

The Palmottu Natural Analogue Project

Technical Report 98-01

Transport of Radionuclides in a Natural Flow System at Palmottu

2nd 12-monthly Progress Report 1997-01-01—1998-01-31

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I INTRODUCTION

The Palmottu Project “Transport of radionuclides in a natural flow system at Palmottu” commenced on January 1st, 1996, as a research study agreed by the European Commission and the four contractors below:

- Geological Survey of Finland (GTK, coordinator),
- Svensk Kärnbränslehantering AB (SKB),
- Empresa Nacional de Residuos Radioactivos S.A. (ENRESA),
- Bureau de Recherches Géologiques et Minières (BRGM),

under the Nuclear Fission Safety research and training programme (Contract No FI4W-CT95-0010, DG 12-WSME).

The Contractors have agreed on associated contracts with the following five partners:

- Radiation and Nuclear Safety Authority, Finland (STUK),
- University of Helsinki, Laboratory of Radiochemistry, Finland (UHRAD),
- Technical Research Center of Finland, Nuclear Energy, Finland (VTT Energy),
- Technical Research Center of Finland, Communities and Infrastructure, Finland (VTT-CI);
- QuantiSci SL, Spain (QuantiSci),

and with the following subcontractors: Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas, Instituto de Medioambiente, (Ciemat), Universitat Politècnica de Catalunya (UPC) and Universidad de Oviedo (UO), all three from Spain, and Conterra AB, Geokema AB, Geosigma AB, Göteborg University, Intera KB (Intera), Kemakta Konsult AB (Kemakta) and ÅF-IPK AB Miljö, all seven from Sweden. Kivikonsultit Oy, Posiva Oy, PRG-Tec Oy, Suomen Malmi Oy and Technical University of Helsinki, have acted as the main subcontractors from Finland.

The project was originally subdivided into two Phases. Phase I studies which were focused on the hydrogeological assessment of the site, were commenced on January 1st 1996 and completed by May 31st, 1997. The following workpackages (WP) and major tasks (with WP and Task Leaders) were covered:

- WP 1 : Understanding the natural flow system (GTK)
 - Task 1.1: Up-dating the structural model of the site (GTK)
 - Task 1.2: Hydraulic testing and interpretation (Geosigma)
 - Task 1.3: Hydrochemistry as an indicator of groundwater flow (Conterra)
 - Task 1.4: Flow modelling and integrated evaluation of hydraulic and hydrogeochemical results (VTT-CI)
 - Task 1.5: Low-contamination drilling (GTK + Conterra)
- WP 5: Performance assessment exercise of Phase I (Kemakta)

The commencement of the Phase II was dependent on the successful completion of Phase I. Therefore, in order to offer the appropriate bases, a report on the hydrogeological evaluation of the Palmottu natural analogue study site was submitted to the European Commission in April 1997 and will be published under EUR (18202) (Blomqvist *et al.* 1998). Based on an project evaluation, the European Commission supported to continue

the Palmottu project for an additional 25 months with Phase II studies, with a starting from August 1st, 1997.

Phase II comprises the following major workpackages:

- WP 2: Geochemical evolution of the water-rock system (GTK)
- WP 3: Redox processes (QuantiSci)
- WP 4: Migration of radionuclides (VTT Energy)
- WP 5: Conclusions relevant to repository performance assessment (Kemakta/SKB)

In the following, the progress of the project activities for 1997 is described separately for Phase I and Phase II studies. With respect to Phase I studies, a conclusive summary of the main results will first be given, with emphasis on the conceptual hydrogeological model of the site, followed by a detailed description of the progress taken within the individual tasks. With respect to phase II studies, the plans and initial progress within each of the four work packages is described for the period 1997-08-01–1998-01-31.

The progress taken has been in good accordance with the plans of the project described in the Technical Annex (see Gantt chart, next page). Eventual modifications to the programme are discussed at the end of each task description. Due to the delay in the starting of Phase II, the original flow chart of the project has slightly been modified for Phase II (with starting from August 1st 1997). The manpower usage of the project is in line with the plans (presented in a separate Annex).

	1996	1997	1998	1999
PHASE 1				
WP1: Understanding the natural flow system				
WP5: Performance assessment exercise of Phase I				
PHASE 2				
WP2: Geochemical evolution of the water rock system				
WP3: Redox processes				
WP4: Migration of radionuclides				
WP5: Conclusions relevant to repository PA				
Meetings and Milestones	1	2	3a-----3d	4
Deliverables: 6-month progress Report (6-M) 12-month Progress Report + Cost Statement (12-M) Annual Programme Report (AR) Hydrogeological Assessment Report (HAR) Final Report (FR)	6-M	12-M AR 6-M HAR	12-M AR 6-M	12-M AR FR
		1. Initial Meeting, 2. Hydrogeological Assessment, 3. Modeling Workshop, 4. Mid-Term Assessment, 5. Final Meeting		

II PHASE I STUDIES CARRIED OUT IN 1997-01-01–1997-05-30

WP 1: Understanding the natural flow system: Summary of results

Objectives and main activities

The objective of WP 1 was to assess the natural flow system of the site in order to be able to proceed with the transport analogue studies of phase II. Therefore WP 1 comprised the following five major tasks: (1.1) Up-dating the structural model of the site, (1.2) Hydraulic testing and interpretation, (1.3) Hydrochemistry as an indicator of groundwater flow, (1.4) Flow modelling and integrated evaluation of hydraulic and hydrogeochemical results, and (1.5) Low-contamination drilling.

The previous activities carried within this workpackage has been documented in the 6- and 12-monthly Progress Reports (Blomqvist *et al.*, 1996, 1997 a, b). The findings of this final study period of Phase I will be described here below, first as a summary of main conclusions of the integrated study of WP I, followed by the task-specific chapters. All Phase I results will be summed-up in the Hydrogeological Assessment Report to be published by the European Commission (Report: EUR 18202, Blomqvist *et al.*, 1998).

Methodology

Both regional and local hydrostructural models of the Palmottu area around the U-Th mineralization were developed. The local site-scale model is the result of exhaustive interdisciplinary studies involving surface and downhole investigations of geology, geophysics, hydrogeology and hydrochemistry. A structural model was produced first which was progressively modified and refined on the basis of hydraulic testing and hydrogeochemical considerations during the course of the investigations. Flow modelling at regional and local scales was also undertaken. According to local scale flow modelling, the Palmottu brook fracture zone acts as the main outlet pathway from the site. Groundwater flow occurs mainly within the uppermost 100 m section of the bedrock, where a predominantly subhorizontal flow can be inferred.

Based on systematic site-scale hydraulic testing from packed-off borehole sections, four major hydrostructural units were identified; subhorizontal structure H1 and three subvertical structural units including the Western granite, the Eastern granite and the Palmottu brook structure. The cross-hole tests indicated that the hydraulic properties of these units are similar and of the same order of magnitude as those for the adjacent fractured rock in the upper part of the bedrock. The test responses showed a local hydraulic anisotropy of the drawdown field along the subvertical structures, mainly parallel to the general direction of schistosity in the bedrock. Over longer periods of time the test responses averaged out and the bedrock behaved as an interconnected network of discrete fractures, approaching the equivalent of a fractured, porous medium with local anisotropy associated with the structures. However, the Eastern granite seems to be hydraulically less connected to the central area, forming a hydrogeological unit of its own.

Since the hydrogeochemical data were obtained from the same borehole intersections where the structural features had been hydraulically tested, the results provided a unique opportunity to integrate hydrogeochemistry with hydrogeology in order to test and assess the groundwater flow conditions at Palmottu. Based on hydraulic data, major ion chemistry and environmental isotopes, the following flow systems were identified (Figure 1): 1. Upper flow system, which is divided into three subsystems, based on topographic features

and head values: Southwestern flow system, Northwestern flow system, and Eastern flow system, 2. Dynamic deep flow system, and 3. Stagnant flow system.

Main conclusions: The conceptual hydrogeological model of Palmottu

The Upper flow system is situated within the subhorizontal structural unit H1, forming the uppermost 100 m layer of bedrock within the central part of the site. Here the bedrock is relatively conductive and the groundwater is of bicarbonate type, generally with a residence time of less than 40 years.

Deeper within the bedrock, below unit H1, along the southern part of the Palmottu brook valley, the bedrock is also relatively conductive, controlling the position of the Dynamic deep flow system. This is characterized by flow conditions towards the southeast, with the subvertical structural units of the Western granite and the Palmottu brook valley being regarded as the most important features controlling flow. The groundwater is of bicarbonate type, similar to that in the Upper flow system, and the distribution of head values as well as the results of the cross-hole tests indicate a close connection between these two flow systems.

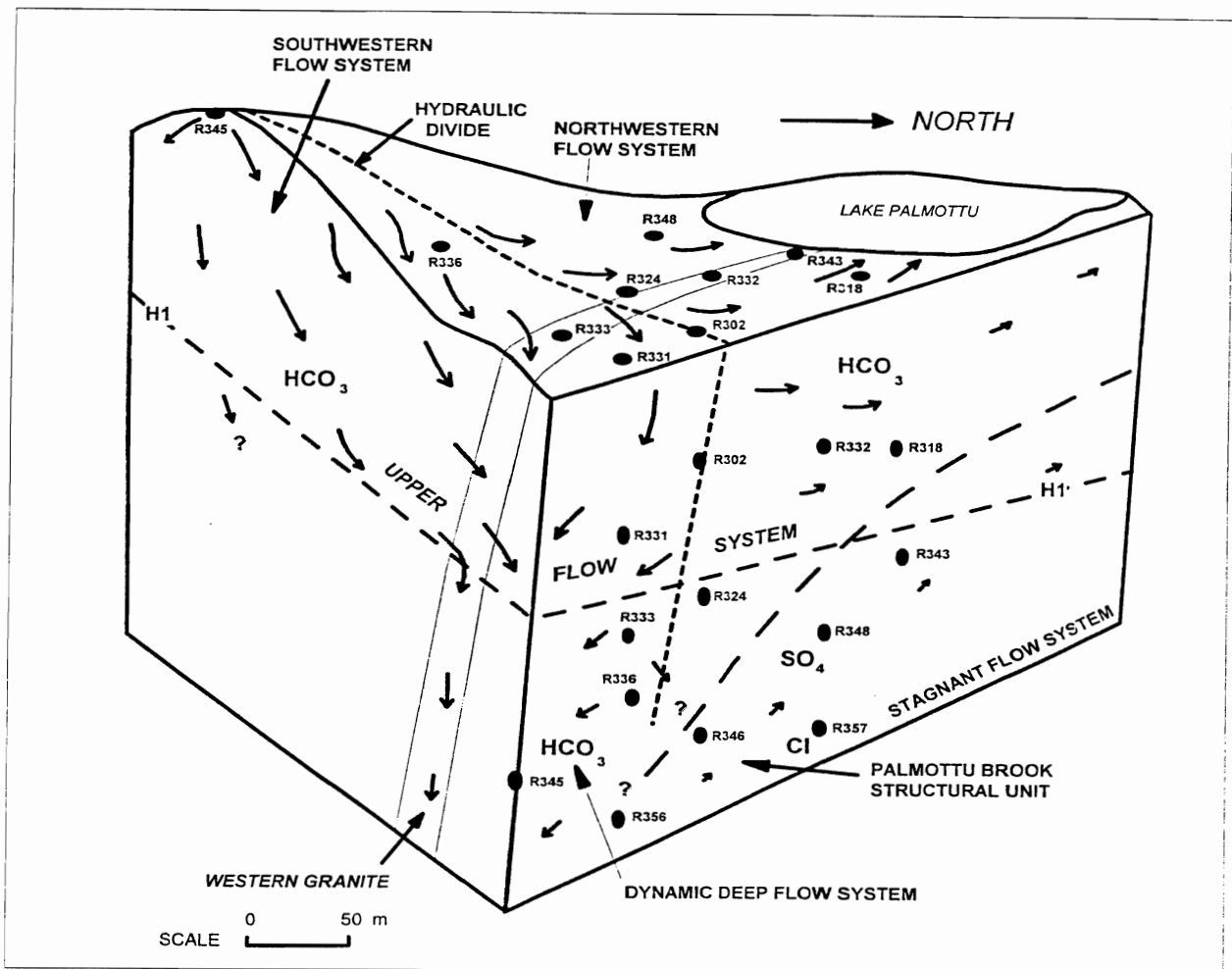


Figure 1. The conceptual hydrogeological model of the Palmottu site. Hydrogeochemical regimes with characteristic geochemical features, and the main flow directions are shown. The Eastern flow system is located to the right of the Palmottu brook structural unit, and is not shown in the diagram.

The rock unit below the Northwestern flow system is characterized by low head values. Groundwater in this Stagnant flow system is brackish and of Na-SO₄ or Na-Cl type. The chemistry and isotopes indicate a long residence time, up to the end of the last glaciation. This groundwater may discharge towards Lake Palmottu, but hydraulic data from the system are still scarce. The Stagnant flow system is separated from the upper dynamic Northwestern flow system by the subhorizontal structural unit H1. The Dynamic deep flow system seems to be isolated from the Stagnant flow system by a NE-SW striking barrier, revealed by the distribution of groundwater types as well as head data.

Task 1.1 Up-dating the structural model of the site

Objectives

The target of the Task was to evaluate and integrate all available structural and hydrogeological data in order to produce an up-dated hydrostructural model of the Palmottu site. This model would serve as the starting point for the planning and execution of forthcoming hydraulic tests (*e.g.* cross-hole testing and tracer tests) and subsequent modelling activities.

Results

Based on the interpretation of the interference tests of 1996, the following re-interpretations and specifications of the structural model were made: 1) the structures V1 and V2 were replaced by the broad Palmottu brook structural unit, incorporating the major uranium horizons and located between the Western and Eastern granites, 2) V4 and V5, delineating the Western granite, were replaced by the tabular Western granite body (*i.e.* Western granite structural unit), 3) the structure V6 in the Eastern granite was modified to incorporate a branch (E1) striking westward towards the south-east corner of Lake Palmottu, and 4) the subhorizontal structure H1 was presented as a broad zone extending from bedrock surface to approximately 100 m depth.

The integration of all available hydraulic and hydrogeochemical information revealed the existence of NE-SW trending (roughly parallel to the drilling profiles) less permeable boundary (a rock unit or a structural feature) located between boreholes R302 and R356. This boundary coincides with the water divide splitting the local flow system in two parts. A considerable amount of efforts were put identify this rock unit: two structural units were proposed, both steeply dipping. Based on the information from the deep geochemical borehole (R385), the depth extension of the structural model could be increased down to 500 m. The majority of open fractures is located in the upper 150 m of the borehole and seem to have a horizontal or subhorizontal dip. Deeper only two hydraulically important zones were detected, at 222 m and 406–408 m, respectively. The vertical structure V2 may be the upper counterpart for the deep 406–408 m structure. However, the borehole radar survey of late 1997 (Carlsten, 1997) clearly showed that this structure forms a subhorizontal tension-type structure and is not likely to be the depth-extension of the structure V2.

The results of the classification of the regional fracture zones (Kuivamäki, 1997) were utilized in groundwater flow modelling on regional and site scales (Koskinen and Kattilakoski, 1997).

Task 1.2 Hydraulic testing and interpretation

Objectives

The hydraulic testing programme was targeted at identifying and characterizing the most prominent hydraulic connections at the Palmottu. Various modelling approaches were used in order to describe the hydraulic inhomogeneity of the rock mass.

Results

Interpretation and modelling of the extensive hydraulic data set, gathered in 1996, was continued. The results were reported by Lampinen *et al.* (1997), Ludvigson (1997) and Floria *et al.* (1997). Major effort was put to combine the hydraulic field data with flow modelling results in order to produce a site-specific hydrogeological understanding of the site. The main conclusions of the hydrogeological work are as follows:

Hydraulic head values vary depending on precipitation and seasonal changes. Within the central area, head values gently follow the local topography. The boreholes R302, R318, R332 and R384, cross-cutting the Eastern granite, however, exhibit relatively higher head values compared to other boreholes. Based on distribution of head values, two main groundwater flow directions can be delineated: one northwestward towards Lake Palmottu and the other along Palmottu brook structural unit towards southeast. Precipitation and seasonal changes do not seem to have any major modification to this flow pattern.

The general results of interference tests at Palmottu indicate that the long-term responses are rather controlled by large-scale structural/lithological units of the bedrock than by narrow fracture zones proposed by the previous structural models (Ludvigson, 1997; Lampinen *et al.*, 1997; Floria *et al.*, 1997). During cross-hole tests, drawdown commonly propagated in the bedrock in a more three-dimensional manner, rather than within any specific fracture zone. The early-time responses indicated that drawdown tended to propagate along preferential directions in the bedrock (anisotropy), commonly following the general bedrock schistosity. With increasing time the test responses averaged out and the bedrock behaved more as an interconnected network of discrete fractures, approaching the equivalent of a fractured, porous medium with local anisotropy along planar structural units.

The cross-hole tests indicated that the following four structural units are present at Palmottu, each generating local hydraulic anisotropy in the bedrock: the subhorizontal unit H1, the Western granite (including zones V4 and V5), the Palmottu brook structural unit (including zones V1 and V2) and the Eastern granite structural unit (including zones V6 and E1).

The estimated transmissivity and storativity values of structure H1, on a site-scale, are in the same range as would be expected for near surface crystalline rock in Scandinavia. This supports the interpretation of structure H1 as a broad structural unit comprising the uppermost approximately 100 m of the bedrock. The calculated hydraulic parameters are rather uniform and no hydraulic connections with significantly higher transmissivities could be identified within this unit.

The proposed structures V4 and V5 of the Western granite are likely to be hydraulically interconnected forming one hydrogeological unit. For the Western granite, the calculated transmissivity is only slightly higher compared to that of the surrounding mica gneiss.

The results of the Palmottu brook structural unit including structures V1 and V2 were inconclusive (mixed responses). Therefore the hydraulic properties and the hydrogeological importance of this structural unit remain uncertain. The estimated

hydraulic properties based on one test seem to indicate similar hydraulic properties for this unit as for the other structural units.

In the Eastern granite (including zones V6 and E1) the response pattern was quite different compared to that of the central area; the responses were restricted only to structures within the Eastern granite, thus indicating that a hydraulic barrier may exist preventing groundwater flow across the Eastern granite to the central study area.

Water-level observations from open boreholes and packed-off sections of boreholes were interpreted (Lampinen, 1997) with special reference to flow implications and seasonal changes of head values.

Modifications to the earlier working programme

The inverse modelling approach was tested with the hydraulic pumping data received from H1. However, the variation of water conductivity values were too small to support effective inverse modelling, and the pumping tests were interpreted using versatile approaches by Geosigma, GTK/VTT and UPC (*op. cit.*). In addition, the detailed monitoring of pressure propagation along major structures during cross-hole testing, offered information on hydraulic connections and anisotropy (*cf.* Lampinen *et al.*, 1997, Lampinen, 1997) that partly could substitute for the information planned to be received by inverse modelling.

The open-borehole head data did not prove to be very useful in establishing realistic flow conditions, but it could be used as supporting evidence in revealing hydrogeological inhomogeneities within the study area (*cf.* Chapter 5 in Blomqvist *et al.*, 1998).

Task 1.3 Hydrochemistry as an indicator of groundwater flow

Objectives

The objective of this task was to compile a reliable hydrogeochemical database, based on groundwaters from packed-off borehole sections, in order to incorporate hydrochemistry to produce an integrated conceptual flow model of the site.

Results

An up-dated version of the hydrogeochemical database was distributed to the partners for modelling purposes (Kaija *et al.*, 1997). The database includes both open-borehole and packed-off samples from the old boreholes, packed-off samples from the deep borehole (R385 and R386) and samples from the overburden groundwaters. Furthermore, database was supplemented by snow and rain samples. The data was classified based on a quality assurance procedure, only data considered reliable could be used in forthcoming modelling purposes.

Based on main cations and anions, four different water types could be distinguished: near-surface dilute Na or Ca dominant bicarbonate groundwaters and deep saline Na-SO₄ or Na-Cl-type groundwaters. Environmental isotopes (³H, δ¹⁸O, δ¹³C and ¹⁴C) revealed at least two different sources for the water itself: groundwater of recent origin (residence time less than 40 years) and a glacial melt water (residence time up to ten thousand years). The former is mainly associated with the bicarbonate water and the latter occurs as a considerable component in both of the saline water types. The high SO₄/Cl and Br/Cl ratios and results from multi-component analysis indicate that seawater is an unlikely component of the saline waters. The calculations indicate (based on δ¹⁸O values) that the most

extreme Na-SO₄ and Na-Cl groundwater types contain 40–60 % glacial meltwater (Blomqvist *et al.*, 1994, 1995, 1997a; Pitkänen *et al.*, 1997; Laaksoharju *et al.*, 1997; Peña *et al.*, 1997; Negrel and Casanova, 1997).

The general hydrogeochemical understanding was successfully utilised in revealing the flow systems of the site scale (see Task 1.4).

Task 1.4 Flow modelling and integrated evaluation of hydraulic and hydrogeochemical results

Objectives

This task comprised: (1) flow modelling performed at regional, local and site scales, (2) evaluation and modelling of a combined pumping and tracer test, and (3) integration and combined evaluation of all hydrogeological and hydrogeochemical information. The target was to understand the site-scale flow system in order to be able to define natural flow paths which would satisfy the needs of a forthcoming transport analogue study.

Results

Based on a three-dimensional modelling approach and the finite element method a regional flow modelling was performed, indicating that predominant regional groundwater flow is toward the southeastern corner of the Palmottu area, *i.e.* from Lake Palmottu along Palmottu brook valley. A comparison of cut planes representing different depths shows that the regional groundwater flow is predominantly horizontal in character. The volumetric fluxes through the horizontal cut planes decrease strongly so that the infiltration below 150 m is only 14 % of the infiltration from the ground surface (Koskinen and Kattilakoski, 1997).

The site-scale flow model comprises a bedrock volume of 0.2 km² × 0.4 km in the close vicinity of Lake Palmottu. The most dominant site-scale structures were incorporated into the model: V1 - V6 and H1. The main inlet of regional groundwater flow is from the west, predominantly along regional east-west striking structures. The Palmottu brook fracture zone (V1/V2) is the main outlet channel from the site-scale area, and is characterized by horizontal flow. Calculations indicate that the groundwater flux along this structure is less than 1 % of the flux of surface water flow along the Palmottu brook itself (Koskinen and Kattilakoski, 1997).

The infiltration rate along structures V4 and V5, *i.e.* the Western granite structural unit, is quite high. The infiltration rate decreases markedly with depth, and for V4 at 150 m it is only 3 % of the flow at the 30 m level. Approximately one third of the flow is trending toward the drainage area (Lake Palmottu), whereas two thirds are trending toward the regional flow field. The flow toward the drainage area is mainly vertical with an upward trend below Lake Palmottu, whereas the flow in the southwestern part of the zone is predominantly horizontal following initial infiltration into the bedrock.

A hydraulic head maximum cross-cuts the Palmottu brook structural unit approximately along drilling profile R336. The head maximum acts as a local water divide separating groundwater flow towards the northeast (Lake Palmottu) and towards the southwest to the topographically lower areas.

The main conclusions from the integration of hydrogeology and hydrochemistry are presented below (*c.f.* Figure 1).

- Upper flow system: the upper part of the bedrock (the subhorizontal structural unit H1), is hydrogeologically relatively conductive. The groundwater is dilute, bicarbonate rich and young (usually less than 40 years). The flow systems mainly respond to topographic features, and therefore the Upper flow system can be divided into three sub-aquifers:

- * Southwestern flow system: the bedrock under the western hill area comprises two sub-aquifers separated by a subvertical NE-SW hydraulic divide which is approximately located on the line formed by the boreholes R345 - R336 - R302. Groundwater from the southern part of the divide flows towards the Palmottu brook valley (V1 and V2), and further to southeast.

- * Northwestern flow system: north of the hydraulic divide groundwater discharges to Lake Palmottu or towards the Palmottu brook valley, and further to the north.

- * Eastern flow system: the Eastern granite zone and the structural unit E1 seems to be hydraulically less connected to the other part of the subhorizontal structural unit H1. Recharging groundwaters, on the eastern hill, flow downward towards the Palmottu brook valley, and further to the north.

Deeper in the bedrock two flow systems have been identified:

- The Dynamic deep flow system is located below the subhorizontal structure H1 in the western hill area and along the Palmottu brook valley southeast from the hydraulic divide. Young, bicarbonate type groundwater similar to that of the Upper flow system, recharge here to greater depths (at least 200 m) along the Palmottu brook valley.

- An aquitard characterized by low head values and brackish Na-SO₄ or Na-Cl type groundwaters is located below the northern part of Palmottu brook structural unit. This Stagnant flow system is separated from the Northwestern flow system by the subhorizontal structure H1, and from the Dynamic deep flow system by the steeply dipping NE-SW trending groundwater divide.

Modifications to the earlier working programme

Stochastic network modelling was abandoned from the working programme after the withdrawal of RMC-E from November 1st, 1996. This modelling exercise was considered inappropriate at this stage, hence, the remaining funding of RMC-E was directed to other activities (support for redox measurements and sampling in borehole R386) with the approval from the Commission.

Task 1.5 Low-contamination drilling

Objectives

The objectives were to improve and test drilling techniques which would allow the sampling of representative formation groundwaters before serious contamination by flushing (or open-hole effects) would have taken place. In practice, hydrochemical sampling and restricted hydrogeological testing would be performed during drilling as soon as water-conducting fractures or fracture zones would be intercepted.

The major aspiration was that such representative groundwater samples would provide the reference hydrochemical (and environmental isotope, colloidal, microbial and organic)

database of Palmottu, to which all existing and future groundwater data can be compared with and quantitatively evaluated.

Results

The four most prominent fracture zones of borehole R385 were sampled for "complete chemical characterization" (CCC) using the mobile field laboratory of SKB: 96 m, 115 m, 222 m and 406–407 m. Drilling activities of boreholes R385 and R386 and different sampling techniques have been described in Ruskeeniemi *et al.*, 1996. For continued utilization of the borehole, a plan for a packer system for the borehole was prepared, and a subcontract to Posiva Oy was given to install necessary packers in the borehole. Accordingly, the five major water-conducting zones were isolated in late January 1997. Each packer section has a length of 5 m, except in the deepest zone at 406–407 m where a 10-m packer interval was required. The packer system allows both head monitoring and hydrogeochemical sampling from each of isolated zones. In October 1997 the packer system was lifted up and the mobile field laboratory unit was put in operation in order to collect the essential reference groundwater samples for the final evaluation of the hydrochemical sampling during low-contamination drilling.

Geological and mineralogical study of fracture infilling and rock types of borehole R385 were continued at Ciemat and GTK. Pèrez del Villar *et al.* (1997) produced a preliminary mineralogical study on selected fracture infilling and rock samples. The study confirmed the results received from previous studies: The main fracture-infilling mineral is calcite associated with variable amounts of clay minerals, usually kaolinite or smectite. They also received microanalytical evidence for a uranium-carbonate phase along with the common uranium silicate minerals in the alteration zone of uraninite.

A geological description of the borehole lithology and water-conductive fracture zones was produced (Ruskeeniemi *et al.*, 1997).

Modifications to the earlier working programme

The final evaluation of the adopted low-contaminating drilling technique required re-sampling from the packed-off sections of borehole R385. Most of these control samples have been taken and the preliminary evaluation will be made during spring.

WP 5, PA-Activities

Objectives

An important part of the Palmottu project is the close integration of the natural analogue study with performance assessment (PA) of repositories for radioactive waste. To ensure that this integration takes place the project includes a special part, Workpackage 5 (WP5) dealing with PA issues. Workpackage 5 activities extend over both Phases I and II of the project. The main objectives for Phase I were to prepare a working programme for Phase II based on the site-characteristic hydrogeological conditions.

Results

The present understanding of the flow systems provided an acceptable basis for planning and optimising the forthcoming PA-oriented studies at Palmottu. The following processes

and phenomena were considered to be the most beneficial with respect to future PA-activities:

Scenario development. The Palmottu natural analogue provides an opportunity to study, document and evaluate the consequences of the glaciations on the hydrogeological and hydrogeochemical system. This task will mainly deal with the near field and far field scenario analyses utilizing the paleohydrogeological results of WP2. The modelling concepts used in the consequence analysis will be studied in Tasks 5.2 and 5.3 of WP5.

Process-orientated studies. These are studies of single phenomena not related to any particular geological or hydrogeological system within the site. Such phenomena that are of obvious concern in PA and that are well suited for studies at Palmottu are:

- * Redox control in groundwater-rock system
- * Propagation of redox fronts (dissolution-retardation mechanisms)
- * Matrix diffusion and sorption
- * Uranium sinks (*e.g.* association with calcites)
- * Occurrence and effects of microbes
- * Colloidal transport of trace elements

Groundwater evolution in infiltration areas. Two main recharge areas have been identified at the Palmottu site, the hills of Western and Eastern granite. Boreholes penetrating plausible flow paths at different locations give the possibility to study several aspects of the evolution of groundwater chemistry of the infiltrating waters. Efforts aiming at the identification of the chemical and physical parameters controlling the uranium transport in these relatively simple sub-systems would contribute to increase understanding of water-rock interaction in oxidizing granitic environments.

The main uranium mineralization. The main uranium mineralization is located along the Palmottu brook valley. The relevant flow systems here are given in Figure 1. The uranium mineralization at Palmottu mainly consists of uraninite, a mineral that resembles the nuclear fuel used in many reactor types. Therefore, studies of the dissolution of uraninite, formation of secondary uranium minerals *etc.* are of value for the identification of dissolution mechanisms that could be included in models used in PA. The long time scale of the evaluation of the Palmottu site may help to observe extremely slow processes that are difficult to identify in laboratory studies.

Transport models of different types will be applied and tested using the data from Palmottu. Tests of models for individual processes, *e.g.* sorption and matrix diffusion, are foreseen to be performed on rather small scales, in the range of centimetres or tens of centimetres. Attempts will also be made to test transport models including advective transport models, and models to couple transport and geochemical reactions.

Biosphere interface. Measurements of uranium concentrations in the Quaternary overburden and in lake sediments make it possible to analyze transport processes at and across the interface between the geosphere and the biosphere. At this interface strong chemical gradients may occur, causing, for example, the redox conditions to change rapidly with the effect of precipitation or mobilization of radioelements. A further benefit of the site is that the time frames for the biosphere transport are well established as the deglaciation stage of the area is possible to date using several independent methods.

III INITIAL PROGRESS OF PHASE II STUDIES (1997-08-01—1998-01-31)

WP 2: Geochemical evolution of the water-rock system (GTK)

Objectives and main activities

The origin and evolution of the groundwaters will be interpreted and conceptual evolutionary models tested. Hydrogeochemical interpretation will be supported by isotope studies of identified fracture coating minerals known to be contact with sampled groundwaters. Testing of selected hydrogeological connections detected during Phase I will be carried out in order to verify flow routes of 1) the surface-close eastern flow system and 2) the depth extensions of central part of the southwestern flow system. WP 2 incorporates the following main tasks:

- Task 2.1 Hydrochemical and geochemical data sets,
- Task 2.2 Detailed characterization of selected water-rock systems,
- Task 2.3 Geochemical modelling of water-rock interaction.

Task 2.1. Hydrochemical and geochemical data sets

Groundwater

The hydrogeochemical results together with hydraulic and flow information were used to produce the conceptual hydrogeological model of the site as presented in the previous chapter (also Figure 1). New analytical and modelling work are presently proceeding in order to expand the previous interpretations into the more rigid understanding needed in the performance of the present studies. Two new boreholes have been drilled in order to characterize more evolved HCO₃-waters of the Palmottu brook structure (borehole R387: ~ 400 m) and to replace destroyed drillcore of borehole R335 in Eastern granite (borehole R388: ~ 100 m).

The groundwater sampling campaign from packed-off borehole sections of the old exploration boreholes started in May after snow melting and continued to December. The objectives of Phase II sampling campaign were:

- to evaluate the representativeness of Phase I sampling campaign and to study possible seasonal changes in groundwater chemistry by
 - * resampling the old exploration boreholes (11 sections) and
 - * resampling and performing redox-measurements of the deep borehole R385 (5 sections)
- to complete the groundwater data sets of different flowsystems by
 - * sampling the boreholes using a modified, narrower packer configuration (2 sections)
 - * sampling from borehole sections not previously sampled (8 sections)
 - * making additional redox-measurements (R318/3 sections)

Deep borehole R385 was resampled between November 1997 and February 1998 using two sampling methods; section 88–93 m was sampled using Posiva's nitrogen driven pump; sections 94–100 m, 112–117 m, 220–226 m and 403–409 m were sampled using SKB's mobile field laboratory unit. During pumping from section 88–93 m, Eh, pH,

oxygen content and electrical conductivity were measured in the flow-through cells at the surface. During pumping from the four latter isolated borehole sections, there was a continuous registration of downhole pH, Eh (electrodes of glassy carbon, gold and platinum) and temperature, backed-up by measurements in the flow-through cell unit installed at the field laboratory. In addition, the oxygen content, electrical conductivity and flow-rate were registered at the surface. For the sake of data comparison, Eh, pH, oxygen content and electrical conductivity were also measured with GTK's electrodes at the surface.

The up-dating and evaluation of hydrochemical data set continued throughout 1997. Besides previously analyzed major and trace elements (*e.g.* Ca, Mg, Na, K, Cl, SO₄, HCO₃, NO₃, Br, F, Si, Fe, Mn, Sr, B, Ba, Li, Al, Rb, V, Cr, Co, Ni, Mo, Cu, Zn, Cd, Pb, As, Sb, Th and U), the following trace elements and REE's were included in the GTK's routine ICP-MS analysis package; Ce, Dy, Er, Eu, Gd, Ho, La, Nd, Pr, Sm, Tb, Tm, Yb, Lu, Sc, Ti and Y. Concentrations of relevant environmental isotopes from selected borehole sections have also been analyzed, *e.g.* ³H, $\delta^2\text{H}$, $\delta^{18}\text{O}$, $\delta^{13}\text{C}$, ¹⁴C, U-series radionuclides, $\delta^{34}\text{S}$, ³⁷Cl/³⁵Cl, ⁸⁷Sr/⁸⁶Sr. Also gas analyzes from borehole R385 have been taken for routine gas analyses as well as for selected isotope analyses.

Some preliminary analysis of ¹¹B/¹⁰B were done at BRGM during 1997 in order to study the origin and evolution of deep groundwaters at Palmottu. Because ¹⁰B and ¹¹B show large relative mass difference and boron has a high geochemical reactivity, significant isotope fractionation produces large variations in ¹¹B/¹⁰B ratios in groundwater samples from different geological environments. Thus, the boron isotopic ratio is a sensitive tracer for hydrochemical evolution of groundwater.

Results

The tritium content in precipitation at Palmottu drainage area is quite stable, being about 11 TU. Seasonal fluctuations in $\delta^2\text{H}$ and $\delta^{18}\text{O}$ are obvious, varying between -162.4 to -52.8 and -22.35 to -7.53 respectively; the lightest ¹⁸O signature value representing snow samples.

The previously documented glacial meltwater signature, $\delta^{18}\text{O}$ being -16.26, from borehole R385/217–222.9 m section nearly disappeared during new sampling campaign. The ¹⁸O signature was clearly heavier, $\delta^{18}\text{O}$ being -13.66. This could be due to slightly different packer configuration, but also due to the fact that during 1996 and 1997 sampling campaigns the pumped volume of groundwater from this section was calculated to be over 3000 l. This may have lead to emptying of an isolated, restricted groundwater storage.

Three groundwater types (R385/403–409 m: Na-Cl, R346/240–246 m: Na-SO₄ and R354/320 m: Na-Cl-HCO₃) were analyzed for ⁸⁷Sr/⁸⁶Sr and $\delta^{11}\text{B}$. Strontium concentrations vary from 30 to 1075 $\mu\text{g/l}$, whereas boron concentrations vary from 264 to 675 $\mu\text{g/l}$. $\delta^{11}\text{B}$ ranged from 33.6 ‰ in sample R354 to 43.5 ‰ in sample R385. The same samples respectively show a ⁸⁷Sr/⁸⁶Sr ratio ranging from 0.744416 to 0.720046. Pure marine ⁸⁷Sr/⁸⁶Sr signature was not found. Groundwaters from R346 and R354 are depleted in $\delta^{11}\text{B}$ with respect to mean sea water, whereas groundwater from R385 is slightly enriched in $\delta^{11}\text{B}$. These studies support the mosaic nature of the Palmottu hydrogeological system. Additional groundwater samples from Palmottu and reference samples from Baltic seawater are presently being analyzed for ⁸⁷Sr/⁸⁶Sr and $\delta^{11}\text{B}$.

Relatively strong variations in uranium concentrations have been revealed especially in groundwaters from mineralized area. For example, uranium concentration in borehole R384/57–94.25 m section varied between 94 $\mu\text{g/l}$ (sampling date 13.5.97) to 132 $\mu\text{g/l}$ (sampling date 18.4.96). Uranium content in borehole R318/50–80 m section varied from

89.5 $\mu\text{g/l}$ (sampling date 12.12.97) to 648 $\mu\text{g/l}$ (sampling date 24.4.96). These kinds of variations are probably connected to seasonal variations, and thus in recharge conditions.

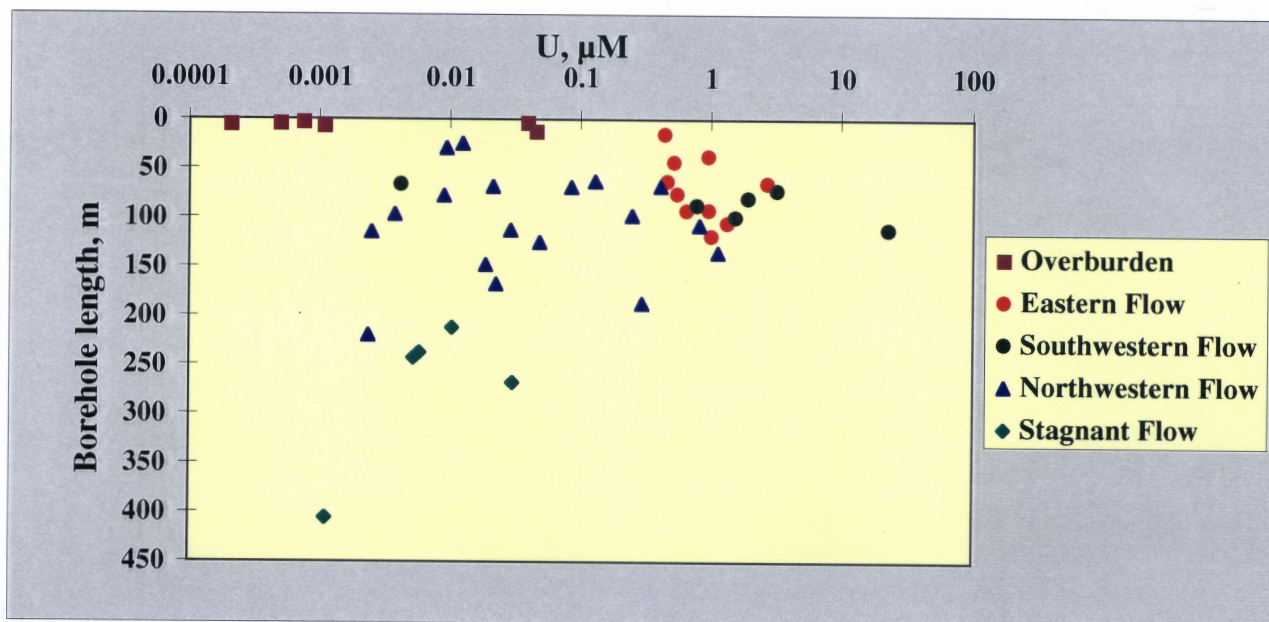


Figure 2. Variation of uranium versus borehole length.

Generally, the enrichment of uranium is associated with shallow depths in the hydraulically active Upper flow system and occurs in proximity of the U-mineralization (Figure 2). The studies of solubility control factors of uranium are presently proceeding.

Redox-measurements

In November-December 1997 redox-measurements were done in borehole R318, which penetrates the uranium mineralization and is part of the Eastern flow system. In borehole R385 redox was re-recorded with the aid of SKB's mobile field laboratory. At the moment redox-measurements are going on in borehole R318 with nitrogen driven pump system. In spring the new deep borehole R387 will be sampled with mobile field laboratory and redox-values will also be registered. Additional redox-measurements will be made in selected boreholes of the Eastern flow system.

The redox-measurements from the previous field seasons at Palmottu mainly concentrated studying groundwater redox-conditions in the fractures of the mineralized zone. Eh-values varied from +55 mV to -92 mV. Also the oxidation state of uranium was analyzed from several samples. The good correlation between analyzed uranium oxidation state and the redox speciation of uranium is striking. This indicates that uranium is in redox equilibrium with the potential determining redox-pair. Two measured sections of borehole R346 contained dissolved uranium predominantly as U(IV) (Ahonen *et al.* 1994).

Most of the redox-measurements in 1996-97 were done in borehole R385, where the bedrock is unmineralized. All Eh-results below about 90 meter of borehole R385 indicate very reducing conditions, close to the lower stability limit of water, which is defined by

the H_2/H_2O redox pair. In spite of extremely reducing conditions, U(VI) seems to be the prevailing redox species of dissolved uranium.

Laser-probe studies on fracture infilling calcites

Six calcites from three boreholes were studied (Wallin, 1998). First a fluorescence analysis was performed for each sample to identify possibly existing different calcite generations. The results were used to direct the laser-probe analyses proper.

The range of $\delta^{13}C$ -values ($-7 - +5$ ‰) suggest two dominant sources for the carbon input. However, a third in-mixed isotopic signature observed in borehole R348 may point to an additional source. Most of the $\delta^{18}O$ values ($+10 - +20$) in all analyzed calcite fracture infillings indicate equilibrium with present groundwaters. The observed light $\delta^{13}C$ -values may be attributed to a hydrothermal source. The depleted $\delta^{18}O$ signatures in some of these samples can be achieved at low temperatures from a mixture of present-day meteoric waters and waters from significantly colder periods. Hence, dissolution of hydrothermal fracture infillings in a cold, possibly glacial water would be one way to explain the observations. The positive carbon isotope values may be derived from the fractionation of carbon between oxidized and reduced components. Light carbon is preferentially bound to reduced phases, for example, graphite, while the heavy carbon fraction is incorporated to carbonates. The intermediate $\delta^{13}C$ values between the extremes may result from mixing of the end-members, although, atmospheric input also introduces carbon with $\delta^{13}C$ around 0 ‰. Organic carbon source with very light isotopic composition seems to negligible at Palmottu.

The data received from the infillings show that a fracture may have a very complex history. Some fracture infillings have precipitated in conditions with constant C and O. However, in most samples the isotope compositions have different kind of trends or regular variation in the course of precipitation (Figure 3). The possible reasons for this include changes in the isotopic compositions of the parent fluid (open/closed system, mixing) or changes in physical conditions (mainly temperature).

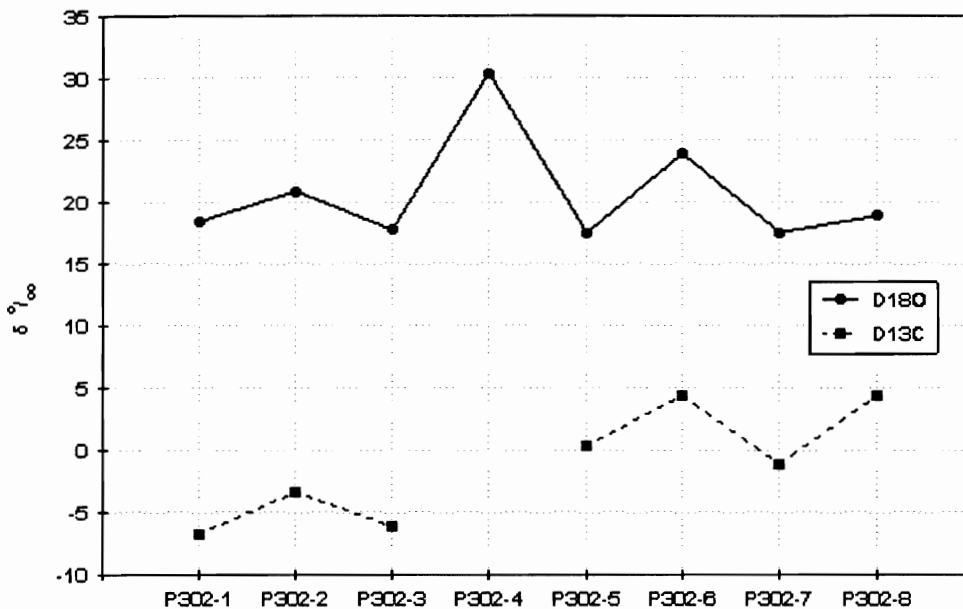


Figure 3. $\delta^{13}C$ - and $\delta^{18}O$ -values across a calcite infilling with a layered texture. The depth in the infilling is increasing from left to the right (R302/52.40 m).

U series disequilibrium studies of calcites in borehole R346

Occurrence of U in fracture calcites was studied at UHRAD in order to see if the changes in the nature of the continuous interaction of the groundwater can be observed. Calcites were sampled from assumedly open fractures in the borehole R346 as a function of depth, starting from fractures in the near surface conditions down to over 174 m where more reducing conditions prevail. In addition to U series disequilibrium measurements also the redox speciation of U in the calcites was studied determining the oxidation states U(IV) and U(VI) (for methodological details see Ervanne and Suksi, 1996). The sample material consisted of seven calcite-bearing fracture infillings of variable thicknesses and with different mineralogical associations. Also groundwater data was available from the borehole enabling correlation of the data from groundwaters and fracture infillings. The groundwater samples were from the sections 65–71 m and 122–128 m, the upper section covering the depth of one calcite sample and lower section three calcites.

Results

The results of the radiochemical analyses and U series disequilibrium measurements are given in Table 1. Duplicate analyses, due to very small sample amounts, could be performed only for the samples 38.02 m, 68.15 m and 174.45 m. The concentration of U ranged from 0.51 ppm to 70 ppm, indicating differences in the migration history of U in the fractures. This view is supported by the variations in the U-234/U-238 activity ratios (0.95-3.7) measured in calcites. The activity ratios of the two calcites within the section 122–128 m are similar as measured in the groundwater. The similar activity ratios suggest that the U in the groundwater in these two fractures may be related to the U accumulated in the calcites over 100 000 years ago.

Further information of the long-term behavior of U is obtained by examining Th-230/U-234 activity ratios. In the calcites with lower U concentrations (10.55 m, 1.1 ppm and 68.15 m, 0.6 ppm) considerable U leaching is indicated by the high Th-230/U-234 activity ratios: 2.9 and 2.1-5.5, respectively. Measured Eh of the corresponding groundwater (*i.e.*, the packered section 65–71 m) is only slightly negative (-11 mV), but clearly higher than in the section 122–128 m (-80 mV), suggesting the mixing of more oxidizing waters in this section. In the calcites deeper U accumulation appears to dominate: Th-230/U-234 values are between 0.3 and 1.0. Noteworthy is the observation of U accumulation in the 38.02 m fracture where, according to the data from the calcites above and below, leaching of U is expected to take place.

The Th-230/U-234 and U-234/U-238 activity ratios also provide a rough estimation of the U accumulation/leaching time frames. The very high Th-230/Th-232 activity ratios measured (in most of the samples) indicate that the measured Th-230/U-234 activity ratios are practically free of detrital (nonauthigenic) components and can thus be considered as the result of only radioactive decay and U accumulation.

The unusually high Th-230/U-234 activity ratios in the calcites 10.55 m and 68.15 m reflect the very recent (of the order of thousands of years) U leaching from the calcites. In the depth of 174 m very low Th-230/U-234 activity ratio (0.3 and 0.53) was measured, indicating U accumulation within the last 70 000 years or even earlier in this calcite.

Table 1. Radiochemical data of fracture coating samples from the borehole R346. The error (in parentheses) is one sigma and is due to the standard deviation of the radioactivity measurement.

Depth (m)	U (ppm)	Th (ppm)	U-234/U-238	Th-230/U-234	Th-230/Th-232
10.55	1.11 (0.15)	1.8 (0.9)	0.95 (0.20)	2.9 (0.7)	5 (3)
38.02	28 (1)	bdl	1.26 (0.07)	0.95 (0.08)	--
"	51 (2)	0.4 (0.5)	1.21 (0.08)	0.75 (0.07)	392 (580)
68.15	0.61 (0.13)	0.26 (0.36)	1.56 (0.43)	5.5 (1.1)	63 (89)
"	0.51 (0.12)	bdl	2.0 (0.6)	2.1 (0.5)	17 (9)
124.15	35 (2)	1.8 (0.8)	1.89 (0.14)	1.03 (0.17)	114 (44)
126.98	70 (3)	1.3 (0.6)	2.66 (0.17)	0.86 (0.16)	374 (180)
127.21	33 (2)	bdl	3.7 (0.3)	0.72 (0.13)	-
174.45	1.0 (0.1)	0.25 (0.17)	3.42 (0.45)	0.30 (0.05)	13 (9)

bdl = below detection limit

-- = could not be determined

The separation of the U(IV) and U(VI) fractions from the calcites was also attempted. Because of the very first attempt, the results obtained should be handled cautiously. However, the interesting feature observed is that the proportion of U(VI) appears to increase deeper in the bedrock. Practically all U found in the calcite at 175 m was in the U(VI) state. This was unexpected, but may indicate the downward transport of the redox-front.

The relatively large amounts of U(IV) in the upper part may be explained by the preferential leaching of U(VI). The occurrence of the U(IV) fractions on the whole remains unclear. Since the separation technique applied is still in its developing stage, the possible experimental artifacts can not be excluded.

This study shows that the changes occurred in the nature of the continuous interaction of the groundwater in discrete fractures can be detected and, importantly, interpreted in a rather reliable way. The reliability of the interpretation can be largely enhanced by increasing the density of the sampling.

Microanalytical studies on U-minerals

The aims of microanalytical studies are to extend the existing geochemical data base of radioactive minerals found at Palmottu and to gain better understanding of the mineralogy and geochemistry of the alteration products of the primary uraninite. About 40 spot analyses of uraninites and its alteration products were selected to the geochemical data base from the large microanalytical data set obtained. This data base can be used both in redox studies and in radionuclide migration studies.

Polished uraninite grains and polished thin sections of mineralized rocks were studied using Cameca SX50 electronprobe microanalyser (EPMA) located in Geological Survey

of Finland. Morphological observations, spot analyses and geochemical profiles and element mapping was done (Hansèn and Ruskeeniemi, 1998).

Results

Previous studies (Ruskeeniemi *et al.*, 1994, Perez del Villar *et al.*, 1997) have shown that the main alteration product of uraninite is a uranium silicate, most likely coffinite. However, the alteration mass is quite heterogenous and also another phases may be present. Elemental distribution maps were a new method in studying the alteration rim. The aim was to see if the distribution patterns of some elements correlate giving indications of discrete mineralogical phases.

Two uraninite samples were chosen for the elemental mapping. In one sample P and Th showed enrichment patterns in certain parts of the sample. Another interesting thing was the absence of Pb. Usually uranium silicate contain a couple of percents of Pb, but in the mapped area the concentrations were close to zero. In second sample Pb shows a normal distribution pattern in uranium silicate. Only in a couple of places where calcium is higher, Pb concentrations are negligible. Th seems to be enriched just around the uraninite grain up to 12 wt-%. The variation in the concentrations of other elements is in the order of one percent making any interpretations unreasonable.

Chemical profiles were made across altered uraninite grains hosted by different silicate minerals to see if there is any variation in the alteration of uraninites and in the chemistry of the alteration products. Uraninite grains are found embedded in biotite, plagioclase or in quartz, the first one being by far the most important host.

The mineralogical zoning around uraninite grains is different depending on the surrounding silicates. These features can partly be attributed to the alteration of uraninite and partly to the damages in silicate lattices caused by radiation. Some indications of geochemical zoning was observed in this study, although more data is needed to confirm these features before making any generalizations.

In quartz only a thin enveloping zone of the alteration product of uraninite, uranium silicate, is observed as the lattice of quartz seems to be resistive against radiation. Compared to "average" U-silicates at Palmottu these show slightly elevated Th and strongly elevated Zr concentrations. Migration of radionuclides into quartz matrix has occurred along some microfissures.

In plagioclase environment the disintegration of the host silicate is distinct. Two different types of clay haloes can be found around uraninites. There is some U in both of these haloes, while Th and Pb are insignificant. Also the fresh plagioclase close to the haloes contains some U. The uraninite is bordered by U-silicate rim.

The effect of radiation in biotites is displayed by a colourless halo. Almost total loss of potassium is the most striking geochemical feature. Migration of U and Pb in to this zone is distinct. U-silicate is also in this system the alteration product of uraninite. Migration of radioactive material along microfissures and cleavage planes is seen sometimes (Figure 4).

Conclusions

The microanalytical work revealed interesting patterns in the distribution of certain elements. However, it is not easy to explain the behaviour of Th, Pb and Zr in the system since it is not uniform or systematic in all samples studied. Some features point to differences in the mobility of the elements, *e.g.* U vs. Th, causing geochemical zonation around the source mineral.

This study was not yet able to identify new U-minerals or compounds from the alteration rim, although the chemical heterogeneity would suggest them to exist. However, the analysis of the extensive amount of chemical data is continued.

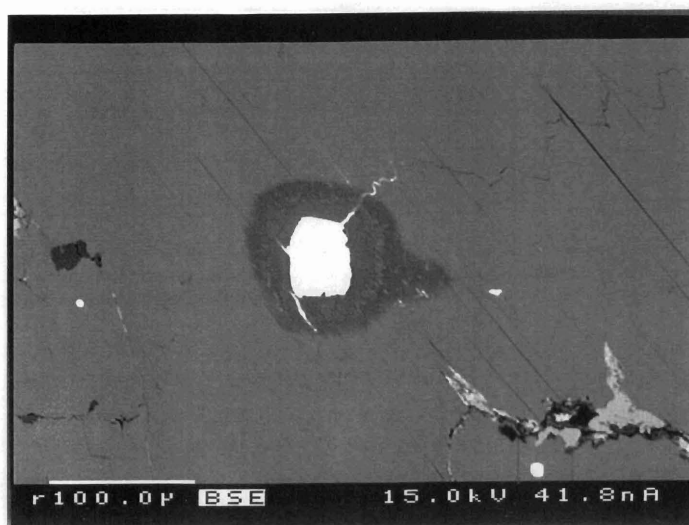


Figure 4. Backscattered electron image of a uraninite grain (white) embedded in biotite. (sample from R304/83.75 m). Uraninite is surrounded by halo characterized by a loss of potassium. Note the migration of heavy elements, mainly U and radiogenic Pb, along the cleavage planes and along microfractures. The diameter of the halo is about 0.1 mm.

Task 2.2 Detailed characterization of selected water-rock systems

Studies on migration route

In order to characterize open fractures and their orientations within the boreholes and to revise the hydrostructural model of the Eastern flow system, a TV-logging was carried out in boreholes R302, R318, R335 and R384. Also 19 other boreholes of the central borehole area have been TV-surveyed (Paulamäki, 1997). The possible water conductive sections determined by the cross-hole tests and spinner measurements were the main focus of interest. This method facilitates the exact location of the open fractures from drill-core sample, as well as, the orientation of the fractures (see Paulamäki *et al.*, 1997).

The mineralogical description of the open fractures was made in late autumn at the drill core archive in Loppi. The most prominent fractures from six boreholes were located from video tapes and the drillcore boxes were marked. The fractures and fracture infillings were described and photographed. Detailed mineralogical study of the major open fractures in the Eastern flow system will be done in co-operation with GTK and Ciemat.

Another aim of the work was to collect testing and background samples for sorption studies (Task 4.1).

Results

About 150 open fractures were located and studied from boreholes R302, R318, R332, R335, R373 and R384. Special attention focused on the tracer-test sections in the Eastern granite. Locating of major fractures was rather successful except in drillcore of R335, which is badly damaged. A new borehole (R388: borehole length ~ 100 m) have been drilled in order to replace this destroyed drillcore.

In many cases one or two open fractures are responsible for the hydraulic anomalies in the Eastern granite, although the core may be strongly broken. The apertures are usually less than 10 mm. The orientation of the major open fractures varies from borehole to another and fractures are mostly subhorizontal. The subvertical fractures are mostly in the direction of the schistosity. As indicated by the cross-hole tests, the boreholes of the Eastern granite are hydraulically connected, but considering the fracture orientations determined by TV-survey, no direct planar connections seem to exist between the boreholes along the structures V2, V6 and E1 of the structural model. This is also supported by the interpretation of the tracer testing.

Figure 5 presents the most likely hydraulic connections between boreholes R302, R335 and R384. According to TV-survey the possible connecting fractures of R302 and R335 are $150/90^\circ$ (and $075/71^\circ$) in R302(91-93 m) and $240-243/63-88^\circ$ in R335 (45.95 m, 62.16 m and 67.50 m). The possible connecting fractures of R335 and R384 include $198/11^\circ$ (34.50 m), $227/37^\circ$ (48.80 m), $228/69^\circ$ (51.45 m) and $198/11^\circ$ (34.50 m) of R384 (Paulamäki *et al.*, 1997).

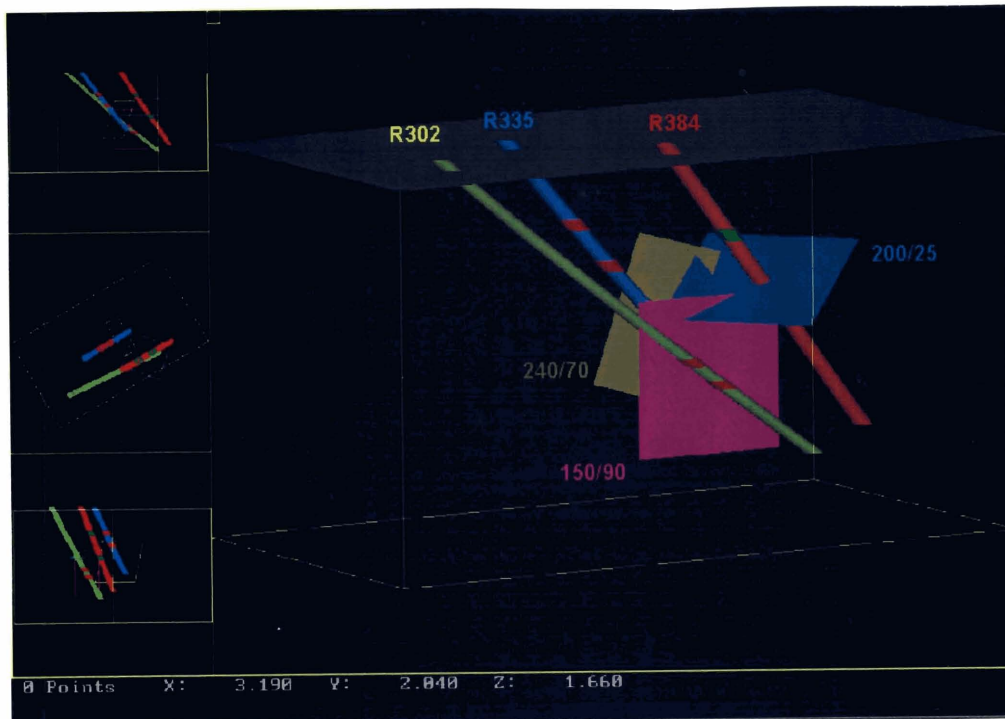


Figure 5. The most likely hydraulic connections (open fractures) between boreholes R302, R335 and R384.

Tracer testing

The plans for conducting the tracer test were completed during summer of 1997, and an extensive tracer test was conducted as joint campaign between Geosigma Ab, GTK and SKB (Gustafsson *et al.*, 1998). The objectives of the tracer test were primarily to verify hydraulic connections within the Eastern granite, with the proposed hydraulic features V6

and E1, and to determine residence time, magnitude of dispersion, hydraulic conductivity of flow paths and flow porosity.

The general idea of the tracer test was pumping in the deepest borehole section R302/88-98 m in the Eastern granite, creating a converging flow field while three non-sorbing fluorescent dye tracers (Amino G Acid, uranine and RdWT) were injected in R384/30-57 m, R335/54-81.5 m and R335/30-45 m packed-off sections (Figure 6). Distances between tracer injection and abstraction were 28-46 metres and the head difference between pumping borehole and injection zones were 26-28 metres. An automatic sampler was used for the water sampling in the pumping borehole.

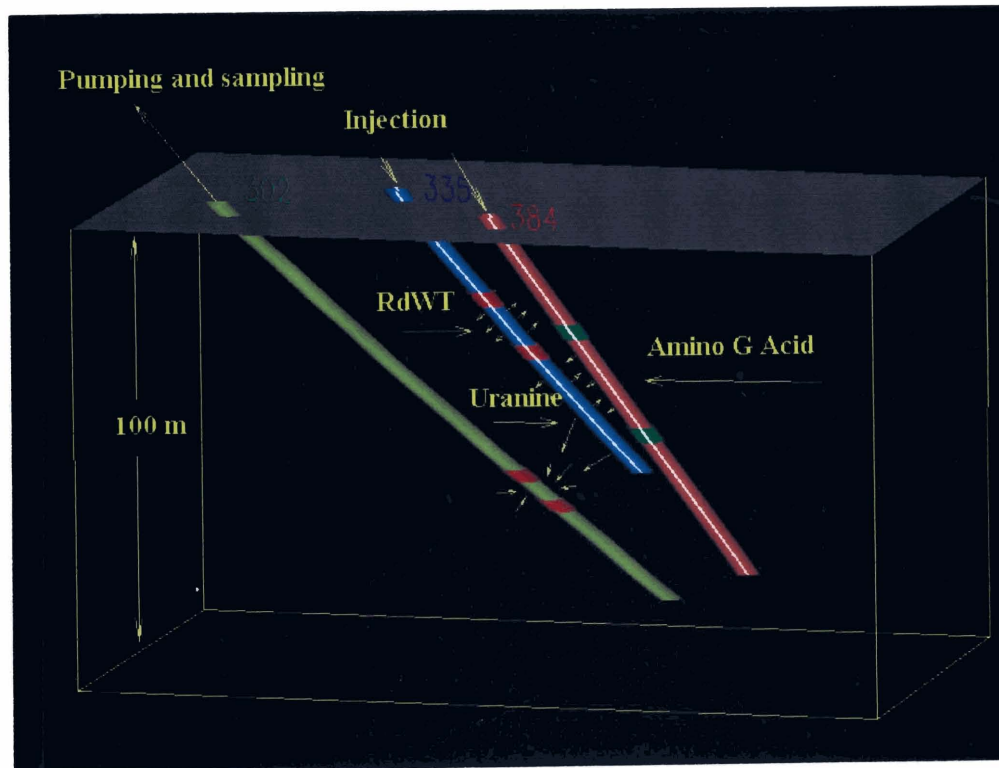


Figure 6. A cross section of the Eastern granite and boreholes R302, R335 and R384. The pumping section in R302 and tracer injection sections are shown.

The results from the tracer test consisted primarily of tracer breakthrough curves. Interpretation of the breakthrough curves using inverse modelling (curve fitting) gave residence times and a measure of dispersion. At a secondary level on interpretation the hydraulic conductivity of the fracture flow paths and fracture porosity were calculated, based on residence times. In addition, the pumping flow rate and groundwater head in the pumping and injection boreholes were measured. A summary of the determined flow and transport parameters is presented in Table 2.

The modelling results show that dispersion is large. Dispersion lengths were determined to 8–13 meters, at travel distances of 28–33 m ($Pe = 4.2-2.1$). The high dispersivity is most likely the result of an interconnected network of discrete fractures approaching the equivalent of a fractured porous medium.

The calculated hydraulic conductivities in route R384 to R302 and in route R335–R302 were 1.4×10^{-4} m/s and 1.6×10^{-4} m/s, respectively. Comparison with some Swedish sites indicates that the flow rates R384–R302 and R335–R302, consist of many short

interconnected flow paths more like a cube of bricks than a few connected continuous fractures.

The mass balance determined fracture aperture e^m , related to the pore volume of the rock unit investigated, is about 1×10^{-2} m in both flow routes and the frictional loss aperture e^f , related to the aperture of the individual flow paths, is about 1.5×10^{-5} m in both flow routes. These values are typical for a shallow fractured bedrock. The e^m/e^f ratio also indicates many small aperture fractures contributing to flow in the investigated rock volume.

Both mass balance ratio, θ_k^m , and conductivity ratio, θ_k^r , determined flow porosities are somewhat higher in route R384–R302 than in R335–R302. The porosity in the investigated rock at Palmottu is slightly lower than shallow fractured granite at compared sites in Sweden and Spain.

The tracer test verified hydraulic connections in the Eastern granite. However, the evaluation of tracer breakthrough curves indicates that transport in the granite between R384–R302 and R335–R302 took place in many, low conductive, interconnected fracture flow paths. The proposed features V6 and E1 can thus not be seen as a set of a few continuous highly conductive fractures. The flow porosity is in the lower margin of what has been determined in other shallow fractured granites. Transport parameters could not be determined for the route R335/30–45 m to R302/88–98 m since the tracer did not arrive during the time of the experiment. Borehole R335/30–45 m locates in the mica gneiss and seems thus more or less hydraulically separated from the pumped borehole R302 deeper in the Eastern granite.

Table 2. Summary of calculated flow and transport parameters.

Parameter	R384 II → R302		R335 I → R302	
	30-57 m	88-98 m	45-81.5 m	88-98 m
Fracture conductivity, K_f (m/s)	$1.4 \cdot 10^{-4}$		$1.6 \cdot 10^{-4}$	
Fracture aperture, e (m)				
Frictional loss, e^f	$1.5 \cdot 10^{-5}$		$1.6 \cdot 10^{-5}$	
Mass balance, e^m	$1.0 \cdot 10^{-2}$		$9.2 \cdot 10^{-3}$	
Flow porosity, θ_k (-)				
Conductivity ratio, θ_k^r	$2.9 \cdot 10^{-3}$		$6.4 \cdot 10^{-4}$	
Mass balance ratio, θ_k^m	$5.6 \cdot 10^{-4}$		$3.9 \cdot 10^{-4}$	
Average hydraulic conductivity, K (m/s)	$3.9 \cdot 10^{-7}$		$1.0 \cdot 10^{-7}$	
Thickness of rock unit, W (m)	18.5		23.2	

Fracture infillings

The mineralogical group at Ciemat (Pérez del Villar *et al.*, 1998) report the following results from fracture infillings of boreholes R385 and R386:

Major minerals:

The semiquantitative mineralogical composition of the 12 samples analyzed by XRD (Table 3) shows that the neoformed phyllosilicates in most of the samples are: smectite, palygorskite, muscovite-illite(?), kaolinite and chlorite, in different proportions. In those samples close to the Eastern granite, kaolinite is the most abundant clay mineral (samples at 451.42 m and 460.40 m). Corrensite, an illite-chlorite interstratified clay mineral, has been detected by SEM+EDX in most of the samples.

Calcite is present, in different proportions, in all the samples. In those samples taken at 81.24 m (R386), 219.70 m, 406.20 m and 460.40 m, calcite is the most important neoformed mineral. Idiomorphic to xenomorphic calcite, with minor Mn and Fe and sometimes showing pit-etching, is present in most of the samples. In some cases, two generation of calcite, with different Mn contents, seem to be present (Figure 7a).

Quartz can be an inherited and/or neoformed mineral. In the second case, SEM observatios indicate the frequent presence of idiomorphic quartz, as well as secondary quartz veinlets. Potassium feldspars and plagioclases are usually inherited from the host rocks, frequently showing well developed pit-etchings (Figure 7b).

Accessory minerals:

The accessory minerals have been classified according to their chemical composition, determined by SEM+EDX. Thus, elemental sulphur has been found at 451.42 m, Fe (Mn) oxyhydroxides are present in most of the samples (Figure 7c), while W oxides and Ti oxides are found in only some samples.

Sulphides, such as pyrite, chalcopyrite, galena, sphalerite, greenockite, pentlandite, bismutinite, molybdenite and complex sulphides with (Fe, Ni, Cu) and (Cu,W,Fe), have been detected, the first three being the most frequent (Figure 7d). Minor sulphoarsenides, such as arsenopyrite and Fe,Co, Ni sulphoarsenides, occur in some samples. Similarly, minor selenides (Bi, Pb) and telurides (Bi) are also present.

Some carbonates, like parisite, smithsonite and cerusite; sulphates, like celestite and Ca-sulphate with REE's, and Pb silicates are very rare.

Finally, inherited zircon, monazite and apatite have been detected in some samples, the first two being strongly altered.

Preliminary conclusions

The mineralogy detected in the fracture fillings studied is very complex, and a better information about the textural relationships among the neoformed minerals is necessary, in order to establish their relative sequence of crystallization and, consequently, the last mineral generation.

The clay minerals detected in the samples indicate very different physico-chemical conditions. Thus, kaolinite indicates acid conditions and very intensive leaching of cations, while palygorskite indicate alkaline conditions and very high SiO₂ and MgO activities.

The coexistence of sulphides and sulphates can be indicative either of a change of Eh conditions or of a range of Eh values in which both minerals are stable.

The preliminary mineralogical results obtained from fracture fillings of borehole R385 and R386, that are in agreement with the hydrogeochemical model of the site (Gimeno *et*

al., 1997), encourage to study fracture fillings of the Eastern granite, as well as in selected fractures of the new borehole R387. To complete this study, the bedrock, adjacent to fracture surface, will be studied using relevant physical, chemical and radiochemical methods, in collaboration with the Oviedo University. In this way, migration/retention processes through the rock matrix will be established, performing true diffusion profiles from the walls of the hydraulically active fractures to the adjacent rock. The joint interpretation of this data and those obtained from the groundwaters, sampled in the same fractures and in different redox conditions, will allow the geochemical modelling of the water-rock interaction processes that occur in the far field of the site. This model will be later used in the PA exercise.

Table 3. Semiquantitative mineralogical composition of the fracture fillings from borehole R385 and R386 obtained by XRD.

DEPTH	Q%	KFd%	Plg%	Cc%	T.Ph%	Chl%	
81.42*	3	2	2	64	29(Sm+Ill)	0	
65.65	5	0	0	6	89 (K+Sm+Ill?)	Tr	
67.12	14	3	3	8	66 (Sm**+Ill+K?)	6	
119.12	17	23	24	15	20 (Pk+Ill?)	0	H.A.Z.
175.55	30	5	8	10	16 (Sm+Pk?+Ill?)	30	
176.75	20	14	14	8	11 (Ill+Sm)	33	
219.70	11	0	0	86	Tr (Sm)	3	H.A.Z.
222.90	9	8	Tr	46	Tr (Sm)	37	H.A.Z.
406.20	6	0	3	86	Tr (K)	5	H.A.Z.
423.10	37	0	42	8	7 (Sm+Pk?+Ill?)	7	
451.42	4	Tr	2	36	58 (K**+Sm+Ill)	0	
460.40	4	6	2	61	27 (K**+Sm+Ill)	0	

*:From Borehole 386; Q:Quartz; KFd:K-Feldspar; Plg:Plagioclase; Cc:Calcite; T.Ph:Total Phyllosilicates; Ill:Illite; Sm:Smectite; K:Kaolinite; Chl:Chlorite
Pk:Palygorskite;**Major clay minerals; H.A.Z.:Hydraulically Active Zone.

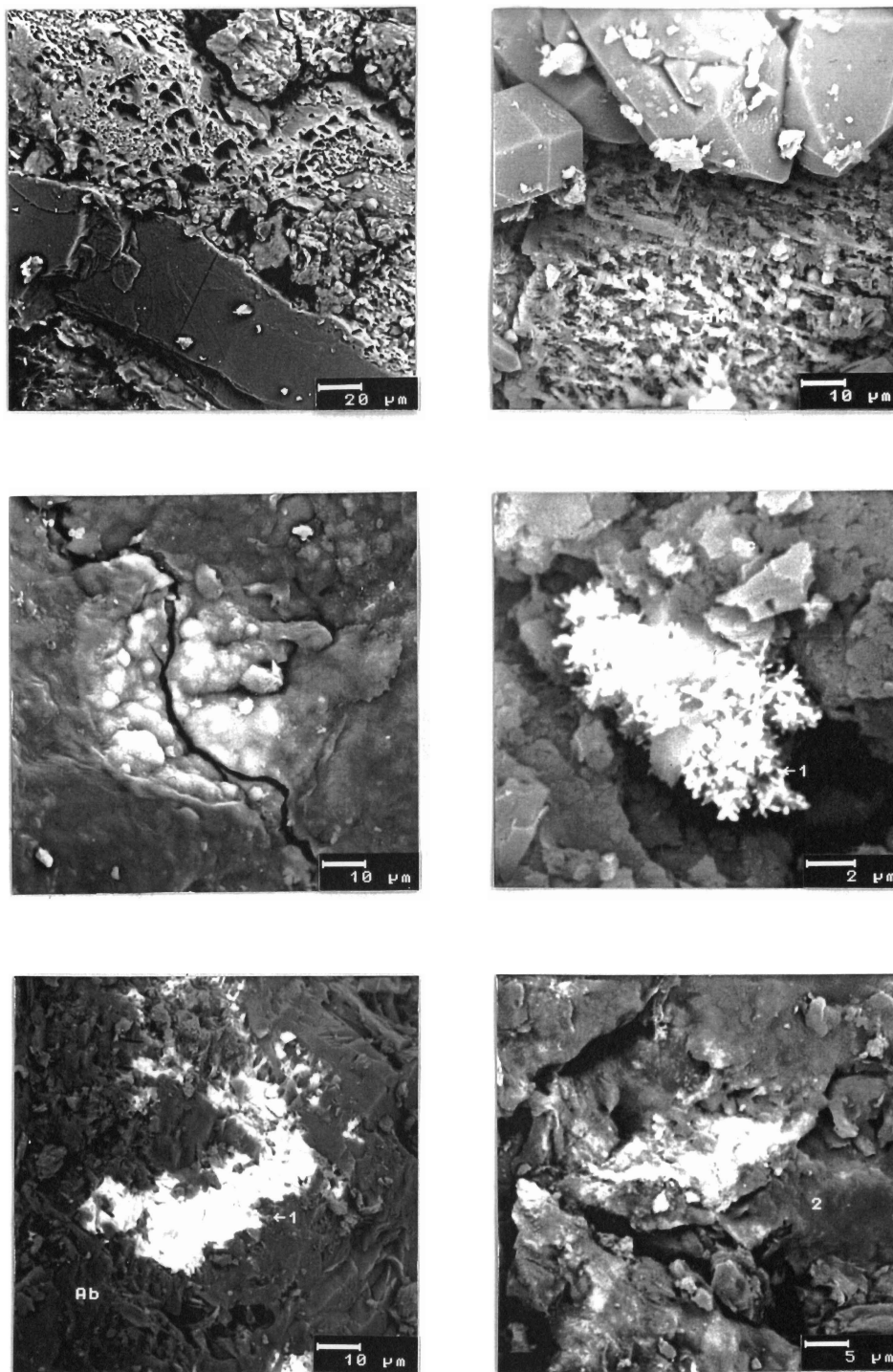


Figure 7. a) two generations of calcite, the first showing pit-etching and the second with veinlet morphology (sample depth 65.65 m), b) idiomorphic neoformed quartz (Q) on K-feldspar (Kfd) showing well developed pit-etching (sample depth 223 m), c) concretionary neoformed Fe(Mn) oxyhydroxides (sample depth 21.64 m), d) dendritic and neoformed (Cu, Fe, W) sulphide precipitated on neoformed calcite (sample depth 223 m), e) U-silicate (1) precipitated on albite (Ab), the latter showing pit-etching (sample depth 460.4 m), f) U-silicate (1) precipitated on neoformed smectite (2) (sample depth 119.12 m).

Task 2.3 Geochemical modelling of water-rock interaction

The main aim of the modelling study of Ciemat group (Gimeno *et al.*, 1998) was to establish the hydrogeochemical conceptual model of the Palmottu site; *ie.* the definition and interpretation of the water-rock interaction processes responsible for the geochemical evolution of groundwaters in order to predict their future behavior and influence in the migration of trace elements.

At first stage a statistical approach was used (multivariate analysis: MCA, PCA and cluster analysis, performed with the code SPSS for windows 3.1) and then the study focused in the reaction path simulations (direct simulation with the codes PHREEQE and EQ3/6) and mass balance modeling (inverse model with NETPATH) in order to reproduce the different geochemical processes. Moreover, the isotopic data of the groundwaters ($\delta^{18}\text{O}$, $\delta^2\text{H}$, $\delta^{13}\text{C}$ and ^{14}C) have been used to support some of the obtained results.

From the statistical analysis it can be seen that all the water samples are placed in a factorial plane forming a triangle whose vertexes represent the end members of different waters: (a) Ca-HCO₃ waters (from the more diluted ones, recharge waters, to the more evolved); (b) Na-SO₄ waters; and (c) Na-Cl waters (Figure 8). In general terms there is an increase in the TDS in the same direction as the increase in depth.

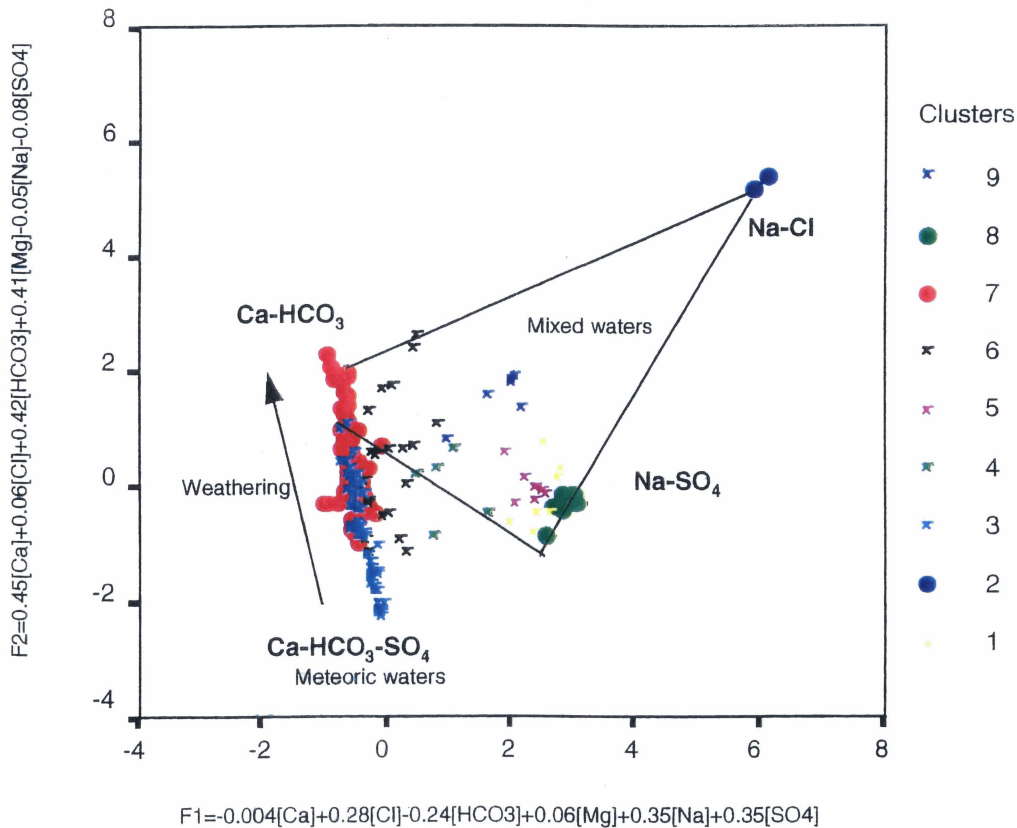


Figure 8. Plot of the different water groups showing the site-specific three groundwater end members, and the recharge water (meteoric) in a factorial plane. The typical weathering process line between the recharge water and the Ca-HCO₃ end member is shown. Cluster 2 = Na-Cl water, Cluster 3 = dilute, shallow, young groundwater, Cluster 7 = groundwater resulting from the weathering processes, Cluster 8 = Na-SO₄ water, Clusters 1, 4, 5, 6 and 9 indicate different combinations between clusters 2, 7 and 8.

The results obtained from the geochemical modeling together with the statistical results, yield to the hypothesis of the existence of two dominant hydrogeochemical processes: (1) the typical weathering process of a crystalline basement, which, when dominant, seems to be the responsible for the waters in the line between the recharge water and the Ca-HCO₃ end member; and (2) the mixture of the previous waters with other deeper, older and more saline ones; this would be the dominant process in the chemical characterization of waters inside the triangle formed by the Ca-HCO₃, Na-SO₄ and Na-Cl end members (Figure 8).

The weathering process is characterized by the following predominant reactions: calcite and Ca-Na feldspar dissolution, CO₂ uptake from the atmosphere, biotite alteration, pyrite (or other sulfides) oxidation and precipitation of silica and kaolinite. And the result is the evolution from the more diluted Ca-HCO₃ waters to the more typical Ca-HCO₃ end member.

Mixed water types have different proportions of the three end members and they are also affected by reactions. Dissolution of aluminosilicates, K-feldspar or even gypsum, and precipitation of calcite, silica and ionic exchange, are the predominant reactions superimposed to the mixing process.

Those results fit very well with the main conclusions that can be extracted from the isotopic analysis; and a coherent conceptual hydrogeochemical model can be drawn.

WP 3: Redox processes (QuantiSci)

Objectives and main activities

WP 3 is intended to be a smooth transition from the interpretative description of the Hydrochemical system at Palmottu to the more predictive requirements of the performance assessment applications to be integrated in WP 5. The study of the redox-related issues will be organized through the following tasks:

- Task 3.1 Geochemical behaviour of U, Th and selected trace elements,
- Task 3.2 Experimental and modelling study of redox capacities and intensities,
- Task 3.3 Development and testing of PA models and submodels.

Mineralogical results

Microanalytical work on alteration products of primary uraninite is presently proceeding (Peréz del Villar *et al.*, 1998; Hansén and Ruskeeniemi, 1998). This study also incorporates analytical work on uranium-rich fracture-infilling material.

The secondary U minerals have been found in the most superficial zones of the borehole R386 (21.64 m), in the hydraulically-active zones of R385 (91.72 m and 119.12 m) and in the fractures close to the Eastern granite (R385/451.42 m and 460.40 m). In the first one, U(Pb) oxides and U(Pb) silicates are present; In the second zone, at 91.72 m depth, only U(Pb,Bi) oxides have been found, while at 119.12 m only U silicates have been observed. In the third zone only U silicates have been detected (Figure 7e and 7f).

These oxides can be explained considering that in these zones the reducing conditions have remained since their formation, at least as microenvironments. In the case that Pb and Bi were of radiogenic origin, these U oxides should be very old. In any case, the presence of secondary U minerals in fracture fillings indicates that this element migrated far from the source term (orthomagmatic uraninite in the granites and pegmatites), and was mainly retained by a precipitation process.

Experimental and modelling results on redox capacities and intensities

Dissolution experiments were performed (Cera *et al.*, 1998) by using natural uraninite samples from Palmottu, in order to test their redox behaviour in the system. The final aim of these experiments were to study the reductive capacity of the samples. Three dissolution experiments were performed in batch conditions and room temperature during a period of approximately 100 days/sample.

The first experiment (in oxidizing conditions) showed a trend of initial uranium increase until reaching a steady-state. The steady state U concentrations agree with the calculated solubility of schoepite. In the second experiment, also performed under oxidizing conditions, the uranium concentration in solution initially increased to a maximum value, but was subsequently decreased and finally reached a steady state value. The steady state uranium concentrations fairly well agree with U concentrations calculated to be in equilibrium with soddyite.

As the phase boundary of soddyite to schoepite transformation is quite close to the phase boundary of quartz and dissolved H_4SiO_4 , small changes on the dissolved H_4SiO_4 concentration in solution could lead to fluctuations across the soddyite/schoepite boundary. This hypothesis remains open for further investigations.

The third experiment was performed under anoxic conditions, and the trend of the evolution of the uranium concentration in solution with time showed an initial release of uranium to reach a steady-state level. The measured U concentrations in solution at the steady state corresponded to uraninite solubility under anoxic conditions. In order to study the reductive capacity of the uraninite sample, two sequential U(VI) inputs were given to this system. The general and reproducible trend of the experimental data indicated a rapid decrease of the induced U concentration to reach the same levels as the ones previously attained in equilibrium with uraninite.

This intriguing behaviour indicates an efficient and reproducible scavenging effect of the U(VI), evidently by the uraninite surface. Two potential mechanisms could be responsible of this phenomenon: (1) sorption of U(VI) at uraninite surface, (2) sorption of U(VI) at uraninite surface followed by the reduction of U(VI) to U(IV). Future work will be devoted to explore the hypothesis proposed for the U(VI) scavenging by uraninite; a combination of solution chemical and surface spectroscopic techniques will be used.

WP 4 Migration of radionuclides (VTT Energy)

Objectives and main activities

WP 4 integrates migration-related information from previous WP's. The basic approach is to apply already existing experimental techniques, modelling concepts and models to assess their usefulness in studying the observed distribution of radionuclides. The work is divided into process-oriented parts (*e.g.* sorption, matrix diffusion) that will be coupled with field data of groundwater flow (from WP 1) into a quantitative migration simulation. The use of PA models in migration simulation will create a direct link to the PA related conclusions of WP 5. Technically WP 4 is divided into four major tasks:

- Task 4.1 The role of sorption processes,
- Task 4.2 The role of matrix diffusion
- Task 4.3 The role of colloids and microbes,
- Task 4.4 Migration modelling.

Detailed planning and initial work

Task 4.1: A planning meeting for Task 4.1 was held (BRGM, Orleans, France) with the purpose to discuss and focus the plans of individual teams. The aims of Task 4.1 were also refined: the ultimate aim is quantitative understanding of sorption mechanisms at Palmottu. The technical aim is to demonstrate that sorption processes can be understood theoretically, and described numerically with concepts that can be based on established experimental methods.

The sorption work conducted by BRGM team during 1997, allowed to test some aspects of the experimental protocol of batch experiments foreseen on undisturbed fracture surfaces. The tests were firstly related to the search of an inert chemical material for the protection of the rock matrix from the uranium sorption during the experiments. The fracture surface area must be the unique part of the sample in contact with the uranium solution. Three products were tested (two PTFE products and one silicon product). The silicon product ("PROSIL") has given the best results. Some additional tests are in progress to improve the quality of the silicon coating on the core sample. The planning of the sorption intercalibration exercise has been completed, rock and water samples for the calibration of U analyses have been distributed, and measurements are on the way.

Task 4.2: A planning meeting for Task 4.2 was held (GTK, Espoo, Finland). The overall aim of Task 4.2 is to demonstrate that matrix diffusion can be observed, quantified and modelled with the experimental and theoretical tools available. The technical aim of the task is to test the classical matrix diffusion model against *in situ* observations made at Palmottu. This aim has relevance with respect to the integrated migration modelling task (4.4) and also to PA aspects (WP 5). The modelling approach consists of parallel use of matrix diffusion and uranium series disequilibria (USD) modelling that provides independent dating of the accumulated uranium system. Besides the concentration profile in the rock matrix being studied by diffusion model, USD dating can be done for fracture surface samples.

Task 4.3: The plans for Task 4.3 will be refined during spring 1998.

Task 4.4: The integrated migration modelling work will be focused at the Eastern flow system. The retardation mechanisms from mechanistic studies (sorption and matrix diffusion) and groundwater flow information from WP 1 and 2 will be applied. In addition, a quantitative understanding of the release rates of U from the rock into the flow system will be needed. This will be based on theoretical (geochemical) and experimental (extraction) considerations. For selecting samples for this Task, the following supporting information is available: structure model of the bedrock, groundwater flow modelling, groundwater chemistry, comparison of groundwater flow modelling and groundwater chemistry, fracture mineralogy, video logging of fracture characteristics *in situ*, and the results of the tracer test conducted around the bore holes of the Eastern flow system. The final criteria to select the proper fractures, is to apply phase-selective extractions to locate appropriate fracture-groundwater pairs.

Two modelling approaches will be used in this Task, namely a simplified PA-oriented one (VTT Energy), and a more detailed coupled modelling approach (BRGM and ENRESA team). Geochemical evolution of the water-rock system will be utilized in the coupled modelling approach together with the groundwater flow field. This parallel use of two different modelling approaches may enable us to draw more detailed quantitative conclusions. The input data for all the models need to be based on experimental data on samples from the actual flow route.

The numerical model will be constructed using the ALLAN and NEPTUNIX modelling aid programming environment, an approach used for treating these problems over the last

5 years by the BRGM. Each developed model configuration will initially be tested in a "closed system" configuration in order to determine whether model-predicted ground water composition is in equilibrium with the ground water and mineral assemblages observed in the Palmottu formation. The solid phase will be composed of unlimited masses of mineral phases identified at specific Palmottu formation observation points. The results of this test will help in evaluating both the proposed mass action laws (stoichiometry, K) and whether ground water residence time is sufficiently long for the system to reach local equilibrium for all kinetically hindered chemical reactions. If successful, the model would then be tested in an "open system" configuration wherein the reactor steady state groundwater composition and mineral phase dissolution and precipitation rates would be calculated for different solution flow rates and input compositions. These tests should allow evaluation of the reasonableness of proposed reaction kinetic constants vis à vis measured groundwater composition and flow rates, and solid phases.

These single reactor tests, once successful, would then be followed by connection of several reactors in series, each corresponding to a specific Palmottu formation observation point along a presumed hydrodynamic, mass-transfer pathway. Each reactor would contain an initial solid phase made up of corresponding observed formation minerals; the initial reactor would be fed by water having the same composition as that circulating at the most upgradient formation point.

WP 5: Conclusion relevant to repository performance assessment PA (Kemakta/SKB)

Objectives and main activities

In WP 5, issues and phenomena with a potential impact on the safety and performance of a repository for radioactive waste will be evaluated and discussed. This will provide possibilities to describe and document scenarios for site evaluation as well as to test models used in PA of repositories, *e.g.* flow models, models of different transport phenomena, chemical speciation models and models describing different types of couplings. The intention of WP 5 is to sum up the information and concepts developed within other WP's with respect to PA-aspects.

A complex natural system like the Palmottu site can provide information relevant to study of a large multitude of phenomena. Processes and phenomena which are of obvious concern in PA and well suited for Phase II studies include: scenario development (*e.g.* effects of different paleo-events), process-oriented studies (*e.g.* redox control in groundwater-rock systems, propagation of redox fronts, matrix diffusion and sorption, uranium sinks, effects of microbes), chemical evolution of infiltrating water, mobilization of uranium in the mineralized zone (*e.g.* dissolution of uraninite and formation of secondary uranium minerals and co-precipitation) and processes interfacing with the biosphere.

The study of these issues are organized within three major tasks:

- Task 5.1 Scenario development,
- Task 5.2 Near field,
- Task 5.3 Far field.

For focusing the work and plans, a workshop was arranged in March 1998 with the purpose to identify the PA issues for which there is a reasonable chance that Palmottu data could give a contribution to the assessment credibility. The target of the workshop was to

document the main contents for the final technical document of WP 5. To ensure this aspect to be fulfilled, the workshop was planned to be implemented as follows:

Discussions on the contributions to scenario development

- main effects of glaciations

Evaluation on the state of the process identification

- redox control
- propagation of redox fronts
- uranium dissolution
- the role of microbes and colloids
- sorption and matrix diffusion
- uranium sources and sinks
- solubility of uranium

Discussion and planning for model testing

- groundwater evolution modeling
- coupled transport
- transport of other tracers (*e.g.* REE, Th)
- transport in the Eastern flow system
- blind predictive geochemical modeling
- biosphere interface modeling
- sorption and matrix diffusion models
- linking hydrochemistry and hydrogeology in interpretation of the flow regime

Discussion and planning of data consistency and building PA database

- laboratory intercomparison on
 - * K_d-measurements
 - * groundwater analyses
 - * sequential extractions
 - * redox measurements

IV PROJECT DOCUMENTS

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