance in the tunnel may be neglected and the copper in many of the capsules is seriously degraded at about the same time. A little hole does not suffice. The top portion of the copper capsule must be degraded. If this happens to some 30 capsules at about the same time it will take about 2 million years for the necessary amount (4-7 tons) of uranium to escape. The time for it to gather again is not included in this figure.

The mechanisms necessary for the reconcentration of the uranium to a critical configuration are known in principle. This may be achieved by a solution of the uraniumoxide in oxidizing water and precipitation of the solved species at some spot where the uranium (VI) is again reduced to uranium (IV).

The series of events leading to this are not easy to imagine. The copper capsules must be not only penetrated but seriously degraded to achieve a hole large enough for the transport of the solved species. The uranium must be oxidized to U(VI) by some mechanism. It should be pointed out that the water at these depths in the rock is reducing. The water must contain large amounts of carbonates or other complexing agents to achieve a high solubility of the uranium. A small spot must be available where all the uranium can be again reduced to uranium (IV).

If all these events occur it will take many millions of years to reach critical mass. Furthermore the gathering will be very slow which eliminates the possibility of a quick blow up.

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Utdiffusion av svårlösliga nuklider ur kapsel efter
kapselgenombrott. KBS TR 80 1978-03-07.
- 2) Neretnieks, I., Transport of oxidants and radionuclides
through a clay barrier. KBS TR 79 1978-02-20

Table 1

Time for a given amount of uranium in one capsule to dissolve and diffuse up to the tunnel bottom.

Length of UO ₂ matrix dissolved m	Amount of U dissolved tons	Time after copper penetration Myers
0.1	0.035	0.36
0.2	0.07	0.75
0.5	0.175	2.1
1	0.35	4.8
2	0.7	12.3
4	1.4	35.3

Diffusivity in clay $2 \cdot 10^{-11} \text{ m}^2/\text{s}$ (2), in UO₂ matrix $10^{-10} \text{ m}^2/\text{s}$
solubility in water $1073 \text{ gU}/\text{m}^3$, compacted clay above the capsule 2 m.

A note on the consequence of hydrogen production in the repository

Background

Hydrogen may be produced in the copper capsule by zircalloy-corrosion and by radiolysis. Hydrogen diffuses slowly in compacted bentonite (1). If the production of hydrogen is fast it will not have time to escape by diffusion at the same rate at which it is produced. A gas bubble may form.

Treatment of the problem

Pusch (2) has treated this case. He concludes that the 4.5 m³ of hydrogen at 50 bar pressure will be confined in a stable bubble, the space being created by the displacement of the tunnel fill.

The question has been posed of what would be the consequences if the bubble escapes. For this to happen a channel must be formed in the compacted clay and in the rock. A local expansion of the rock might be conceived if the pressure of the hydrogen rises to 130-140 bar or more. This is the pressure of the overlying rock. A widening of the fissures in the nearby rock will facilitate the extrusion of bentonite in the fissures. This is further facilitated by the increased pressure difference. If a channel is formed, some clay will probably escape. As the clay will keep its swelling properties even at very low clay densities (2), a minor loss of clay does not leave a void. The permeability decreases but will still be much less than that of the rock (10^{-9} m/s) even after a major loss (> 80 %) of clay (2).

After the escape of the bubble the locally expanded rock will settle down again. The fissures that were opened will probably not settle to their original width. This is partly because other points on the surfaces will touch after resettling and partly because clay has crept out into the fissures. Any widened fissure which is blown free of clay will fill very quickly if the fissure width is larger than about 0.2 mm. Pusch (3) has investigated the extrusion velocity of bentonite in fissures experimentally, as well as theoretically. In two months the clay creeps about 6 mm in a 0.5 mm fissures. Larger fissures will fill faster and smaller are of no importance as the water flow and its transport capacity of radionuclides is low in such fissures (4). A 6 mm deep layer of clay in a fissure has about the same resistance to diffusion as the clay surrounding the capsule in the hole. This is mainly because the fissure area is very small in comparison to the area of the wall in the hole with the capsule.

The clay will continue to extrude in the fissure but at a continually decreasing rate. The resistance to diffusion will thus continually increase.

If the fissure(s) become very large 1-3 mm, the clay will fill these fissures much quicker and the resistance to diffusion will increase proportionally.

In the part of the fissure which has not yet been filled with clay the retention time of the water will increase compared to the undisturbed rock. There is at present no method to describe in detail the fissures in the disturbed rock. The following example will however give an indication of the consequences to be expected if all fissures increase from 0.1 mm to 0.5 mm after the escape of the gas. Originally we have the following

relative resistances for escaping nuclides (from table 5 ref 4).

	fissure width mm	relative resistances		"film" resistance
		clay in hole	clay in fissure	
original case	0.1	1	0	7.1
after H ₂ escape	0.5	0	1 (6 mm clay)	3.2

After the event, clay has extruded 6 mm into the fissure and restored the resistance which may have been lost in the hole. The film resistance has decreased to less than half. In total the resistance has decreased to half the original.

Conclusion

No grave consequences are expected if the escaping hydrogen opens a channel in the clay and in the rock. This is because:

- If a limited amount of clay is lost the clay still retains strong swelling properties and will quickly fill the channel in the clay mass.
- A large (> 0.5 mm) fissure will quickly fill with clay and considerably decrease the escape of diffusing species.
- The escape of a limited amount of clay from the hole does not increase the permeability of the clay above that of the rock.
- The "film resistance" will decrease with increasing fissure widths. Even very large fissures (3 mm) will not decrease the total resistance to mass transfer to more than half the original.

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FÖRTECKNING ÖVER KBS TEKNISKA RAPPORTER

- 01 Källstyrkor i utbränt bränsle och högaktivt avfall från en PWR beräknade med ORIGEN
Nils Kjellbert
AB Atomenergi 77-04-05
- 02 PM angående värmeledningstal hos jordmaterial
Sven Knutsson
Roland Pusch
Högskolan i Luleå 77-04-15
- 03 Deponering av högaktivt avfall i borrhål med buffertsubstans
Arvid Jacobsson
Roland Pusch
Högskolan i Luleå 77-05-27
- 04 Deponering av högaktivt avfall i tunnlar med buffertsubstans
Arvid Jacobsson
Roland Pusch
Högskolan i Luleå 77-06-01
- 05 Orienterande temperaturberäkningar för slutförvaring i berg av radioaktivt avfall, Rapport 1
Roland Blomqvist
AB Atomenergi 77-03-17
- 06 Groundwater movements around a repository, Phase I, State of the art and detailed study plan
Ulf Lindblom
Hagconsult AB 77-02-28
- 07 Resteffekt studier för KBS
Del 1 Litteraturgenomgång
Del 2 Beräkningar
Kim Ekberg
Nils Kjellbert
Göran Olsson
AB Atomenergi 77-04-19
- 08 Utlakning av franskt, engelskt och kanadensiskt glas med högaktivt avfall
Göran Blomqvist
AB Atomenergi 77-05-20

- 09 Diffusion of soluble materials in a fluid filling a porous medium
Hans Häggblom
AB Atomenergi 77-03-24
- 10 Translation and development of the BNWL-Geosphere Model
Bertil Grundfelt
Kemakta Konsult AB 77-02-05
- 11 Utredning rörande titans lämplighet som korrosionshärdig kapsling för kärnbränsleavfall
Sture Henriksson
AB Atomenergi 77-04-18
- 12 Bedömning av egenskaper och funktion hos betong i samband med slutlig förvaring av kärnbränsleavfall i berg
Sven G Bergström
Göran Fagerlund
Lars Rombén
Cement- och Betonginstitutet 77-06-22
- 13 Urlakning av använt kärnbränsle (bestrålad uranoxid) vid direktdeponering
Ragnar Gelin
AB Atomenergi 77-06-08
- 14 Influence of cementation on the deformation properties of bentonite/quartz buffer substance
Roland Pusch
Högskolan i Luleå 77-06-20
- 15 Orienterande temperaturberäkningar för slutförvaring i berg av radioaktivt avfall
Rapport 2
Roland Blomquist
AB Atomenergi 77-05-17
- 16 Översikt av utländska riskanalyser samt planer och projekt rörande slutförvaring
Åke Hultgren
AB Atomenergi augusti 1977
- 17 The gravity field in Fennoscandia and postglacial crustal movements
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- 18 Rörelser och instabilitet i den svenska berggrunden
Nils-Axel Mörner
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- 19 Studier av neotektonisk aktivitet i mellersta och norra Sverige, flygbildsgenomgång och geofysisk tolkning av recenta förkastningar
Robert Lagerbäck
Herbert Henkel
Sveriges Geologiska Undersökning september 1977

- 20 Tektonisk analys av södra Sverige, Vättern - Norra Skåne
Kennert Röshoff
Erik Lagerlund
Lunds Universitet och Högskolan Luleå september 1977
- 21 Earthquakes of Sweden 1891 - 1957, 1963 - 1972
Ota Kulhánek
Rutger Wahlström
Uppsala Universitet september 1977
- 22 The influence of rock movement on the stress/strain
situation in tunnels or bore holes with radioactive con-
sistors embedded in a bentonite/quartz buffer mass
Roland Pusch
Högskolan i Luleå 1977-08-22
- 23 Water uptake in a bentonite buffer mass
A model study
Roland Pusch
Högskolan i Luleå 1977-08-22
- 24 Beräkning av utlakning av vissa fissionsprodukter och akti-
nider från en cylinder av fränskt glas
Göran Blomqvist
AB Atomenergi 1977-07-27
- 25 Blekinge kustgnejs, Geologi och hydrogeologi
Ingemar Larsson KTH
Tom Lundgren SGI
Ulf Wiklander SGU
Stockholm, augusti 1977
- 26 Bedömning av risken för fördröjt brott i titan
Kjell Pettersson
AB Atomenergi 1977-08-25
- 27 A short review of the formation, stability and cementing
properties of natural zeolites
Arvid Jacobsson
Högskolan i Luleå 1977-10-03
- 28 Värmeledningsförsök på buffertsubstans av bentonit/pitesilt
Sven Knutsson
Högskolan i Luleå 1977-09-20
- 29 Deformationer i sprickigt berg
Ove Stephansson
Högskolan i Luleå 1977-09-28
- 30 Retardation of escaping nuclides from a final depository
Ivars Neretnieks
Kungliga Tekniska Högskolan Stockholm 1977-09-14
- 31 Bedömning av korrosionsbeständigheten hos material avsedda
för kapsling av kärnbränsleavfall. Lägesrapport 1977-09-27
samt kompletterande yttranden.
Korrosionsinstitutet och dess referensgrupp

- 32 Egenskaper hos bentonitbaserat buffertmaterial
Arvid Jacobsson
Roland Pusch
Högskolan i Luleå 1978-06-10
- 33 Required physical and mechanical properties of buffer masses
Roland Pusch
Högskolan i Luleå 1977-10-19
- 34 Tillverkning av bly-titan kapsel
Folke Sandelin AB
VBB
ASEA-Kabel
Institutet för metallforskning
Stockholm november 1977
- 35 Project for the handling and storage of vitrified high-level waste
Saint Gobain Techniques Nouvelles October, 1977
- 36 Sammansättning av grundvatten på större djup i granitisk berggrund
Jan Rennerfelt
Orrje & Co, Stockholm 1977-11-07
- 37 Hantering av buffertmaterial av bentonit och kvarts
Hans Fagerström, VBB
Björn Lundahl, Stabilator
Stockholm oktober 1977
- 38 Utformning av bergrumsanläggningar
Alf Engelbrektson, VBB
Arne Finné, KBS
Stockholm december 1977
- 39 Konstruktionsstudier, direktdeponering
ASEA-ATOM
Västerås
- 40 Ekologisk transport och stråldoser från grundvattenburna radioaktiva ämnen
Ronny Bergman
Ulla Bergström
Sverker Evans
AB Atomenergi 1977-12-20
- 41 Säkerhet och strålskydd inom kärnkraftområdet.
Lagar, normer och bedömningsgrunder
Christina Gyllander
Siegfried F Johnson
Stig Rolandson
AB Atomenergi och ASEA-ATOM 1977-10-13

- 42 Säkerhet vid hantering, lagring och transport av använt kärnbränsle och förglasat högaktivt avfall
Ann-Margret Ericsson
Kemakta november 1977
- 43 Transport av radioaktiva ämnen med grundvatten från ett bergförvar
Bertil Grundfelt
Kemakta november 1977
- 44 Beständighet hos borsilikatglas
Tibor Lakatos
Glasteknisk Utveckling AB
- 45 Beräkning av temperaturer i ett envånings slutförvar i berg för förglasat radioaktivt avfall Rapport 3
Roland Blomquist
AB Atomenergi 1977-10-19
- 46 Temperaturberäkningar för slutförvar för använt bränsle
Taivo Tarandi
Vattenbyggnadsbyrån Stockholm 1978
- 47 Teoretiska studier av grundvattenrörelser
John Stokes
Roger Thunvik
Inst för kulturteknik KTH maj 1978
- 48 The mechanical properties of the rocks in Stripa, Kråkemåla, Finnsjön and Blekinge
Graham Swan
Högskolan i Luleå 1977-09-14
- 49 Bergspänningsmätningar i Stripa gruva
Hans Carlsson
Högskolan i Luleå 1977-08-29
- 50 Lakningsförsök med högaktivt franskt glas i Studsvik
Göran Blomqvist
AB Atomenergi november 1977
- 51 Seismotectonic risk modelling for nuclear waste disposal in the Swedish bedrock
F Ringdal
H Gjöystdal
E S Husebye
Royal Norwegian Council for scientific and industrial research
- 52 Calculations of nuclide migration in rock and porous media, penetrated by water
H Häggblom
AB Atomenergi 1977-09-14
- 53 Mätning av dissusionshastighet för silver i lera-sandblandning
Bert Allard
Heino Kipatsi
Chalmers tekniska högskola 1977-10-15

- 54 Groundwater movements around a repository
- 54:01 Geological and geotechnical conditions
Håkan Stille
Anthony Burgess
Ulf E Lindblom
Hagconsult AB september 1977
- 54:02 Thermal analyses
Part 1 Conduction heat transfer
Part 2 Advective heat transfer
Joe L Ratigan
Hagconsult AB september 1977
- 54:03 Regional groundwater flow analyses
Part 1 Initial conditions
Part 2 Long term residual conditions
Anthony Burgess
Hagconsult AB oktober 1977
- 54:04 Rock mechanics analyses
Joe L Ratigan
Hagconsult AB september 1977
- 54:05 Repository domain groundwater flow analyses
Part 1 Permeability perturbations
Part 2 Inflow to repository
Part 3 Thermally induced flow
Joe L Ratigan
Anthony S Burgess
Edward L Skiba
Robin Charlwood
- 54:06 Final report
Ulf Lindblom et al
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