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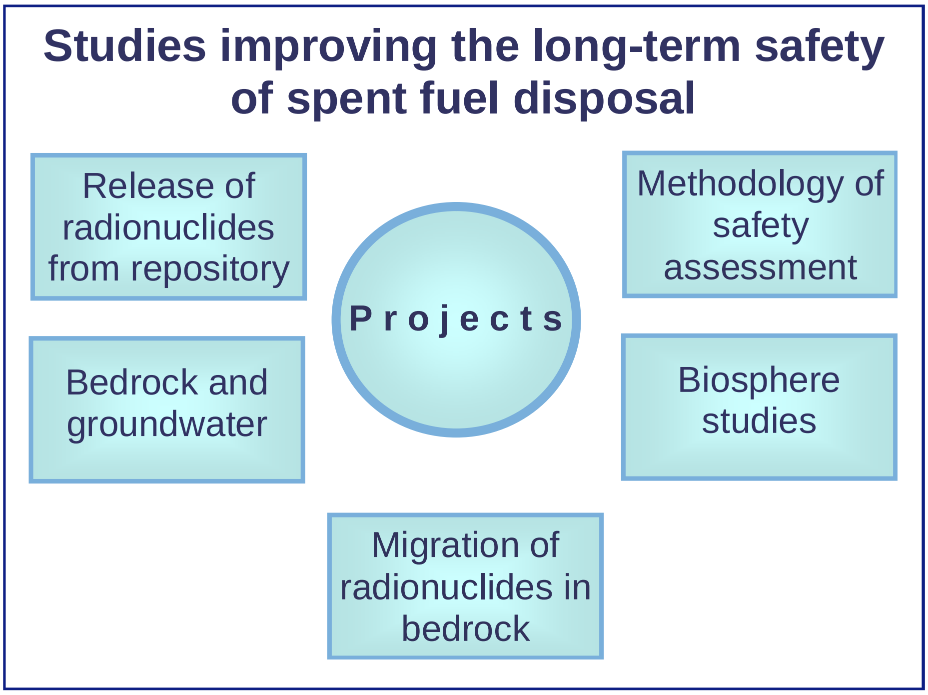
VTT RESEARCH NOTES 2337



The Finnish Research Programme on Nuclear Waste Management (KYT) 2002- 2005

Final Report

VTT TIEDOTTEITA – RESEARCH NOTES 2337

The Finnish Research Programme on Nuclear Waste Management (KYT) 2002–2005

Final Report

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VTT

ISBN 951–38–6786–2 (soft back ed.)
ISSN 1235–0605 (soft back ed.)

ISBN 951–38–6787–0 (URL: http://www.vtt.fi/publications/index.jsp)
ISSN 1455–0865 (URL: http://www.vtt.fi/publications/index.jsp)

JULKAISIJA – UTGIVARE – PUBLISHER

VTT, Vuorimiehentie 3, PL 1000, 02044 VTT
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Technical editing Leena Ukskoski

Valopaino Oy, Helsinki 2006

The Finnish Research Programme on Nuclear Waste Management (KYT) 2002–2005. Final Report. Kari Rasilainen (ed.). Espoo 2006. VTT Tiedotteita – Research Notes 2337. 246 p. + app. 45 p.

Keywords nuclear waste management, Finland, repositories, safety, radionuclides, migration, bedrock, ground water, spent fuel, reactor wastes, environmental protection

Abstract

The producers of nuclear energy are responsible for the safe handling, management, and disposal of their wastes, as well as for the costs arising, according to the Finnish nuclear energy legislation. National authorities supervise the nuclear waste management and issue regulations for this purpose. The research needs of the authorities have traditionally been supported by public research programmes. The four-year KYT programme (2002–2005) followed the earlier public nuclear waste research programmes that started as long ago as in 1989.

The main research area in KYT was in studies promoting the assessment of the long-term safety of the geological disposal of spent nuclear fuel. This wide area was divided into five sub-areas: (1) the methodology of safety assessment; (2) the release of radionuclides from the repository; (3) bedrock and groundwater; (4) radionuclide transport in bedrock, and (5) biosphere studies. The second research area covered strategic alternatives in spent nuclear fuel management. Studies in this area were focused on advanced fuel cycles, including e.g. partitioning and transmutation (P&T). In addition, there were limited studies on reactor wastes resulting from the operation of nuclear power plants.

KYT promoted Finnish know-how in nuclear waste management for the use of national authorities. It acted as a national forum for discussion and co-operation between the authorities, the nuclear industry, and the research community. Furthermore, it trained new experts in the field, as the most experienced nuclear waste specialists are approaching their retirement.

The next major milestone in the Finnish nuclear waste management programme is the construction licence application for the disposal facility for spent fuel, due around 2012. The licensing process increases the need for high-quality research on the technical safety of the facility. The successor to the KYT programme, called KYT2010, will be carried out from 2006 to 2010.

Foreword

This is the final report of the Finnish Research Programme on Nuclear Waste Management (KYT). The KYT programme, covering the years 2002–2005, was originally planned as a public research programme funded by individual actors in Finnish nuclear waste management. The Nuclear Energy Act was amended at the beginning of 2004 to ensure the level of funding. In the amended Nuclear Energy Act the research needs of the Finnish authorities were emphasised. After the beginning of 2004 the funding for KYT came from the dedicated Nuclear Waste Research Fund established within the State Nuclear Waste Management Fund. As a research programme, KYT is a direct continuation of the previous public nuclear waste research programme, JYT (first stage 1989–1993, second stage 1994–1996, third stage 1997–2001).

The co-ordinator of the KYT programme was Dr. Kari Rasilainen from the Technical Research Centre of Finland (VTT). The Steering Group of the programme included representatives from the authorities, as well from the nuclear power companies. The authorities were represented by the Ministry of Trade and Industry (KTM) and the Radiation and Nuclear Safety Authority (STUK). The nuclear power companies were represented by Teollisuuden Voima Oy, Fortum Power and Heat Oy, and Posiva Oy. Furthermore, the National Technology Agency (Tekes) was represented in the group. Ms. Anne Väätäinen (KTM) acted as the chairperson of the Steering Group during 2002–2003 and Mr. Esko Ruokola (STUK) during 2004–2005.

The execution of the research programme was based on the overall Framework Programme outlined by the Steering Group. The Framework Programme was specified in the Annual Research Plans based on the individual plans of the actual project proposals. Each year there was a public call for project proposals, from the yield of which the Steering Group selected the proposals to be funded.

The programme consisted of several projects that were carried out by various research institutes of VTT, the Geological Survey of Finland (GTK), Helsinki University of Technology (TKK), the University of Helsinki (HYRL), and the University of Jyväskylä (JYFL). In addition, STUK and Fracom Oy participated in the research.

This report has been edited by Kari Rasilainen on the basis of contributions from the different research projects. The most interdisciplinary contributions were co-ordinated by the respective project managers. The writers of individual subsections are mentioned in the text and the scientists who contributed to the actual results are mentioned in Annex A for each project. The language of the manuscript was checked by Ruth Vilmi Online Education Ltd.

Contents

1. Introduction[1]

1.1 Nuclear waste management policy in Finland

SCHEDULE

The Nuclear Energy Act and Degree provide a clear framework for nuclear waste management in Finland. According to the legislation, the producers of nuclear waste are responsible for all measures needed for the safe management of the waste and for the costs that arise. The authorities supervise the management of nuclear waste and issue regulations for this purpose. The Nuclear Energy Act was amended at the end of 1994, and it now[2] stipulates that all radioactive waste produced in Finland must be handled and disposed of in Finland.

The objectives and timetables for nuclear waste management in Finland, as well as for the related research and planning, were defined in a policy decision issued by the Government in 1983. Subsequently, the authorities have made decisions on the more detailed principles and requirements that power companies have to comply with. The Finnish spent fuel management programme has so far kept to its original schedule, at least partly as a result of the clearly-defined division of responsibilities between the energy-producing industry and the authorities.

There are currently two operating geological repositories for low- and intermediate-level reactor waste in Finland. The reactor waste repositories are located at the two nuclear power plant (NPP) sites, one at Loviisa and one at Olkiluoto. At both repositories there are plans for extensions to take care of the decommissioning waste of the respective NPPs in the future.

The Parliament ratified the Government's favourable Decision in Principle (DiP) to establish a spent fuel disposal facility in the Olkiluoto area in the Eurajoki municipality on May 18, 2001. As a whole, the DiP process proceeded rapidly, as Posiva submitted the application to the Government on May 26, 1999. The application was backed by, inter alia, a performance assessment (Vieno & Nordman 1999) and an extensive Environmental Impact Assessment (EIA) (Posiva 1999). Posiva's EIA covered four candidate municipalities (Kuhmo, Loviisa, Eurajoki, Äänekoski) that were included in Posiva’s detailed site investigations preceding the site selection.

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[1] By Kari Rasilainen, VTT.

[2] The reactors of the Loviisa nuclear power plant were acquired from the former USSR. The owner of Loviisa NPP, Imatran Voima Oy, initially made contractual arrangements for the whole fuel cycle service with the USSR, including the return of spent fuel. Spent fuel shipments to Russia were halted after 1996.

In conjunction with the discussion of the DiP for the fifth nuclear power reactor (Olkiluoto 3), on May 24, 2002 the Parliament ratified an amendment to the DiP for the final disposal concerning the nuclear waste from the new reactor.

In line with the DiP, the excavation of the ONKALO underground rock characterisation facility at Olkiluoto was started in summer 2004. ONKALO will not only be an underground rock laboratory but it is meant eventually to be part of the underground repository at Olkiluoto.

The current timetable of Finnish spent nuclear fuel management is shown in Figure 1.

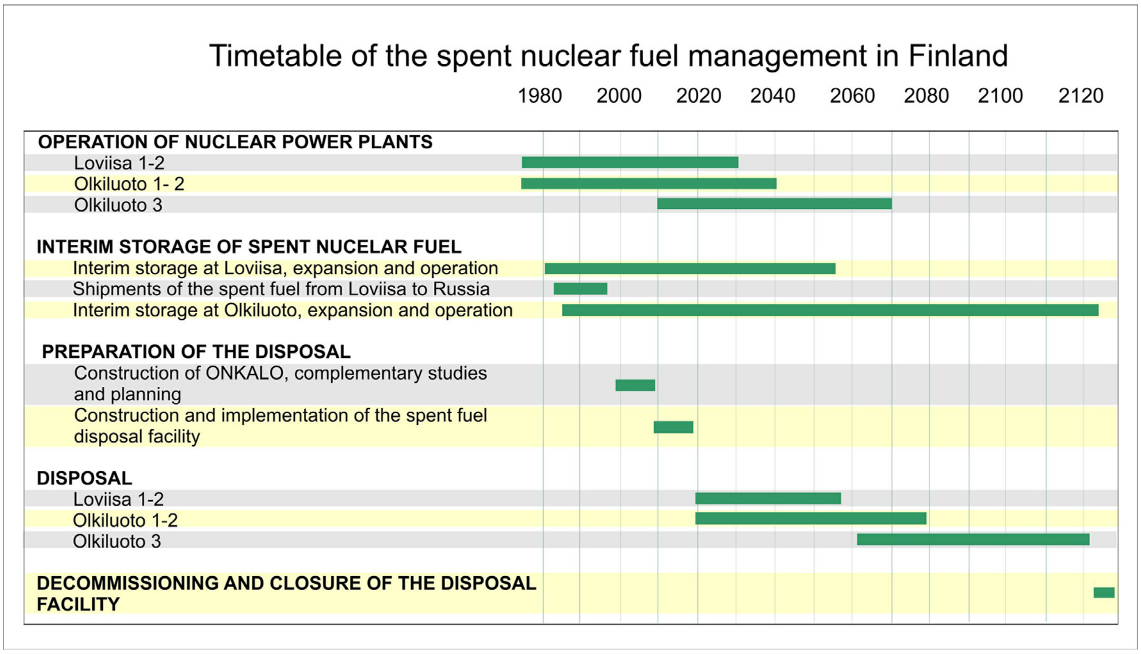

Figure 1. Overall schedule of spent nuclear fuel management in Finland. The spent fuel is assumed to come from all currently existing reactors and from Olkiluoto 3, which is under construction.

The next major milestone in the planning of nuclear waste management is the preparation for the construction licence application for the spent fuel disposal facility and the handling of the application by the authorities around 2012. The application for the operating licence is planned to submitted around 2020.

ACTORS AND FUNDING

There are three main actors in Finnish nuclear waste management, each with different responsibilities. In order to fulfil their responsibility to take care of their spent nuclear fuel in Finland under the amended Nuclear Energy Act, the nuclear power companies[3] Teollisuuden Voima Oy and Fortum Oyj (previously Imatran Voima) formed a jointly-owned company, Posiva. Posiva was founded in 1996, and its mission is to plan and implement the disposal of spent nuclear fuel generated in Finland.

Overall leadership and control in nuclear energy matters in Finland is the responsibility of the Ministry of Trade and Industry (KTM). The ministry prepares the relevant national legislation and international agreements, as well as monitoring compliance with them. The Radiation and Nuclear Safety Authority (STUK) is responsible for the supervision of nuclear and radiation safety. The STUK prepares the national safety requirements for spent fuel disposal.

To make sure that the remaining activities in the national nuclear waste management programme can be implemented under all conditions, the power companies are obliged to set aside money in the State Nuclear Waste Management Fund. The Fund is governed by the basic principle that it can, at all times, cover all the future costs of all currently existing nuclear waste.

The power companies must present cost estimates for the future management of their nuclear wastes to the KTM annually. The cost estimates are based on the power companies' latest technical plans, and they also include the decommissioning of nuclear power plants. The Fund is administered by the KTM, and, on the basis of the cost estimates and on expert reviews of these, the KTM decides, annually, the contributions to be made to the Fund. For uncovered costs, the power companies must furnish securities. At the end of the year 2004 the money that had been paid in covered 100% of the liability.

1.2 Context of the KYT Programme

KYT follows the tradition of public research programmes on nuclear waste management, as these are considered to be a practical way to pool the necessary expertise. The publicly financed research on nuclear waste management in Finland was initiated by the Advisory Board of Atomic Energy as long ago as the early 1970s. Co-ordinated, publicly administrated research programmes on nuclear waste management have been carried out in Finland since 1989. The first phase of the research programme (JYT) was conducted in 1989–1993 (Vuori 1990, 1991, 1993), the second phase (JYT2) in 1994–1996 (Vuori 1997), and the third phase, JYT2001, in 1997–2001 (Rasilainen 2002).

[3] Both companies own two nuclear reactors, Teollisuuden Voima at Eurajoki and Fortum at Loviisa.

The KYT programme was started in 2002 as programme of independent sponsors. The amounts of funding provided by individual sponsors are shown in Annex D. The original idea of the research programme was that the research would benefit all funding organisations involved. All issues linked to licensing were excluded from the very beginning, however, as the research of a public research programme must be neutral.

The Nuclear Energy Act was amended from the beginning of 2004. The general aim of the amendment was to ensure the level of funding for the public research programmes on nuclear safety. For the KYT programme the amendment meant that a dedicated Nuclear Waste Research Fund was established in the State Nuclear Waste Management Fund, which is controlled by the KTM. The capital for the research fund is collected from the producers of nuclear waste. Every producer will have to contribute 0.08% of its respective assessed nuclear waste management liability (confirmed annually by the State Nuclear Waste Management Fund). With current liabilities the annual funding level of KYT is around 1 M€.

In the amended Nuclear Energy Act the research needs of the authorities were emphasised. This appears to be a somewhat different focus as compared to the original one, but all licensing issues were excluded from the new KYT too. As the practical aim has always been to fund only studies relevant to the national nuclear waste management programme, the implementers can still benefit from the results.

1.3 Structure of the KYT Programme

The research in the KYT programme was divided into two main categories: (1) strategic studies of nuclear waste management, and (2) studies improving the long-term safety of geological spent fuel disposal. Support for the performance assessment of long-term safety was emphasised in the framework programme (Rasilainen 2003). Of the two categories above, the latter one was dominant both in terms of funding and in the number of research projects.

STRATEGIC STUDIES

The research evaluating strategic alternatives of nuclear waste management supports the robust implementation of Finnish nuclear waste management. Strategic studies aim to ensure a knowledge base of our own that is sufficiently broad also to implement nuclear waste management in changing conditions; see Figure 2. Although Finland is proceeding purposefully towards the geological disposal of spent nuclear fuel, we must be aware of potential alternatives in case the present plans do not come to fruition.

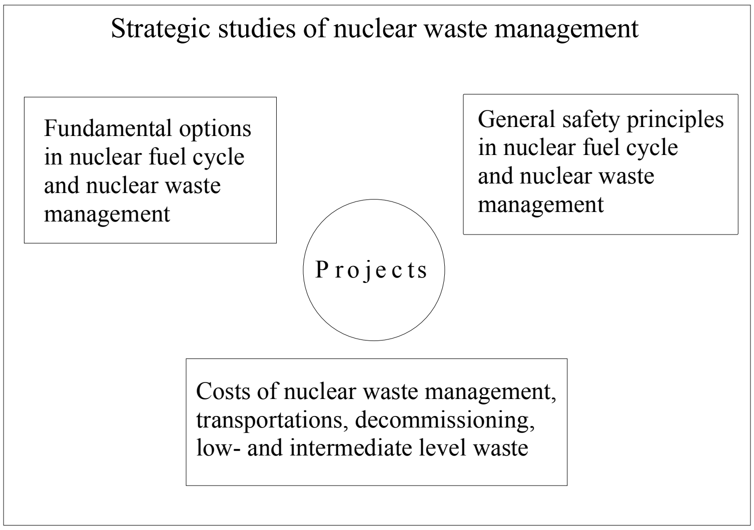

Figure 2. Strategic studies in the KYT programme.

Issues belonging to the basic policy definitions of the nuclear fuel cycle and waste management are, for example, reviews of future alternatives, such as partitioning and transmutation (P&T) or long-term storage. There is a need in Finland to follow international developments in these research fields. In addition, possible participation in international joint ventures presumes sufficient initial know-how.

General safety issues connected to nuclear waste management and the fuel cycle include the key principles of safety and radiation protection, the retrievability requirement of disposal, human intrusion into the repository, and the EIA process. Additionally, technical reviews associated with potential post-closure monitoring can be included in the general safety issues.

The disposal of spent fuel is by far the largest research area of the KYT programme. For practical reasons, the strategic studies in the KYT programme include all technical, economic, and natural science research that does not directly serve the disposal of spent fuel. These are, for example, cost reviews and estimates of nuclear waste management, as well as reviews related to the management of reactor waste, transport of nuclear waste, and decommissioning of nuclear power plants. The follow-up of international development work (EU, IAEA, and NEA) around these topics is justified.

LONG-TERM SAFETY OF SPENT FUEL DISPOSAL

A central part of the research programme is investigations that contribute to the long-term safety of the geological disposal of spent nuclear fuel, and especially to the improvement of capabilities to assess long-term safety; see Figure 3. The need for research is analysed from the performance assessment point of view so that the limited resources can be allocated as reasonably as possible.

The aim of the KYT programme was not to make a complete performance assessment of geological disposal, independent of Posiva. Rather, the aim was to concentrate on investigations that can improve the potential of making analyses and to diminish uncertainties inherent in the subsystems, and that can be accomplished using available resources. The intention was that the KYT programme should include investigations with relevance to nuclear waste management. Such studies include the development and testing of experimental and theoretical methods and tools.

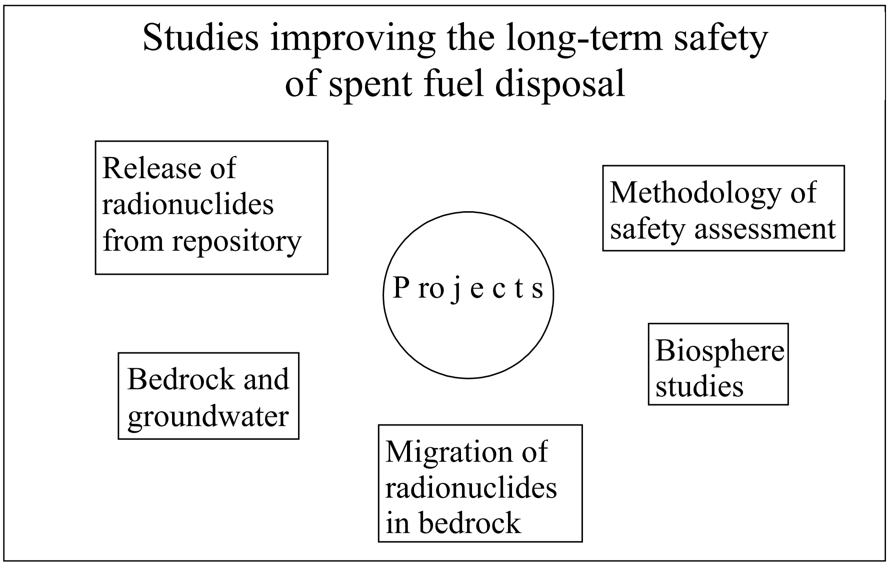

Figure 3. Studies improving the long-term safety of geological spent fuel disposal in the KYT programme.

The methodology of the safety assessment of the disposal of spent nuclear fuel includes issues such as scenario analysis and uncertainty analysis. The systematisation of the derivation of scenarios makes it easier to communicate with people outside the nuclear waste management community and to ensure that all scenarios relevant to performance assessment are considered. Supplementary safety arguments have been considered by international expert groups. These would be less mathematical in nature and thus more easily accessible to non-professionals. They can be extracted from e.g. natural analogues and different safety indicator projects.

The release of radionuclides from a repository comprises the disposal facility itself, its near field, and all engineered barriers. The detailed technical planning of the engineered barrier system (EBS) and the manufacturing techniques fall beyond the scope of the KYT programme, however. The development of a research methodology for the near field conditions and the coupled processes is, however, of the utmost importance. Copper canister studies are included as the canister is the most important single engineered barrier of the disposal concept. The coupled THM (thermal-hydrological-mechanical) behaviour of compacted bentonite is not yet known in detail.

Bedrock and groundwater are important research topics because Finnish nuclear waste management is based on geological disposal. The Olkiluoto-specific investigations are unambiguously Posiva's responsibility, but the development and testing of methods to study the internal structure and heterogeneity were an important part of KYT. The coupled thermal-hydrological-mechanical (THM) behaviour of the near field of the repository, integration methodology of the geochemical interpretation of groundwater and the results from groundwater flow modelling, and paleohydrogeological methodological research were included in KYT.

The transport of solutes and radionuclides in the bedrock depends on the groundwater flow field and chemistry. Therefore, from the process and data point of view the transport studies have a tight connection with all the previously-mentioned research areas. Important issues include the migration modelling methodology, the uncertainties related to inevitably imperfect site observations, and changing conditions resulting from long time periods.

Biosphere modelling is a procedure to convert radionuclide release rates (Bq/a) to dose rates (Sv/a). All Olkiluoto-related biosphere studies are naturally the responsibility of Posiva, but methodological studies have been pursued in KYT. In recent years, international expert groups have discussed the radiation protection of life forms other than human.

1.4 Structure of the Final Report

In the report, a brief review is provided of the most important results obtained by the KYT programme during 2002–2005. The space available in the report does not permit us to go deeply into detail and therefore the interested reader is advised to consult the original publications.

The results are grouped into four main chapters, in line with the overall structure of the research programme. The studies which contribute to the long-term safety of geological disposal are described in Chapter 2; these studies formed the central emphasis of the research programme. The studies dealing with strategic alternatives in spent fuel management are described in Chapter 3. The reactor waste studies performed within the KYT programme are described in Chapter 4. In Chapter 5 a summary of communication between the research programme and domestic and international interest groups is provided.

Each research project is described formally in Annex A. A list of the most important publications is given in Annex B. The organisation of the research programme is described in Annex C. The funding of KYT is shown in Annex D. Finally, the evolution of the emphasis of the research programme is shown in Annex E.

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2. Long-term safety of spent fuel disposal

The long-term safety of the geological disposal of spent nuclear fuel formed the main emphasis of the research programme. In the following sections the safety-related studies are summarised briefly. The extremely wide research area is divided into studies on the methodology of safety assessment, on the release of radionuclides from the repository, on bedrock and groundwater, on radionuclide transport in bedrock, and on biosphere-related issues.

2.1 Methodology of safety assessment

The safety assessment methodology-related studies consist of three basic components. KYT participated in the international DECOVALEX projects in an effort to increase understanding of coupled processes in repository-bedrock systems. The International Atomic Energy Agency (IAEA) organised a co-ordinated research project of natural safety indicators in which KYT participated in order to increase its expertise in this safety-relevant topic. Uranium migration in granitic rock was studied in international collaboration in order to obtain experience in a real modelling case, with the aim of increasing Finnish expertise in the area of reactive transport modelling. The last component was a separate extension of the IAEA project.

2.1.1 DECOVALEX studies in KYT programme[4]

INTRODUCTION

The international DECOVALEX project studied coupled Thermo-Hydro-Mechanical processes in a nuclear repository environment. The understanding of coupled processes is essential in the assessment of the performance of the disposal system.

Thermal effects arise mainly from spent fuel. Groundwater is found in fractured media. This means that hydraulic, i.e. groundwater effects should be taken into account. The repository will be constructed in a more or less fractured and non-homogeneous geological medium, and thus mechanical effects should be incorporated in the analysis. What is more, different temporal and spatial scales should be dealt with.

[4] By Esko Eloranta, STUK.

The name 'DECOVALEX' is an acronym, and stands for 'an international co-operative project for the DEvelopment of COupled models and their VALidation against EXperiments in nuclear waste isolation'. The project was managed by SKI (Statens Kärnkraftinspektion, Sweden). During the KYT period (2002–2005) two phases of DECOVALEX projects went on: DECOVALEX III, which was active in the years 1999–2003, and DECOVALEX THMC, ongoing since 2004.

The organisational structure of the project was the same as in the previous DECOVALEX phases from 1991. SKI from Sweden was the managing participant. The Royal Institute of Technology (Stockholm) acted as a secretariat, and the funding organisations formed the Steering Committee of the project. The chairman of the Steering Committee was Dr. Chin-Fu Tsang from the Lawrence Berkeley National Laboratory, USA. Funding organisations also supported and supervised their research teams. There were more than ten international research teams dealing with the specified modelling tasks.

GENERAL OUTLINE OF THE NATIONAL WORK

The main interest in the project was different ways to model the coupled Thermo-Hydro-Mechanical (T-H-M) phenomena occurring in a spent nuclear fuel repository. These processes are especially important in consideration of radionuclide release and transport from the repository. The main emphasis was on fractured rocks and buffer materials and the role of Performance Assessment (PA). International co-operation also provided exchanges of laboratory and field data for validation purposes and prepared statements on coupled T-H-M issues for performance assessment.

In the DECOVALEX III project Finnish interests were concentrated on three issues: (1) the modelling of bentonite behaviour; (2) the upscaling and homogenisation of the hydro-mechanical properties of fractured rock, and (3) the modelling of the formation of permafrost and its influences on the repository. The modelling of the bentonite buffer was studied at the Institute of Mathematics of Helsinki University of Technology, while the upscaling and homogenisation of fractured rock were studied at Uppsala University in Sweden and at the Laboratory of Rock Engineering of Helsinki University of Technology. The permafrost issues were studied at the Laboratory of Structural Mechanics of Helsinki University of Technology. The national reports on these issues were published by the STUK (Eloranta 2004a). The general outline of the project is described by Eloranta (2004b).

In addition to these issues a master's thesis was written at the Institute of Mathematics of Helsinki University of Technology. The thesis dealt with the mechanical stability of a canister in a bentonite buffer (Lahtinen 2002).

After the DECOVALEX III project, a new phase called DECOVALEX THMC has been ongoing since 2004. The letter ‘C’ in the acronym means “Chemistry”.

CASES STUDIED IN FINLAND IN DECOVALEX III

Full descriptions of the Tasks and Bench Mark Tests (BMT) can be found on the DECOVALEX web pages: www.decovalex.com. In the following section short summaries are given of the Tasks and BMTs studied.

Task 1. FEBEX in-situ T-H-M experiment

The test case was based on the now-completed FEBEX in situ experiment carried out in Switzerland. Two large-scale experiments were performed: 1) an in situ field test of a heater-buffer-rock system with a long period of heating, followed by 2) a large-scale laboratory "mock-up" test. The aim of the FEBEX project was to demonstrate the present capabilities to build bentonite barriers in conditions similar to those encountered in actual repository design and to provide monitoring data to make it possible to understand coupled THM and THG (Thermo-Hydro-Geochemical) processes in the near field. Large quantities of monitoring data regarding stress, deformation, water content, water pressure, and temperature distributions and their histories over time at a large number of monitoring places were recorded in situ and a large number of rock/buffer property parameters were also measured in laboratory tests.

On the basis of this data package, a benchmark test (BMT) problem was proposed. The object of the BMT was to apply different numerical models and codes to simulate the FEBEX in situ field test, with the support of available monitored in situ system responses and laboratory property data, for the coupled hydro-mechanical or coupled thermo-hydro-mechanical processes of the fractured rock-buffer-heater system. The aim was to verify the currently available numerical models for coupled THM processes, improve confidence levels in numerical modelling, and deepen understanding of the coupled behaviour of fractured rock-buffer interactions during heating. The BMT was divided into two subtasks: A) simulation of the hydro-mechanical behaviour of the fractured rock mass with respect to the tunnel excavation; and B) the simulation of the coupled thermo-hydro-mechanical responses of the complete rock-buffer-heater system during the whole heating period.

The first subtask required predictions of the redistribution of the water head field, flow rate field, stress field, and deformation field in the rock mass induced by tunnel boring. It was possible to use numerical models and calibrate them against monitored data on geological and hydrological characterisation. The hydraulic tests carried out before the excavation of the tunnel were also used in this context.

The second subtask required predictions of the responses of buffer and rock mass and their interactions, including temporal evolutions and spatial distributions of temperature, water content, water pressure, and stress and deformation of the buffer material and rock mass near the tunnel. The results were compared at selected points. As a global measure of the rock-buffer-heater system, it was also intended to predict the time history of the total system power input to the heater. It was possible to maintain the prediction-calibration cycle throughout the BMT so as to enhance the numerical capability and improve confidence.

The national contributions to Task 1 Subtask B are presented by Jussila (2004). In the present report, Chapter 2.2.5 also deals with the Finnish contribution. The bentonite buffer modelling work has also continued in the DECOVALEX THMC project since 2004.

BMT 2

The homogenisation and upscaling of the BMT were basically concerned with the relationship between an equivalent continuum (which could be heterogeneous) and detailed discrete representations of fractured rocks, and the extrapolation of rock properties obtained from small-scale tests and observations on a large repository scale, with analysis for uncertainties. The main PA measures were the methods of derivation of the flow and deformation properties of the fractured rock, from a small detailed model to a large-scale equivalent continuum model. The database developed at Sellafield for Task 1 of DECOVALEX II was used for the detailed technical definition of the BMT.

The details of the national contribution have been presented by Öhman et al. (2004) and Antikainen (2004). The academic dissertation by Johan Öhman (2005) at Uppsala University deals with upscaling.

BMT 3

The Glaciation BMT was mainly concerned with the hydro-mechanical impacts of a cycle of glaciation and deglaciation on the long-term (up to 100,000 years) performance of a hypothetical post-closure repository, without considering the thermal effect. Many different scenarios could be included as alternative contents, such as permafrost, different ice-rock interface conditions, 2D-3D transition, inland/coastal repository locations, sea level changes, saline water intrusion, fracture initiation, propagation and creeping, etc. The main PA measures were the maximum deformation, changes in permeability fields, flow patterns, and formation of critical flow paths, ground surface subsidence, and rebound. Only long-lasting and large-scale changes in PA measures were significant.

The Finnish research team at Helsinki University of Technology (TKK) was the only one which studied permafrost and the thermal influences of the repository on permafrost. The numerical simulations concerning permafrost were performed by Hartikainen (2004).

The achievements of the DECOVALEX III project have been published in the international literature as research papers and as a volume in a book series on Geo-engineering edited by Stephansson, Hudson, and Jing (2004).

DECOVALEX THMC PROJECT FROM 2004

The bentonite buffer modelling work carried out by Jussila (2004) has continued in the DECOVALEX THMC project since 2004; see Chapter 2.2.5.

In the DECOVALEX THMC project the time-dependent fracturing of rock mass has also been studied. The national work is being performed by Rinne from Fracom Oy, and the results gained so far are presented in Chapter 2.3.2 of the present report.

THE NATIONAL DECOVALEX III GROUP

As the Finnish co-ordinator of DECOVALEX research, the STUK organised *The National DECOVALEX III Group* (NDG). The Group was open to all who were interested in the problems of the international project and who wanted to contribute in one way or another to the national effort. The group had eight meetings during 1999–2003. The last meeting was held at the STUK's premises on October 6, 2003. It was at the same time a seminar in the KYT programme.

After the DECOVALEX III project a smaller national group for the DECOVALEX THMC project was established. The STUK’s representative has acted as a chairman also in the national DECOVALEX THMC group.

CONCLUSIONS

The experiences gained during international DECOVALEX projects from 1991 onwards have meant an increasing and deepening understanding of the coupled processes of repository conditions. Coupled processes have been acknowledged as being central phenomena in repository conditions. In DECOVALEX III (1999–2003), large-scale and realistic modelling problems were simulated and analysed. The work also required the initial development of appropriate conceptual models. This kind of work has also continued in the DECOVALEX THMC project since 2004.

The projects have also had an impact on the national level through the education of new experts on nuclear waste management. A few academic theses have been and others will be published in the near future. This is an important aspect when considering the general KYT programme, whose one aim is to guarantee the existence of expertise in Finland.

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2.1.2 Natural geochemical safety indicators[5]

INTRODUCTION

The International Atomic Energy Agency (IAEA) started the coordinated research project (CRP) "The use of selected safety indicators (concentration; fluxes) in the assessment of radioactive waste disposal" in 2000; the project was completed in 2005. Nine countries participated in the CRP: Argentina, Brazil, China, Cuba, the Czech Republic, Finland, Japan, Sweden and the United Kingdom.

The objective of the CRP was to contribute to the assessment of the long-term safety of radioactive waste disposal by means of additional safety indicators based on the observation of natural systems. The main focus of the programme was on long-term safety aspects.

In the following section the most relevant results from the Finnish point of view will be reviewed briefly. The final report of the CRP was published in the IAEA TECDOC Series at the end of 2005 (IAEA 2005).

STUDY AREAS

A look at the groundwater flow systems at the four different repository candidate sites in Finland studied in detail by Posiva is indispensable if a deeper understanding of the geochemical flow systems in the country is to be achieved. These four sites are unique in that there is no other place in Finland which has been studied with a comparable comprehensive use of advanced characterisation methods. Two of the investigation sites are coastal sites (Olkiluoto, Loviisa (Hästholmen)) and two are inland sites (Romuvaara, Kivetty). A comparable level of detailed knowledge is also available at the Palmottu natural analogue study site, the location of a uranium mineralisation that is also included in the CRP.

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[5] By Karl-Heinz Hellmuth, STUK.

ELEMENTAL AND RADIONUCLIDE CONCENTRATION DATA

The selected countrywide concentration data from the various compartments (till, stream sediments, stream water, groundwater from dug and drilled wells and lake sediments) are summarised in Table 1. In addition, a map showing the areal distributions for U in till is given in Figure 4. Significant variation is caused by differences in the elemental content of the parent rocks. Concentrations of As, Cu, K, Rb, Th, U are given for till, while for other materials the list of analysed elements is shorter. The missing data are Rb for stream sediments, As, K, Rb, and Th for lake sediments, and Th for dug and drilled well groundwaters; Ra values are only available for well waters. Cs concentrations were not analysed and of the Sn concentrations in bedrock groundwater that were analysed only 10 out of 1000 were above the detection limit. Element-specific results are given and discussed according to their relation to geological provinces.

Considering the present state of geochemical elemental transport cycles working between bedrock, bedrock groundwater, and the surface environment, it seems that for geological, topographic, and climatic reasons, the main direction of transport is downwards within the overburden profile and horizontally along the surface layers. Most of the fluxes seem to occur in the upper soil layers, where the intruding rainwater is soon buffered and becomes less aggressive when reaching the C-horizon or the bedrock surface and the rock fracture system. In a larger global context these observations are to be expected, as the prerequisites for active elemental transport by deep groundwaters to the biosphere, which can be observed, for example, in large parts of central Europe – high relief, high thermal gradients, deep-reaching conductive tectonic structures, and/or the release of gases from great depths – are not encountered in Finland.

Detailed site-specific concentration data from Posiva's investigation sites, as well as from the Palmottu study site, are available mainly from earlier studies. These are from boreholes and focus on rock and groundwater. In baseline studies radioactivity and the concentration of radioactive elements at the nuclear waste repository candidate sites were compared with average values for the whole country. Uranium concentrations in groundwater often show large scatter with depth, and it is of dubious value to calculate average concentrations without understanding the processes that influence these concentrations. The observed values depend on the groundwater flow pattern in the rock fracture system, groundwater chemistry, and the geochemistry of the flow paths. The primary value of these data is their use in understanding and modelling the geochemistry of a site, but they are also valuable for mass balance estimations. As an example, the concentration distribution of uranium in groundwater at Palmottu is given (Figure 5); there, oxidising and reducing types of groundwater can be clearly distinguished. There are two types of oxidising waters with enhanced U contents that are practically limited to the upper 150 m of the system.

Table 1. Statistical values of element concentrations in different compartments in Finland.

		Count	Min	5 %	Median	Mean	S.D.	95 %	99 %	Max
Lake sediments										
Cu	mg/kg	15304	1.00	2.00	18.0	37.5	78.2	129	284	3280
U	mg/kg	15304	0.008	0.824	3.15	7.52	58.9	18.3	63.9	5020
Till: Reconnaissance scale										
As	mg/kg	1054	0.13	0.257	2.56	3.59	4.60	11.5	25.8	44.0
Rb	mg/kg	1054	<15.0	40.9	72.6	76.1	25.1	123	152	239
U	mg/kg	1054	<0.3	1.61	3.08	3.42	2.04	6.37	8.86	48.2
Till: Regional scale										
As	mg/kg	82065	<40.0	<40.0	<40.0	.	.	<40.0	63.2	2550
Cu	mg/kg	82065	<8	<8	21.9	28.4	25.5	69.4	111	1640
K	mg/kg	82065	<1300	<1300	1950	2470	1855	6090	9450	24800
Th	mg/kg	82065	<20	<20	<20	.	.	26.1	42.4	639
Stream sediments										
As	mg/kg	1166	<0.05	0.751	2.89	5.41	14.2	14.7	69.1	268
Th	mg/kg	1166	<0.04	1.52	4.94	5.69	3.47	12.7	17.6	31.2
U	mg/kg	1166	<0.01	0.611	2.01	4.31	14.9	11.8	41.7	379
Cu	mg/kg	1166	<1	5.03	12.4	15.9	18.4	35.6	59.9	476
K	mg/kg	1166	<200	502	1550	2220	1999	6647	10562	14100
Stream water										
As	µg/l	1154	<0.05	0.059	0.359	0.551	0.627	1.58	3.85	6.5
Cu	µg/l	1154	0.059	0.17	0.638	0.932	1.32	2.36	6.04	24.5
K	mg/l	1154	0.068	0.203	0.695	1.18	1.38	3.83	7.2	17.1
U	µg/l	1154	<0.01	<0.01	0.073	0.186	0.437	0.728	2.35	6.28
Groundwater: Drilled bedrock wells										
As	µg/l	263	<0.05	<0.05	0.16	0.999	2.82	6.74	15.0	23.6
Cu	µg/l	263	<0.04	<0.04	9.14	32.3	78.3	146	357	917
K	mg/l	263	0.23	0.719	3.00	4.42	4.93	14	23.9	40.2
Rb	µg/l	263	0.03	0.153	1.87	3.19	4.71	10.6	27.6	42.7
Th	µg/l	263	<0.02	<0.02	<0.02	.	.	0.096	0.154	1.41
U	µg/l	263	<0.01	0.01	0.68	13.7	55.1	54.5	309	643
Rn	Bq/l	263	1.00	8.87	142	311	593	1390	2980	4880
Groundwater: Dug wells										
As	µg/l	739	<0.05	<0.05	0.14	0.353	1.00	1.33	4.74	19.7
Cu	µg/l	739	<0.04	0.136	2.53	14.1	33.3	76.2	176	410
K	mg/l	739	0.19	0.587	2.79	4.97	7.36	17.3	43.9	92.3
Rb	µg/l	739	0.04	0.227	2.73	4.98	7.45	15.9	41.1	73.3
Th	µg/l	739	<0.02	<0.02	<0.02	.	.	0.140	0.583	1.50
U	µg/l	739	<0.01	0.01	0.09	0.846	3.03	3.83	21.5	36.6
Rn	Bq/l	739	0.10	2.00	12.0	37.8	82.8	194	526	893

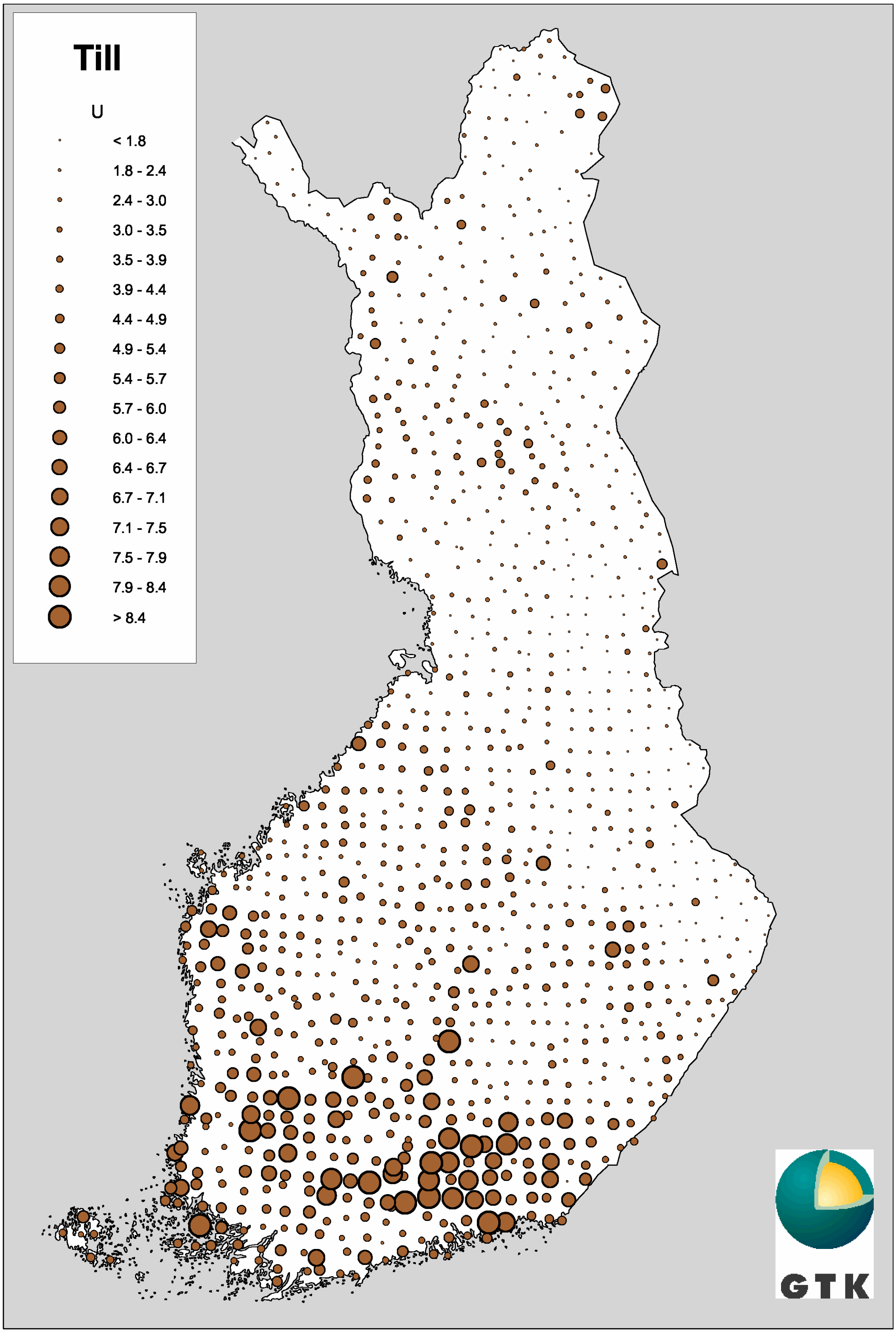

Figure 4. Uranium (U) concentration (mg/kg) in till in Finland.

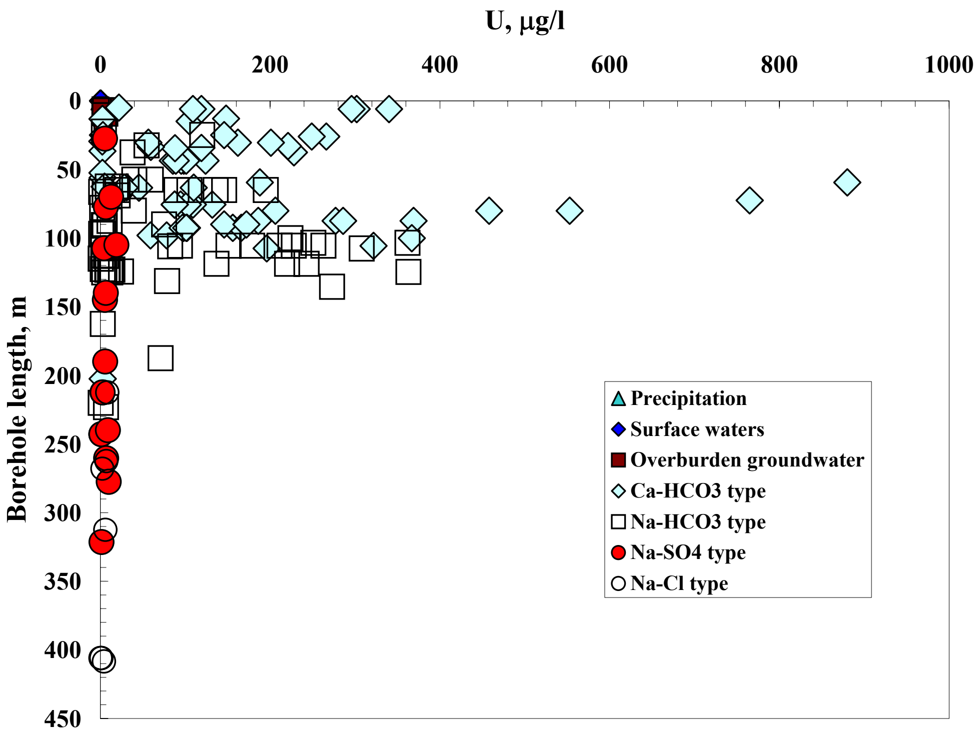

Figure 5. Uranium concentration in groundwater versus borehole length, Palmottu.

ELEMENTAL AND RADIONUCLIDE FLUX DATA

There are numerous approaches used to measure or estimate elemental fluxes caused by weathering and chemical erosion, but they often require certain assumptions. Therefore, there can be more certainty about the results when the rates obtained by various independent methods are compared. Some methods are more reliable in regions of short regolith weathering histories, such as glaciated terrain, as in Finland. The direct use of abundant and versatile till geochemical data available in Finland improves weathering rate estimates by giving more reliable information concerning weatherable base cations than bedrock data.

From an investigation programme on geologically and hydrogeologically well-defined small catchments over a longer time period, a number of representative sites were selected. Groundwater chemistry and the observed large and rapid variations in the chemistry lead to the conclusion that groundwaters flow mostly in the Quaternary in all five catchments and represent shallow groundwaters characterised by short contact times with the geological substrate. Those geochemical fluxes that are worth mentioning seem to be confined to the superficial geological layers, which include overburden and the most fractured uppermost layers of bedrock. The influences of fluxes, which might bring substances from greater depths, cannot be distinguished in these catchments. The results show a significant scatter of groundwater chemical erosion rates in the different catchments. From the average amount of K removed in the catchments since the end of the latest glaciation a theoretical weathering depth of about 40 mm can be derived for the average granitic composition with about 3% K under the assumption that weathering is limited to the surface layer only, a number which seems reasonable. A thicker overburden seems to lead to higher trace metal concentrations in water as a result of more intense water-rock interaction, but the presence of rock fractures does not find unambiguous expression in element contents or fluxes. It is clear that the observed differences depend on the geochemistry of all branches of the relevant flow paths and not only on the properties of the parent material, and conclusive explanations would require very detailed site investigations. In most cases the chemical weathering rate estimates give at least some insight into the order of magnitudes in areas where the anthropogenic input is low. For the future it can be expected that weathering rate estimates will be improved as a result of the increasing availability of selectively leached, mobile metal ion data.

It is emphasised here that the comparison of fluxes can hide the problem of high concentrations of harmful elements in small streams with low runoff. It has been recommended to compare the concentrations of metals in regional maps with the lowest concentrations likely to cause injurious effects to aquatic organisms, the *so-called* "lowest known levels of effect". These values were exceeded for Cu in four streams examined in this study, although the estimated Cu fluxes for these four headwater streams were lower than the mean value for the 30 streams studied.

The uranium mass balance estimation for the Palmottu mineralisation (about 1000–1500 t of U) showed that very little U has been lost from the system during its long geological history. When considering a block of rock containing most of the ore, it is interesting to note that the same order of magnitude of U is actually contained in the rock matrix itself (about 1500 t). Most of the small fraction of the inventory that has been mobilised is at present found adsorbed on fractures or fixed in fracture coatings in the oxidising zone comprising the upper 100 m (8 respectively 23 t of U). The respective values in the reducing zone (lower 150 m) are 6 and 0.2 t. The amounts of U in groundwater are much lower: flowing bedrock groundwater in fractures in the oxidising zone 0.15 kg; in the reducing zone: 0.002 kg; stagnant pore water in rock matrix in the oxidising zone: 3 kg, and in the reducing zone: 0.045 kg. The measured U concentrations in lake and river water are very low; in the groundwater of the overburden the U inventory is also low, about 0.72 kg. The major sinks for U in the biosphere are: a lake (0.08 kg U); lake sediment (130 kg U), and two peat bogs (84 + 17 kg U). In the overburden 11 tonnes of U are found, but only 0.2% of that amount is mobile. The peat bogs do not seem to be the main groundwater discharge locations. The time scale for the accumulation of U in secondary sinks is about 10,000 a.

The U mass balance and the fluxes to the biosphere are displayed in Figure 6. The key conclusion is that the U flux from the upper, oxidising zone is much higher (160 g/a) than that from the lower, reducing zone (0.002 g/a), but in the same order as the U flux caused by surface bedrock erosion and chemical weathering (70 g/a). Natural uranium fluxes are very limited under reducing conditions, but under oxidising conditions too mineralisation processes and organic complexation limit or delay the transport of U. Considering long-term perspectives, there are indications that during past glaciations melt waters may have intruded more deeply into the flow system at Palmottu, but reducing conditions were largely maintained by the redox buffering capacity of the rock.

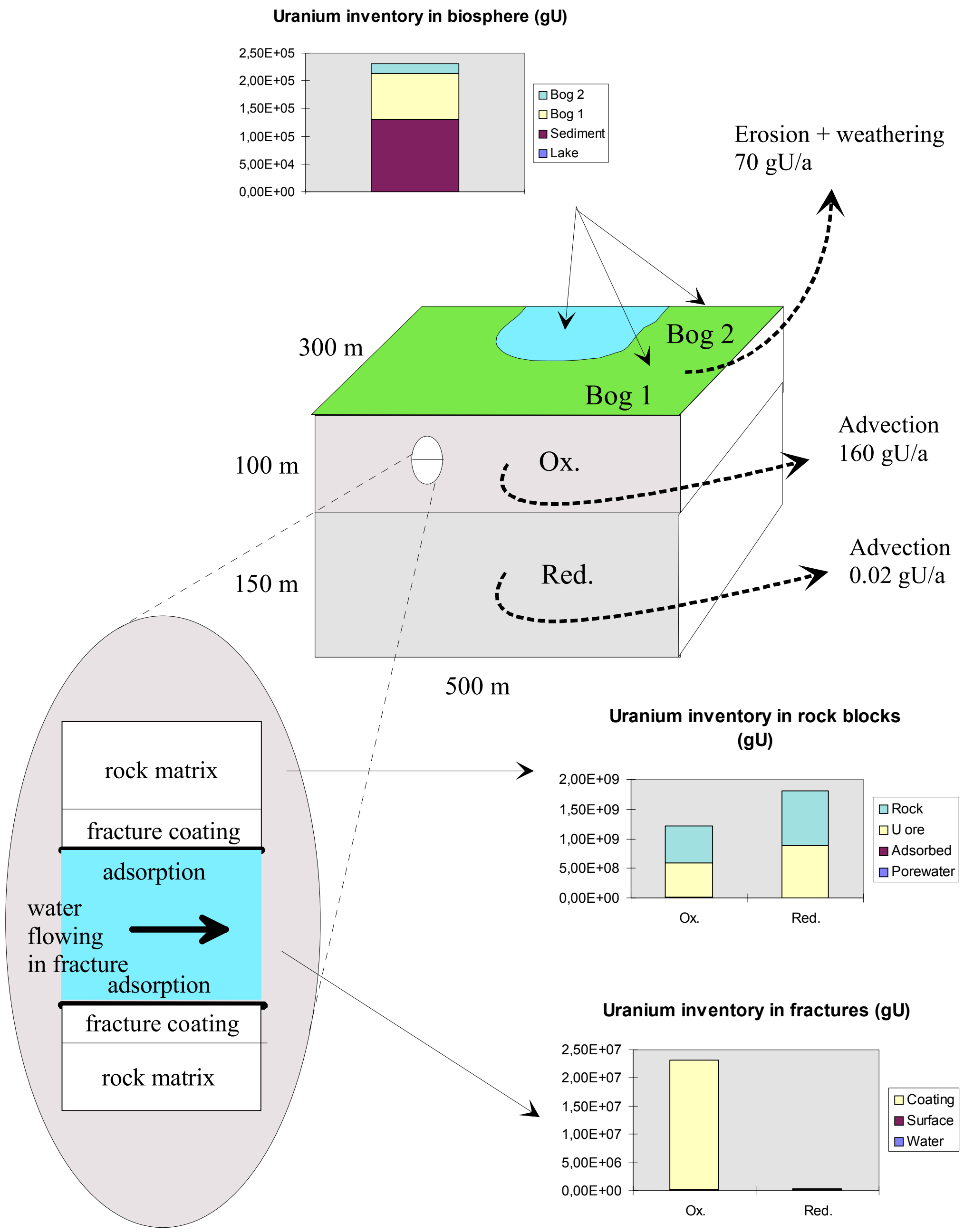

Figure 6. U inventories and fluxes for a bedrock block at Palmottu.

INTERPRETATION AND APPLICATION OF THE CONCENTRATION AND FLUX DATA

The geochemical mapping data for Finland were used for areas where concentrations of harmful elements exceed permissible limits and for epidemiological studies. The focus was on F, As, and U, but also on some other heavy metals. The limit for As was exceeded in about 1% of the drilled wells and very high values were found in individual cases. There seemed to be an association between high As values and bladder cancer. In baseline studies at repository candidate sites it was found that U contents in drinking water from drilled wells in bedrock were roughly one order of magnitude higher than in municipal water supplies and the significant role of U in the radiation exposure of the population was emphasised. In drilled wells the local value for Loviisa was about twice the average for the whole country. In Finland it is estimated that in about 100,000 springs and drilled wells the limit for U of 2 µg/l is exceeded, which means that about 800,000 people may be exposed to high U levels. The maximum U content measured in a drilled well in bedrock was 12,400 µg/l. The role of concentrations as a measure that can be used to evaluate possible harmful effects is emphasised here, as the comparison of fluxes alone can hide the problem of high concentrations of harmful elements under certain circumstances.

In the Finnish literature the results of performance assessment simulations for the long-term chemical effects on the environment of the substances released from the materials in a nuclear waste repository are reported. The models used were partly simplified from the recent TILA-99 performance assessment exercise and thus gave rough overestimations of releases, which were accompanied by considerable uncertainties. The resulting concentrations in drilled well water were compared with current drinking water standards. Although the solubility limits of many of the substances are higher than the permissible concentrations in drinking water, because of the low water flow the calculated release rates are very low and the dilution factor is high, so that the resulting concentrations in the well water are at least four orders of magnitude below permissible limits.

Natural concentrations and fluxes from Finland have been reported to the EU project "Testing of safety and performance indicators" and were included in the list of reference values, but obviously no detailed comparisons with performance assessment results could be conducted within the framework of the project. Recently, site-specific data from Palmottu have been compared with performance assessment predictions (Read et al. 2003). The TILA-99 performance assessment was designed to be transparent and is readily amenable to comparison with alternative approaches. Dose calculations relate to a well for drinking water that is assumed to be located in the vicinity of the repository or in the groundwater discharge zone. This is the only exposure pathway considered. The model employed assumes that the annual releases from the repository into the biosphere are diluted in 100,000 m^3 of water and that an individual drinks 500 dm^3 of this water per year. For cases where the canister is assumed to 'disappear', the maximum release rate for U from the geosphere is given as only 1.2 Bq y^{-1} (and then after 1 million years). This equates to a concentration of $4x10^{-15}$ mol dm^{-3}. In comparison, the uranium concentrations measured at Palmottu range from around $4x10^{-9}$ mol dm^{-3} for the most reducing waters to $>10^{-6}$ mol dm^{-3} nearer the surface. The safety case considered release from only one canister; however, scaling up to match the total inventory at Palmottu (~1000 t), is insufficient to account for such a large discrepancy. If all the canisters were assumed to disappear there would still be 10^3–10^6 times less uranium in the simulated well water than is actually observed in the geologically similar Palmottu system (Figure 7). The difference is entirely due to assumptions made regarding the geochemical behaviour of uranium. This is not only a feature of TILA-99 but of all current performance assessment calculations. It is interesting to see nuclide-specific constraints for the release of activities to the biosphere according to the Finnish regulations (Guide YVL 8.4) when they are converted to mass units in relation to natural fluxes; a limit of 0.3 GBq/a for U isotopes (averaged over < 1000 a) is equivalent to as much as 30 kg/a.

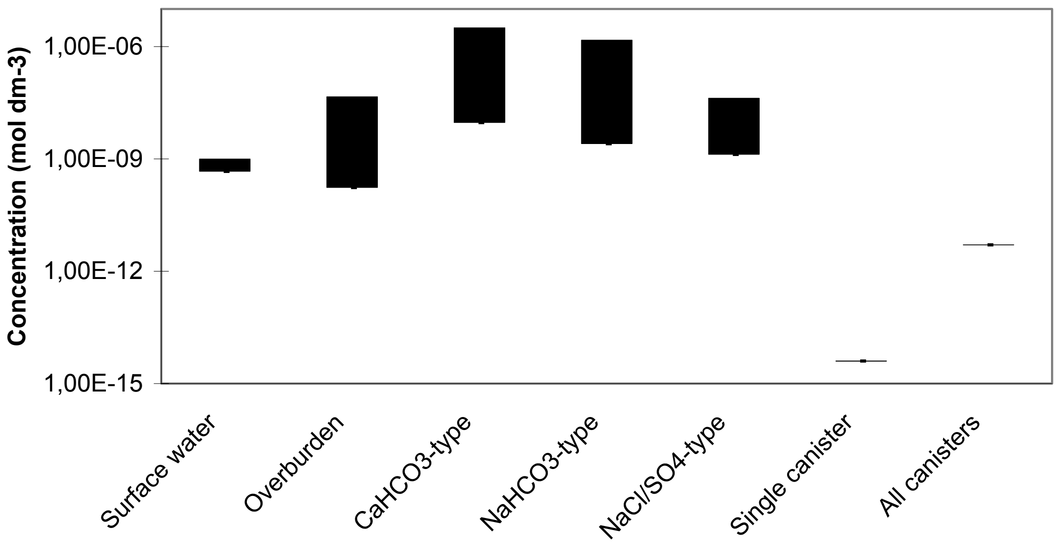

Figure 7. Uranium concentrations in Palmottu surface and groundwaters compared to performance assessment predictions for a drinking water well (Read et al. 2003).

In the future it is expected that on the basis of a better understanding of natural U fluxes more realistic models for radionuclide migration in the geosphere can be developed and the development of performance assessment tools can benefit from these. As a first step the preliminary results of these alternative models can be compared with the results obtained by conventional performance assessment tools. When the behaviour of U at the Palmottu site can be simulated satisfactorily, then attempts can also be made to simulate the behaviour of spent fuel at the same site and thereafter the alternative modelling approach can be tested in the Olkiluoto system.

CONCLUSIONS

The benefits of the CRP for Finland can be divided into three main areas:

- A comprehensive database of natural concentrations and fluxes has been compiled for the derivation of reference values in different scales (for example, country-wide, according to geochemical provinces, and site-specific), as well as for various compartments or materials.

- The basic understanding that has been achieved of the processes leading to the mobilisation and fixation (dispersion and enrichment) of elements in geochemical cycles and the consideration of the global context in terms of geology, topography, and climate implies a significant increase of confidence in the ability to understand and quantify the long-term behaviour of a nuclear waste repository. The only available geological option in Finland which is characterised by water-saturated, fractured crystalline rock, glaciated terrain, peneplain, secondary sinks etc. can now be better evaluated against alternative geological options all over the world. For example, the geological conditions in Finland do not favour large geochemical fluxes mediated by groundwater from deep formations to the surface. In addition, more detailed knowledge of the complexity of natural processes facilitates the drawing of simplifying conclusions that can be beneficial in communication with non-geoscientific and non-technical audiences.

- Detailed and quantitative knowledge of local, element-specific natural geochemical fluxes is being used to develop better modelling tools to be integrated into a safety case, which take account of the interactions between geochemical and hydrological processes and do not fundamentally misrepresent the known geochemical behaviour of the elements.

The assumption in repository performance assessments of the simple dispersion and dilution of ions released from the waste, transported through the geosphere by groundwater and finally reaching the biosphere, is not supported by observations from natural anomalies. In nature complex mobilisation, migration, and, in particular, reconcentration (followed eventually by remobilisation) processes prevail. The nearer to the topsoil layers metal ions migrate, the more complex their behaviour and the more difficult predictions can be made.

Experiences from Finland show that under the influence of the presence of mostly unweathered, largely crushed bedrock, low hydraulic conductivity in till and groundwater saturation with groundwater levels often relatively near to the top level of the regolith profile, in connection with a low relief, often leading to less oxygen-rich or partly reducing conditions already below less than a metre in depth as a result of the high buffer capacity of the fresh, fine-grained rock material and the low groundwater flow rate, chemical fronts of pH, redox conditions, anion and complexant concentrations etc. can act as effective chemical barriers limiting the movement of elements and leading to secondary enrichments, depending on their respective chemical properties. When these natural geochemical characteristics are taken into account in future repository performance assessments, then the comparison with natural elemental concentrations and fluxes makes sense, can support a safety case, and conforms to the protection goals of the Finnish regulations giving nuclide-specific release limits.

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LITERATURE

In this section only a summary of the most prominent results contributed to the CRP is given. For detailed references (original citations) we give a list of reports that contain all the information from the literature on the methods and data, as well as results, of supporting studies putting the Finnish situation in a more global context. These reports were all published before the end of 2003.

Hellmuth, K.-H., Tarvainen, T., Backman, B., Hatakka, T., Vesterbacka, P., Savolainen, H. 2003. IAEA Coordinated Research Project, The Use of Selected Safety Indicators (Concentrations, Fluxes) in the Assessment of Radioactive Waste Disposal, Report 4, Natural Geochemical Concentrations on the Baltic Shield of Finland for Use as Indicators of Nuclear Waste Repository Safety., Rep. YST-109, Geological Survey of Finland, Nuclear Waste Disposal Research, GSF, Espoo, Finland, 2003.

Kaija, J., Rasilainen, K., Blomqvist, R. 2003. IAEA Coordinated Research Project, The Use of Selected Safety Indicators (Concentrations, Fluxes) in the Assessment of Radioactive Waste Disposal, Report 6, Site Specific Natural Geochemical Concentrations and Fluxes at the Palmottu U-Th-mineralization (Finland) for Use as Indicators of Nuclear Waste Repository Safety. Rep. YST-114, Geological Survey of Finland, Nuclear Waste Disposal Research, GSF, Espoo, Finland, 2003.

Pitkänen, P., Löfman, J., Luukkunen, A., Partamies, S. 2003. IAEA Coordinated Research Project, The Use of Selected Safety Indicators (Concentrations, Fluxes) in the Assessment of Radioactive Waste Disposal, Report 7, Site Specific Natural Geochemical Concentrations and Fluxes at Four Repository Investigation Sites in Finland for Use as Indicators of Nuclear Waste Repository Safety., Rep. YST-115, Geological Survey of Finland, Nuclear Waste Disposal Research, GSF, Espoo, Finland, 2003.

Read, D. 2003. IAEA Coordinated Research Project, The Use of Selected Safety Indicators (Concentrations, Fluxes) in the Assessment of Radioactive Waste Disposal, Report 8, Natural Uranium Fluxes and Their Use in Repository Safety Assessment. Implications for Coupled Model Development, Rep. YST-116, Geological Survey of Finland, Nuclear Waste Disposal Research, GSF, Espoo, Finland, 2003.

Read, D., Hellmuth, K.-H., Kaija, J., Ahonen, L. 2003. Natural Uranium Fluxes and Their Use in Repository Safety Assessment. In: Merkel, B.J., Planer-Friedrich, B., Wolkersdorfer, C., Uranium in the aquatic environment, Springer, Berlin, 2003. P. 115–127.

Tarvainen, T., Backman, B., Hellmuth, K.-H., Hatakka, T., Savolainen, S. 2003. IAEA Coordinated Research Project, The Use of Selected Safety Indicators (Concentrations, Fluxes) in the Assessment of Radioactive Waste Disposal, Report 5, Chemical Weathering Rates on the Baltic Shield of Finland for Use as Indicators of Nuclear Waste Repository Safety., Rep. YST-113, Geological Survey of Finland, Nuclear Waste Disposal Research, GSF, Espoo, Finland, 2003.

Tarvainen, T., Hellmuth, K.-H., Backman, B. 2005. Natural geochemical concentrations and fluxes of U, Th and Cu in Finland. Geochemistry: Exploration, Environment, Analysis, Vol. 5, 2005, pp. 41–50.

Traber, D. 2002. IAEA Coordinated Research Project, The Use of Selected Safety Indicators (Concentrations, Fluxes) in the Assessment of Radioactive Waste Disposal, Report 3, Geohydraulic and Geochemical Basis of Geochemical Fluxes from Deep Groundwaters Observed at the Earth's Surface, Appendix: Lahermo, P., Groundwater in Finland – with Special Reference to Groundwater Conditions in Central Europe, Rep.YST-108, Geological Survey of Finland, Nuclear Waste Disposal Research, GSF, Espoo, Finland, 2002.

Vaganov, P. 2002. IAEA Coordinated Research Project, The Use of Selected Safety Indicators (Concentrations, Fluxes) in the Assessment of Radioactive Waste Disposal, Report 1, Geochemical Cycles and the Dispersion and Concentration of Elements in the Earth's Crust – Global, Regional and Local Scale. Appendices: 1. Vaganov, P., Natural Average Geochemical Fluxes of Radioactive and Chemotoxic Elements, 2. Hellmuth, K.-H., Heavy Metal Soils, 3. Hellmuth, K.-H.,Geochemical Halos Associated with Ore Bodies in Finland, Rep. YST-106, Geological Survey of Finland, Nuclear Waste Disposal Research, GSF, Espoo, Finland, 2002.

Vaganov, P. 2002. IAEA Coordinated Research Project, The Use of Selected Safety Indicators (Concentrations, Fluxes) in the Assessment of Radioactive Waste Disposal, Report 2, Dispersion Haloes and Fluxes of Chemical Elements in the Cryolithozone (Permafrost or Periglacial Regions), Rep. YST-107, Geological Survey of Finland, Nuclear Waste Disposal Research, GSF, Espoo, Finland, 2002.

2.1.3 Uranium migration and retention in granitic rock[6]

INTRODUCTION

At the beginning of 2000, the IAEA initiated a Co-ordinated Research Project (CRP) entitled "The use of selected safety indicators (concentrations, fluxes) in the assessment of radioactive waste disposal" in an attempt to achieve an international consensus on how such indicators might be applied. The Finnish contribution to this study was a report series dealing specifically with the geochemical and hydrological recycling of trace elements in a peneplained shield setting.

In parallel the STUK, the Finnish participant in the CRP, embarked on the development of a coupled chemical transport model based on previous work undertaken in Russia. Therefore, a decision was made to test the SONE model in a Pilot Study at the Palmottu natural analogue site. The aim was to demonstrate the feasibility of applying a reactive chemical transport model to a complex natural site. Additionally, it was seen as important to establish data requirements to prepare the more exhaustive tests that would be needed if the model were applied in the safety assessment of a nuclear waste repository.

SONE (modified to SONE_PLM) was used to simulate uranium transport along a hypothetical crosscut section through the Eastern Granite at Palmottu (Oziabkin and Oziabkin 2005; Read 2003). This section was chosen on the basis of the data available from studies within the main Palmottu EU project (Blomqvist et al. 2000). A stream-tube of unit cross-sectional area was constructed and the displacement of the initial solutions (groundwater at various depths) by reaction and mixing with the boundary fluids (rain water and overburden pore water) was simulated.

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[6] By Nuria Marcos, TKK; Karl-Heinz Hellmuth, STUK.

The results of the Pilot Study were reasonable and seemed to agree fairly well with data from the site (Blomqvist et al. 2000). However, a number of uncertainties remained. First, the modelled flow route is largely hypothetical; there is some indication of hydraulic connections between the boreholes, but no substantial evidence of uranium mass transfer in the direction postulated. The land is rising as a result of postglacial rebound and the flow regime may have been different in the past (e.g. Pitkänen et al. 2002). Second, information on mineral transformation rates and associated uranium release or fixation was limited at the time of the calculations of the Pilot Study. The primary (IV) phase, uraninite, is known to be very old (> 10^9 years) but definitive age determinations for U secondary (VI) phases were lacking. Third, further information was needed to decipher the relationship between the rock microstructure and the position of uranium phases. This is a key issue in understanding the role of the rock matrix and the fracture network in water-rock interactions.

The objectives of the supplementary studies described in Read et al. (2004) are to:

- constrain time frames for uranium mobilisation and fixation
- determine whether uranophane precipitation is an ongoing process with successive generations of crystal formation possibly linked to episodic climatic events
- establish the relative contribution of secondary uranophane and residual uraninite to high uranium concentrations in shallow groundwaters.

At Palmottu, U-series dating of well-characterised mineral phases was carried out, thus avoiding the uncertainties introduced by the selective leach of complex samples. The U-series isotopic data obtained were specific to uranophane.

Primary and secondary uranium phases co-exist within the rock matrix in the vicinity of water-conducting fractures. Surveying beta/gamma autoradiography and alpha radiography using polycarbonate films were used to localise and select zones of uranium enrichment for microscopic investigations. Autoradiographies of ^{14}C-PMMA-impregnated samples made possible the correlation of uranium occurrences with the conductive pore network from fracture surfaces exposed to weathering to the rock matrix.

Calculations were performed to investigate the controls on uranophane formation as a function of depth.

COUPLED REACTIVE CHEMICAL TRANSPORT AT THE PALMOTTU NATURAL ANALOGUE SITE

The most recent attempt to model the evolution of the Palmottu ore body is described in Oziabkin and Oziabkin (2005). Although this was essentially a demonstration exercise, it nevertheless represents an advance over earlier modelling studies. A dual porosity model is used to represent fractures and matrix coupled to chemical reactions. It is also possible to change rates of mineral precipitation-dissolution in response to physical constraints (variations in flow direction and rates).

Oziabkin and Oziabkin (2005) describe the application of the SONE code to the Palmottu natural analogue site and, in particular, uranium transport along a hypothetical model section through the Eastern Granite. The modelling work is based on existing knowledge and data from Palmottu, which are compiled, described, and evaluated before integration into the model.

The report contains a detailed model description, including the treatment of the various physical and chemical processes, as well as a description of the numerical modelling approaches and various code verification exercises. It is presented in its original form following translation and has been edited only insofar as to make it understandable to a reader broadly familiar with the technical context.

A novel approach to describing the influences of changing external conditions on the system was further developed after the completion of the pilot modelling study to cover a wider range of scenarios. These results, describing the evolution of the modelled system under different initial conditions and its response to temporary or continuous changes of boundary conditions, are detailed in Appendix 1 in Oziabkin & Oziabkin (2005). A User Manual for SONE_PLM is included as Appendix 2 in the same report.

The time frame considered was 10,000 years, roughly the period since Palmottu emerged from the sea. According to the model calculations, around 44% of the uranium released by the dissolution of the main ore mineral (uraninite) re-precipitates within the model domain as uranophane (Ca(UO2)2(SiO3OH)2(H2O)5), with 56% ‘lost’ as outflow (Figure 8). The proportion fixed by ion exchange or co-precipitation with fracture minerals was predicted to be negligible (Oziabkin and Oziabkin 2005 and references therein).

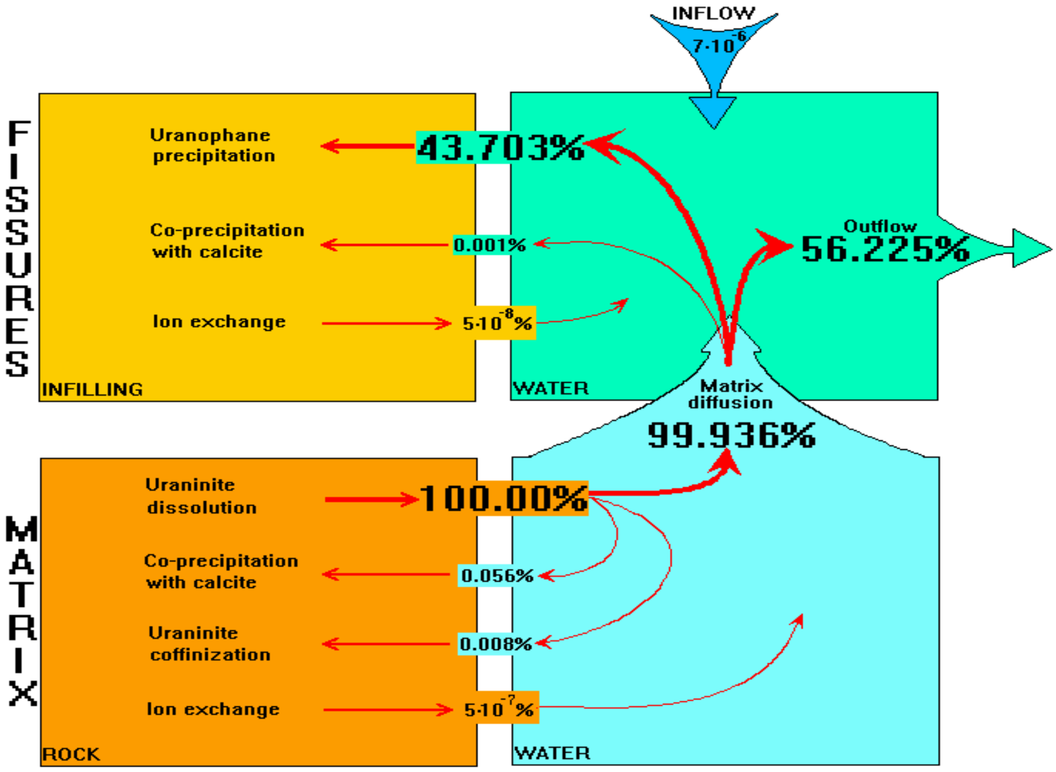

Figure 8. Post-glacial uranium fluxes at Palmottu, near-surface part of eastern flow system (Oziabkin and Oziabkin 2005).

The model accounts for the main geochemical features of the site and provides a description of uranium cycling that is consistent with current understanding. However, during the course of the investigation it became apparent that additional information was needed to constrain the hydrogeochemical interpretation of the Palmottu system, its evolution, and the timescales over which uranium migration has taken place. The necessary experimental work has now been carried out as part of a separate KYT project and is reported elsewhere. Because of time constraints in the project the coupled modelling exercise could not be updated using the new experimental findings.

This study highlighted and demonstrated the potential usefulness of a reactive transport model in a safety assessment framework. It accurately represents our understanding of radionuclide migration processes, employs fundamental thermodynamic, kinetic, and site data at a mechanistic level, and can be verified directly against observations from the field. Such models also provide new insights in terms of evaluating alternative scenarios of system evolution. They could fulfil a similar function as part of a repository safety case.

TIME CONSTRAINTS OF SECONDARY U MINERALISATION IN THE GEOSPHERE: IMPLICATIONS FOR PALAEOHYDROGEOLOGICAL INTERPRETATION

Our understanding of the Palmottu uranium deposit and its evolution has increased considerably in recent years (Blomqvist et al. 2000; Kaija et al. 2003; Blyth et al. 2004), permitting reasonably credible simulations of uranium release, migration, and fixation to be constructed (Oziabkin & Oziabkin 2005 and references therein). Nevertheless, prior to this study several important issues remained to be resolved.

The first of these concerned the relative contribution of primary uraninite and secondary uranophane to groundwater. It is now apparent that the re-working of uranophane dominates in the shallow groundwaters and this phase probably constitutes the main control throughout the oxidised section. Only in deeper waters (≥ 100 m), where incipient oxidation of the primary ore takes place, does the situation change.

The migration of U derived from uraninite through microfissures has been demonstrated by autoradiography of the fractured rock (Figures 9 and 10). Uranium is subsequently precipitated as uranophane from solution, often in open fractures. The results of this study indicate that uranium has migrated on the order of centimetres from its source before precipitating. No evidence has been found of replacement *in situ*. Other U secondary phases that are known to replace uraninite upon oxidation, such as (meta)schoepite, have not been found. The high silica content in the Palmottu groundwaters may be the key factor; equilibrium calculations suggest that uranophane is thermodynamically favoured (Figure 11).

The solubility of uranium determined by its equilibrium with uranophane is lower in natural waters than described above because of the presence of calcium and silica in solution. Figure 11 plots the theoretical solubility of uranium in equilibrium with crystalline (solid line) and synthetic (dotted line) uranophane using the Palmottu groundwater compositions employed in earlier modelling studies. These are fairly typical in terms of major element chemistry. The calculations were performed using PHREEQC (Parkhurst & Appelo 1999) and thermodynamic data were taken from Wolery (1992) and Casas et al. (1994, 1997).

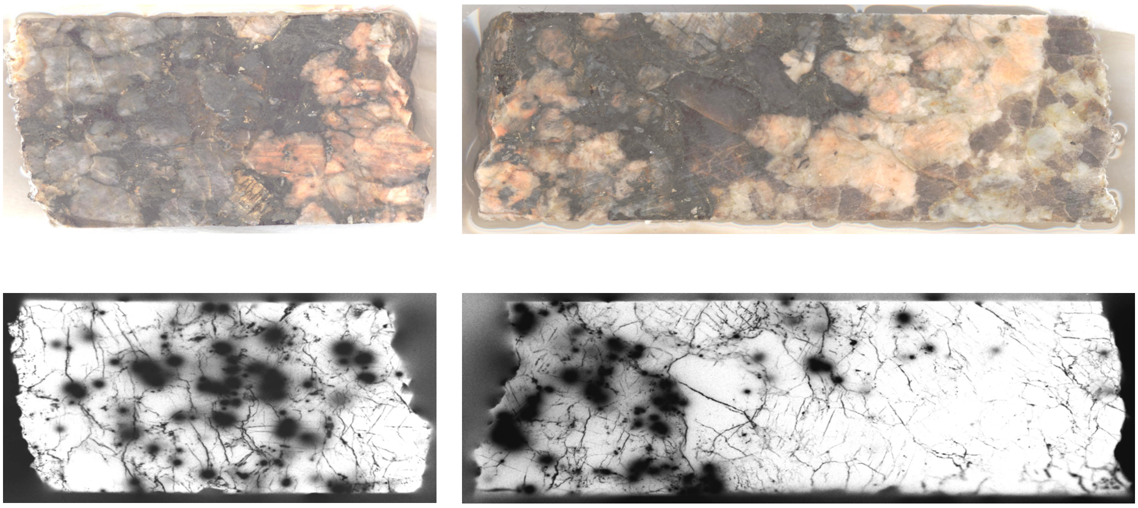

Figure 9. Photograph (above) and corresponding C-14-PMMA autoradiograph (below) of a near-surface sample (length 11 cm). The left-hand side is in contact with the atmosphere. Natural radioactivity of primary U phases (dark halos) inhibits locally structural characterisation by the C-14-PMMA method.

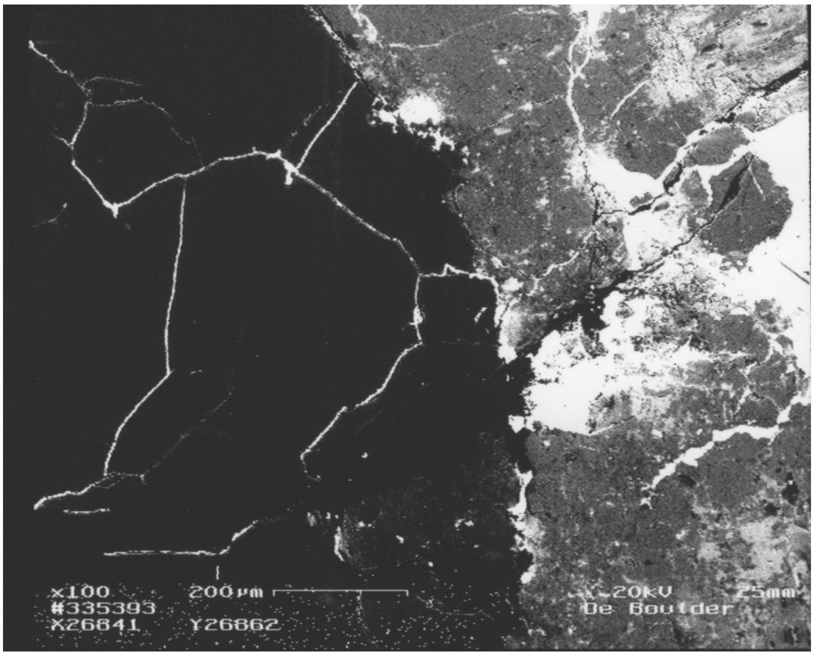

Figure 10. Secondary U phases in the rock matrix: backscattered electron image of another surface sample. Uranophane (white) found in microfissures transecting quartz grain (left) and spread in porous feldspar grain and its microfissures (right).

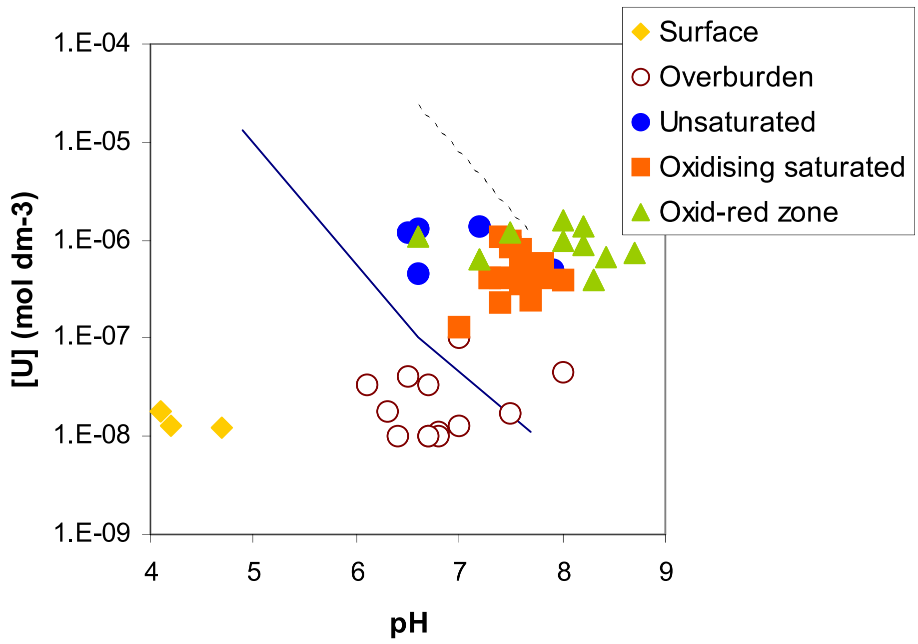

Figure 11. Measured U concentrations in surface and ground waters compared to theoretical solubility of crystalline (solid line) and amorphous uranophane (dotted line).

At shallower depths, within 20 m of the surface, waters are clearly oxidising (~300 mV), with pH values clustering around 7.5 (Kaija et al. 2003). The relatively constant composition of the groundwater from a number of boreholes (R318, R324, R325, R384, R389, R390; see Kaija et al. 2003) is reflected in the close grouping of measured uranium concentrations (Figure 11). They fall between the theoretical solubility of crystalline and amorphous uranophane known to be present. It is suggested that this phase maintains stable levels. As with the deeper samples, a substantial influx of acidic water would be required to promote dissolution. Samples from the unsaturated (or, more strictly, variably saturated) zone also occupy a region between the predicted solubility of crystalline and amorphous uranophane, though they are generally more acidic (Kaija et al. 2003).

In principle, the upper limit of uranium concentrations is determined by the solubility of uranophane as a function of pH (Figure 11); it could exceed the values observed during prolonged periods of acidic recharge. However, in dry spells the pH rises as the water table falls as a result of mixing and reaction with the host rock. Therefore, uranium concentrations in the unsaturated zone will show considerable seasonal variation. It is evident from textural microscopic studies that uranophane is being continually re-worked. It is noted, however, that only a small part of the inventory needs to dissolve for there to be significant changes in solution composition.

The age of the uranophanes constrains the time period over which the oxidation of the ore body has taken place. Previous work on fracture fillings gave ages of 90–120 ka for

R390 (8.61 m) and 189–241 ka for R389 (32.1 m), respectively (Blomqvist et al. 2000). These values were obtained by leaching ill-characterised minerals ('fracture calcites') from drill cores, several of which have now been shown to contain uranophane. Model ages obtained for well-characterised uranophanes in R384 (41.7 m) and R390 (8.5 m) have been given as 110 ± 9.0 ka and 15.3 ± 1.5 ka, respectively (Read et al. 2004).

It was postulated that the uranophane near the surface could be very recent. This has now been confirmed (average age ≤ 2000 years). Further evidence of dissolution fabrics agrees well with the results of groundwater monitoring in shallow wells (Kaija *et al.* 2003).

The final objective, to determine whether uranophane precipitation is an ongoing process or only a process linked to discrete climatic events (i.e. oxidation by glacial melt waters), is partly answered in the discovery of recent, post-glacial uranophane in the near-surface zone (Figure 12). The relatively continuous dissolution and precipitation of uranophane is an ongoing process, especially near the surface. At greater depths, however, the situation is less clear.

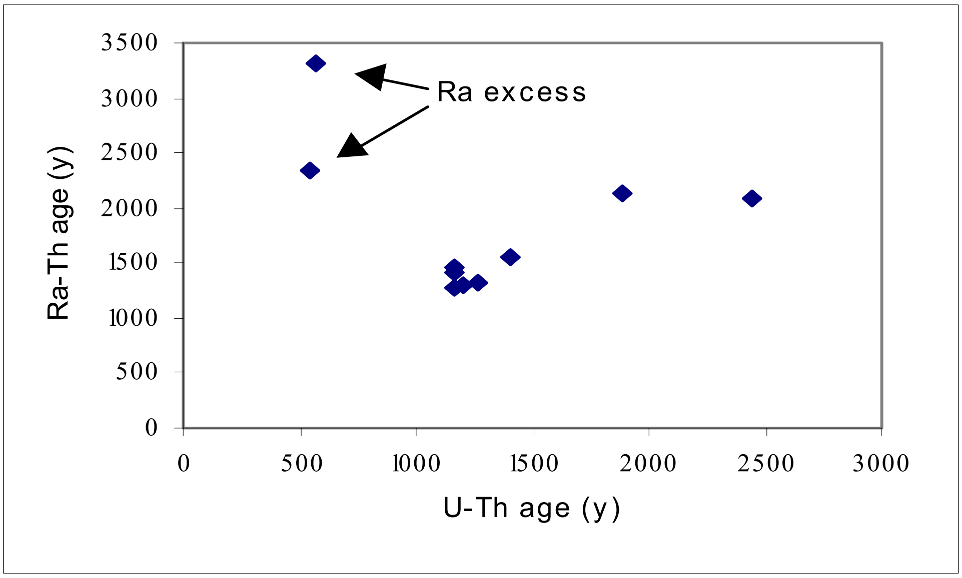

Figure 12. Evidence of recent Ra and U accumulation: Palmottu data (Read et al. 2004).

Episodic formation of secondary uranophane should give rise to discrete ages rather than a continuum. In fact, the results obtained for uranophane (Read et al. 2005) of 110 ± 9.0 ka and 15.3 ± 1.5 ka, correspond to the periods immediately following the interval between the last two glaciations, Saalian and Weichselian, and after the last glaciation. This observation does not preclude the formation of uranophane at other times since the sample set is limited. However, it does imply that no significant modification of these samples has occurred subsequent to these dates.

Active dissolution and re-precipitation is confined to the variable saturated zone. Consequently, continual re-working reduces the average age of near-surface deposits. Further down, uranophane is stable. However, groundwater oxidises to a depth of 100 m and may lead to the dissolution of uraninite in the matrix. The uranium released from primary ore would be expected to form uranophane and add to the inventory already present. This too would have the effect of reducing the average age of secondary U phases.

DISCUSSION

The first objective, to supplement existing evidence concerning the (geologically) recent evolution of the southern Baltic region, has been largely achieved. The advantages of USD dating in the present context are that it furnishes direct rather than inferred information on the timing of past events. Interesting information has been gathered via USD dating of uranophanes from two sites in southern Finland, Palmottu and Hyrkkölä. Published ages for secondary U phases found in southern and central Sweden have been re-evaluated in the light of these findings, providing a substantial and coherent set covering a large geographical area. The positions of Palmottu and Olkiluoto represent similar situations with respect to the possible influence of glacial melt water at different times in past, present, and future evolution (Figure 13).

It is evident that uranium mobilisation and subsequent fixation in the form of secondary U phases has been occurring for at least 100,000 years. The process has not been continuous; none of the samples analysed gave a U-Th age between 40,000–70,000 years BP. Another hiatus is apparent at ~20,000 BP, coinciding with the Last Glacial Maximum (LGM). These results suggest that the trigger for U release is the intrusion of oxidising, glacial melt water. Uranium (VI) minerals form during interglacial periods by sub-aerial weathering. The process can be observed now at Palmottu (Read et al. 2004) and at a number of the sites in Sweden examined by Löfvendahl and Holm (1981) (Figure 14). In this case, however, the much lower hydraulic heads limit the effect to the topmost few centimetres of the sequence.

The interpretation of USD data needs to be viewed in conjunction with other, independent lines of evidence, including stable isotope and fluid inclusion studies on fracture minerals. According to Blyth *et al.* (2004), calcite fracture fillings at Palmottu were formed under late-stage metamorphic or magmatic conditions in the Precambrian and have not been affected by subsequent glacial events. They cite this as evidence that the fractures were not transmissive, even during the loading and unloading of ice sheets. The results presented in this report indicate that the fractures were indeed transmissive, since uranophane in fractures, whether or not associated with calcite, represents precipitation from groundwater. The preservation of calcite only requires the water to be buffered as it moves down through the sequence. The measurements of present-day groundwater demonstrate the effectiveness of pH buffering in the system (Kaija *et al.* 2003).

In addition to providing information on glacial evolution, the results of this study describe the long-term (100,000-year) geochemical behaviour of uranium in conditions relevant to spent fuel disposal. The shallow groundwaters at Palmottu are calcium-rich and, consequently, the presence of uranophane as a secondary alteration product of the primary ore is not unexpected. It also occurs at Hyrkkölä (Marcos and Ahonen 1999) and is the most common secondary mineral reported from similar sites in Sweden. Most of these U occurrences are located in crystalline rocks. Equilibrium solubility calculations carried out as part of the Palmottu study indicate that uranophane is thermodynamically favoured (Read *et al.* 2004). The occurrence of boltwoodite or phospuranylite, as found at several Swedish localities (Löfvendahl and Holm 1981), would require much higher levels of potassium or phosphate, respectively.

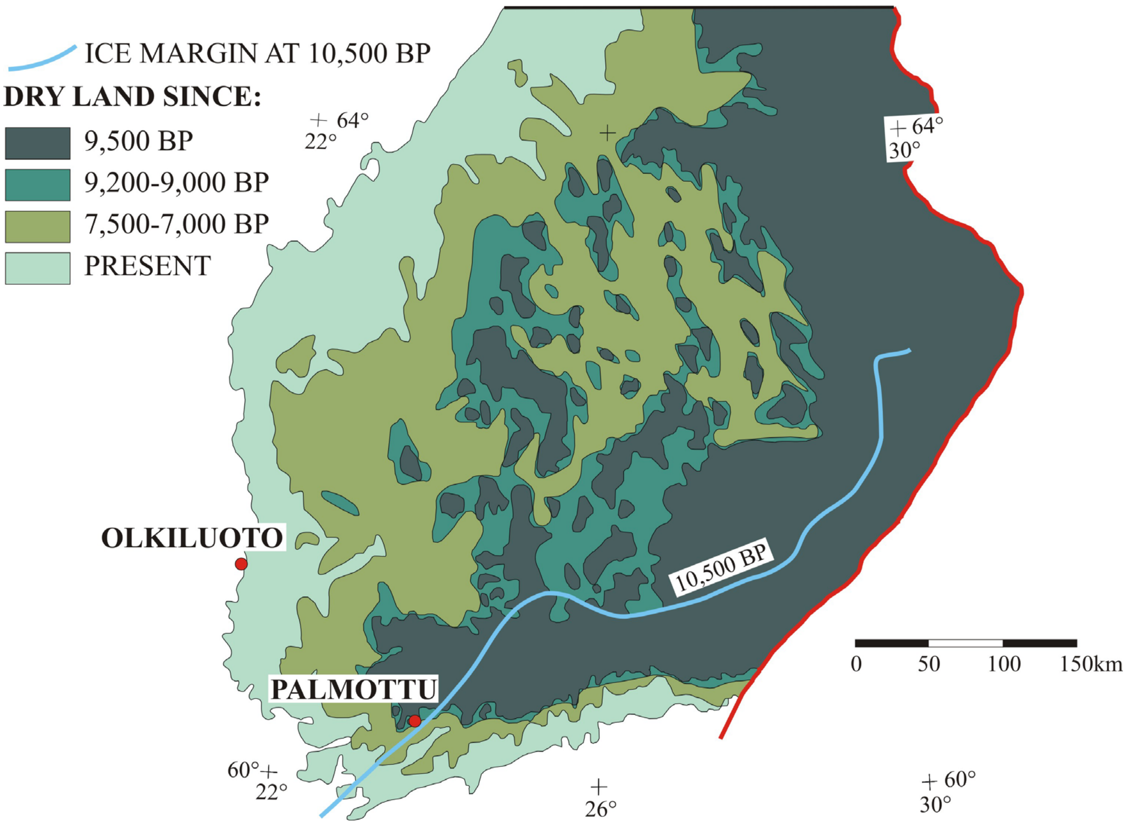

Figure 13. Shoreline displacements in southwest Finland over the last 10 ka (after Pitkänen et al. 2002).

The third objective, to provide realistic scenarios and boundary conditions for model calculations, has been accomplished. The information necessary for constructing a credible model of U release and fixation over the past 100,000 years now exists and can be verified by observations of U occurrences throughout southern Fennoscandia. Thus, many aspects of the model would be transferable to a waste disposal situation in Finland or Sweden, since the geological and geographical conditions are similar.

RELEVANCE OF FINDINGS TO THE SAFETY CASE

Many ‘natural analogue’ sites show little resemblance to the locations chosen for radioactive waste disposal (Miller et al. 1994). This is not the case at Palmottu, which shares a number of features with the proposed repository site at Olkiluoto, some 150 kilometres to the northwest (Read et al. 2002). The geology (gneiss and granite pegmatite) is similar, the hydrogeology is fracture-controlled, and the deposit contains uranium mainly in the form of UO_2 and it extends to at least repository depth (> 400 m). Both sites experienced continental ice margin conditions but represent different stages in post-glacial uplift. Olkiluoto emerged above sea level around 2000 years ago (Pitkänen et al. 2004), in comparison to 10,000 years in the case of Palmottu (Kaija et al. 2003). In this respect, Palmottu provides an indication of the likely future evolution of Olkiluoto over the next few thousand years (Figure 13).

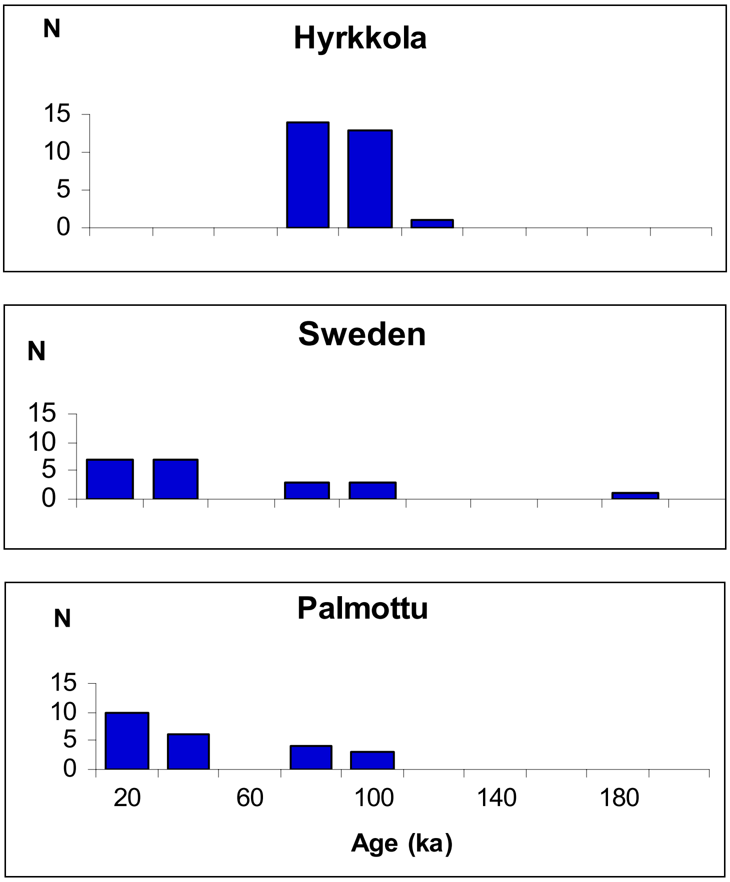

Figure 14. Regional validity: U-Th ages for uranium silicates from Hyrkkölä, Palmottu, and several sites in Sweden (Read et al. 2005).

A major concern for a repository at this latitude is the potential disturbance caused by future glaciations. Previous glacial advances have clearly perturbed Palmottu; uranium has been oxidised to a depth of around 100 m.

Two points often mentioned in safety case considerations, e.g. Rasilainen (2004; p. 145), may be commented on. These are: (i) the poor overlap between safety assessment time frames and those governing mass transport in nature, and (ii) that natural uranium enrichment typically occurs where there is mass transport from oxidising to reducing conditions. The first point is evidently an over-simplification, as uranium mobilisation at Palmottu and many other sites around the world has occurred since the formation of the deposit until very recently, that is, in a time span of millions of years to a few hundred. The second point is supported by the fact that all secondary deposits of U(VI) minerals have been formed by the oxidation of primary ores, usually uraninite/pitchblende. This is also the case at Palmottu and Hyrkkölä. There is abundant evidence that oxygen penetrating along microfissures has mobilised uranium and that this has been transported and subsequently precipitated as uranophane. However, there is no indication that oxidised uranium has been transported from open fractures to the less permeable matrix and reduced there. These findings have important consequences for the safety case, where considerable emphasis is placed on 'rock matrix diffusion' as a retardation mechanism. Here the general distinction often made in safety case considerations that, as opposed to U deposits, the transport of radionuclides is from a deep repository in a reducing environment to oxidising conditions near the surface, appears poorly supported.

Under current plans around 6000 t of spent fuel will be disposed of at Olkiluoto (POSIVA 2003). If the uranium were to come into direct contact with oxidising groundwaters, it is reasonable to assume that its fate might be similar to that of the natural deposit at Palmottu, although the presence of engineered barriers complicates the situation. The presence of uranium in association with copper at Hyrkkölä may provide further insights and merits more detailed investigation.

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2.2 Release of radionuclides from repository

The studies concerning the release of radionuclides from a repository consist of five basic components. Copper corrosion analysis was studied experimentally with partial funding from the Swedish Nuclear Power Inspectorate (SKI). The combined creep and corrosion of canister copper was also studied experimentally with partial funding from SKI. The interaction between copper corrosion products and uranium was studied by means of a literature review. Likewise, a literature study was conducted on the freeze-thaw effects on bentonite and bentonite mixtures. The coupled behaviour of bentonite buffer (i.e. compacted bentonite) was studied within the international DECOVALEX projects, with an emphasis on coupled THM modelling.

2.2.1 Copper corrosion analysis[7]

INTRODUCTION

The failure of the copper canister as a result of corrosion may be caused by three different mechanisms, namely general corrosion, pitting corrosion, and stress corrosion cracking. On the basis of mass balance calculations, the overall extent of general corrosion would not be expected to exceed 300 μm (Laitinen et al. 2001). However, because of the possibility of the localisation of the oxidant supply, the extent of general corrosion could be much higher locally. Such localisation would be expected to rise e.g. from uneven swelling of the bentonite or uneven distribution of trapped oxygen around the disposal hole. Also, there is some uncertainty as to the rate of consumption of trapped oxygen in compacted bentonite in other reactions than copper corrosion. Thus, there is a need to develop tools to make possible the experimental verification of calculated estimates of rates of different processes competing in the consumption of oxygen in compacted fully saturated bentonite. The work reported here concerns the development of a reference electrode, a redox electrode, and an on-line corrosion sensor capable of operation in fully compacted bentonite and the effect of compacted bentonite on the general corrosion rate of copper in a final disposal vault environment for radioactive waste.

SENSOR DEVELOPMENT

In the first phase of the work an experimental facility was developed to make possible the simulation of the corrosion environment which copper will "see" in compacted bentonite after water saturation and the development of full hydrostatic and swelling pressures.

The reference electrode design used in bentonite is based on the conventional AgCl/Ag reference electrode, where the sensor is an Ag rod covered with AgCl coating placed in a container filled with KCl solution. The sensor container was further connected to the compacted bentonite container (Figure 15) via a tube 1.6 mm in diameter filled with bentonite which had been soaked in HSGW (high salinity groundwater). The bentonite which had been soaked in HSGW was found to operate as an efficient salt bridge through which the potential signal was transmitted without disturbance.

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[7] By Timo Saario, Petri Kinnunen, VTT.

The redox electrode consisted of a Pt rod partially covered with PTFE (PolyTetraFluoro-Ethylene) and placed directly in the compacted bentonite through an opening in the bentonite container. The opening for the Pt rod was sealed using a graphite box seal on the part of the Pt rod covered with PTFE.

The on-line copper corrosion rate electrode (Figure 15) was developed on the basis of the well-known wire-resistance probe principle (Saario et al. 2001). In this probe a direct current I passes through a wire made of the material under study, and the resulting voltage U is monitored as a function of time. Because U = R×I and

$$R = \rho \frac{l}{A} \qquad (1)$$

where ρ is the specific resistance of the wire material, l the length of the wire, and A the cross-sectional area of the wire. The decrease in the cross-sectional area (i.e. corrosion rate) is detected as an increase of the measured voltage signal U.

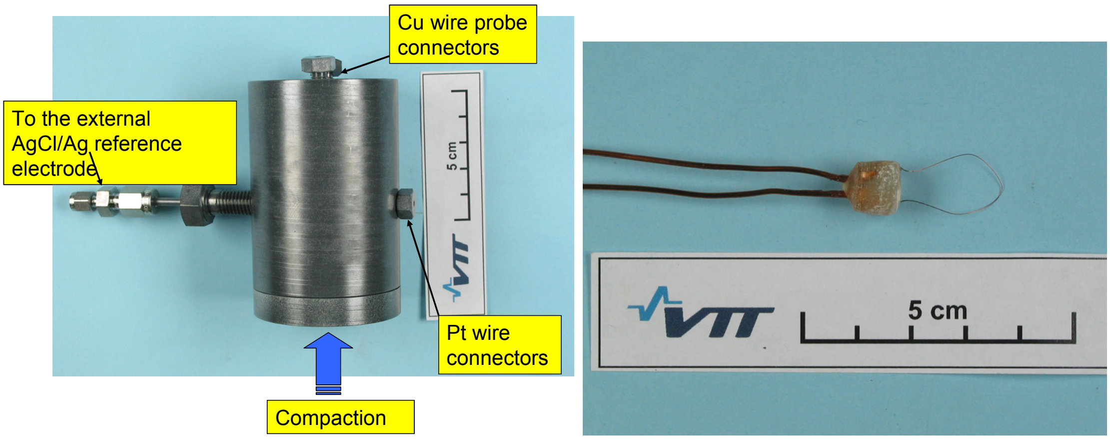

Figure 15. The bentonite container and the on-line corrosion rate probe.

EXPERIMENTAL

All experiments were carried out in highly saline groundwater (HSGW) containing 53,800 mg/l (1.52 M) of chloride (Table 2) at 80°C and a water pressure of 14 MPa. The HSGW is a brine near-field reference water representing Olkiluoto-type brine groundwater which has been in contact with bentonite (Vuorinen & Snellman 1998). The water pressure was chosen to simulate the estimated maximum hydrostatic pressure of 7 MPa (corresponding roughly to a depth of 700 m) and the maximum estimated bentonite swelling pressure, 7 MPa.

The measurements were performed in a Ti-clad autoclave inside which a smaller vessel (bentonite container made of oxidised Zr) and a Pt sheet to measure the redox potential of groundwater and a conventional pressure-balanced AgCl/Ag reference electrode were installed (Figure 16). The conventional AgCl/Ag reference electrode was used to check the stability of the modified AgCl/Ag reference electrode filled with bentonite soaked in HSGW. The Zr vessel contained the compacted bentonite, a Cu wire probe to measure the corrosion rate, two weight loss coupons, and a Pt wire to measure the redox potential in the bentonite. Oxygen-free phosphorus microalloyed copper (Cu OFP, 45 ppm P and 1.5 ppm O) was employed as the material for the weight loss coupons.

The wire resistance probe was made of Cu (99.95%, Goodfellow, diameter 0.125 mm, length 45 mm). Bentonite was mixed with the highly saline groundwater and soaked at room temperature for more than 10 h before compaction. The force used to compact the bentonite in the Zr vessel was produced using springs. A DC current of 85 mA was fed through the wire resistance Cu probe and the resulting voltage drop was continuously recorded in order to monitor the wire probe resistance and thereby the instantaneous corrosion rate.

Table 2. Composition of the highly saline groundwater (HSGW) (mg/l), see Vuorinen and Snellman (1998) for details.

Cl^-	SO_4^{2-}	Mg^{2+}	Ca^{2+}	Na^+	K^+	HCO_3^-
53800	1200	700	9900	22700	190	4.8

It is worth mentioning that in the test runs in which the compaction degree was not controlled (i.e. non-compacted bentonite slurry), the test results varied significantly from test to test and no firm conclusions on the corrosion rate could be reached. In some test runs the copper wire continued to corrode during the whole three-week exposure period. These observations demonstrate the importance of controlling the degree of compaction of the bentonite during the experiments.

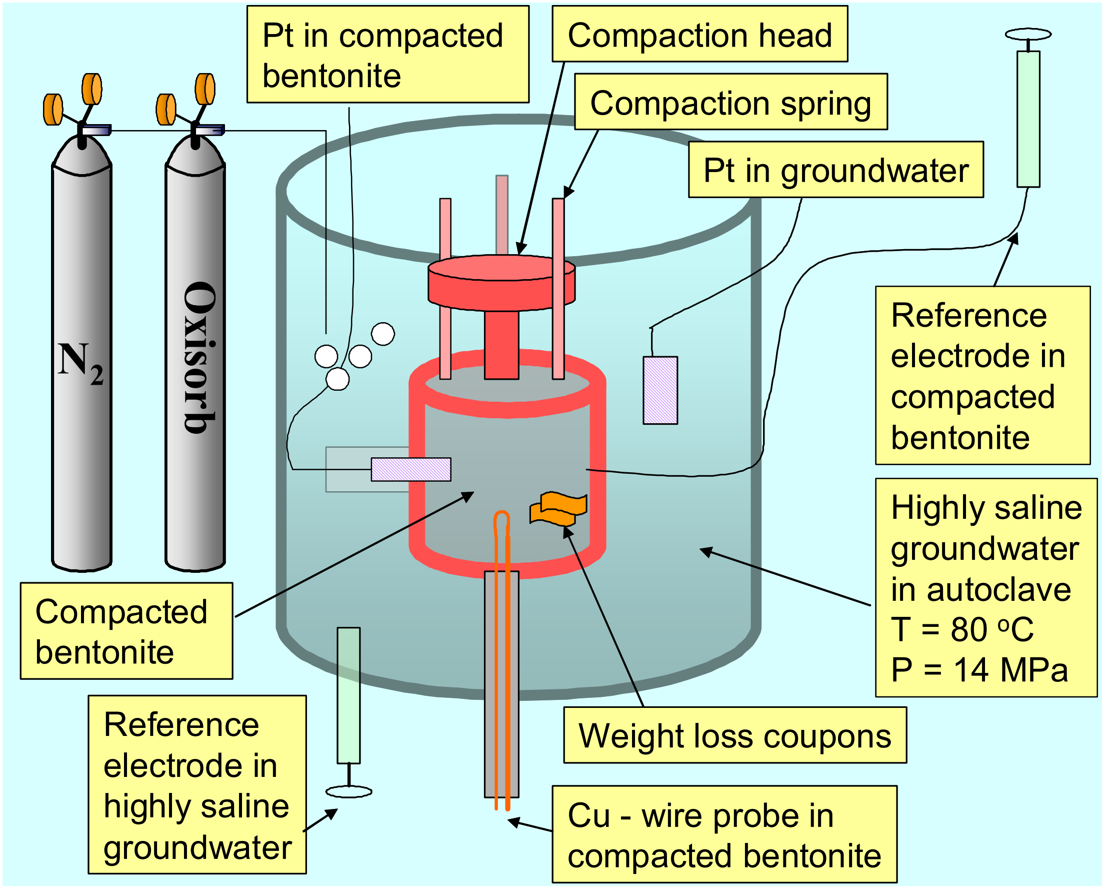

Figure 16. Scheme of the measurement set-up.

RESULTS

The <u>first 1-month test</u> in compacted bentonite was performed with a bentonite compaction pressure of 4.5 MPa. The measured corrosion depth penetration and corrosion rate of Cu are shown in Figure 17a. The potential of the Cu corrosion probe and the potential of Pt in highly saline groundwater and in bentonite are collected in Figure 17b. Results are shown vs. the modified AgCl/Ag reference pair, which is connected to the Zr vessel containing Cu samples via a salt bridge formed by HSGW and bentonite.

The depth of corrosion penetration, d_{corr}, can be estimated from the measured resistance according to the following formula (Ueno et al. 2003)

$$d_{corr} = r_0 \left(1 - \sqrt{\frac{R_0}{R}} \right) \qquad (2)$$

where r_0 and R_0 are the initial values of the radius and the resistance of the probe and R is the resistance during corrosion. Differentiating this formula with respect to time makes it possible for us to estimate the instantaneous corrosion rate as

$$\Delta_{corr} = 0.5 r_0 \sqrt{\frac{R_0}{R}} \frac{1}{R} \frac{dR}{dt} \qquad (3)$$

As can be seen in Figure 17a, the corrosion of Cu takes place mainly during the first 100 hours, resulting in a total corrosion penetration of about 0.0011 mm (1.1 µm). The latter fact means that the Cu corrosion rate tends to very low values at exposure times exceeding 100 h. After ca 200 h of exposure the depth of corrosion penetration seems even to slightly decrease, which may be caused by the redeposition of Cu. However, the signal coming from the Cu wire sensor fluctuates to some extent during the whole measurement period, which may be caused by small changes in temperature during the measurement. It can be estimated that a change of one degree in temperature changes the resistance of the Cu wire ca. 0.43%. In addition, this change in resistance would correspond to a change of ca. 0.14 µm in the corrosion depth penetration. From the results shown in Figure 17a, the fluctuations can be calculated to be of the order of ±0.07 µm, which corresponds to a change of ca. 0.5^{o} centigrade in temperature. This is typical for the accuracy of temperature control in high-temperature autoclave measurements. As a consequence of this small temperature fluctuation, the calculated instantaneous corrosion rate also shows some fluctuation. This is due to the fact that the derivative term including the change in resistance in the equation used to estimate the corrosion rate (see above) is very sensitive to temperature fluctuations. Therefore, the fluctuations in the instantaneous corrosion rate after a 100-hour exposure time can be attributed purely to temperature fluctuations and the accuracy of the calculation procedure, and the average corrosion rate is very low, close to zero.

The potential of the Cu probe in compacted bentonite with a compaction pressure of 4.5 MPa stays between -0.25 and -0.2 V vs. modified AgCl/Ag (-0.13–-0.08 V_{SHE}, potential in Standard Hydrogen Electrode scale) for the whole experiment and after the heat-up, showing a slightly increasing trend towards the end of the test (Figure 17b). According to the theoretical equilibrium calculations (Beverskog & Pettersson 2002, Chen et al. 1983) these potential values are close to the equilibrium potential of the relevant copper-containing redox system (e.g. $CuCl_3^{2-}/Cu$ redox pair). These potential values are, however, higher in comparison with our earlier tests in simulated groundwater without bentonite (Laitinen et al. 2001, Saario et al. 2001), as well as in wetted uncompacted bentonite (Betova et al. 2004). One possible explanation for this feature may be that the diffusion of ionic copper-containing species away from the wire in compacted bentonite is slower than in ground water and thus the concentration of Cu species close to the wire stays higher, resulting in a higher potential. However, the total change in the Cu wire potential, ca. 0.04 V, can be considered to be relatively small and comparable with a drift in the modified reference electrode.

The potential of Pt in compacted bentonite follows that of the Cu probe during the whole measurement period, showing values roughly 0.08 V lower than that of the Cu probe (Figure 17b). These rather high potential values for Pt indicate that some residual oxygen probably remains in the bentonite after compaction. Additionally, the existing ionic Cu species can increase the potential of Pt. The potential value of Pt in bentonite is close to that obtained earlier in wetted uncompacted bentonite (Betova et al. 2004).

It has been previously shown in pure water without Cu or any other soluble species that the purging procedure used removes oxygen from the solution very efficiently (Bojinov & Mäkelä 2003). Thus, the slow decrease in the potential of Pt in the free electrolyte (see Figure 17b) is most probably due to impurities in the solution released from the bentonite. The Pt potential reaches a value of ca. -0.35 V vs. modified AgCl/Ag, (-0.23 V_{SHE}) after 660 h, which is almost similar to that obtained in wetted uncompacted bentonite (Betova et al. 2004).

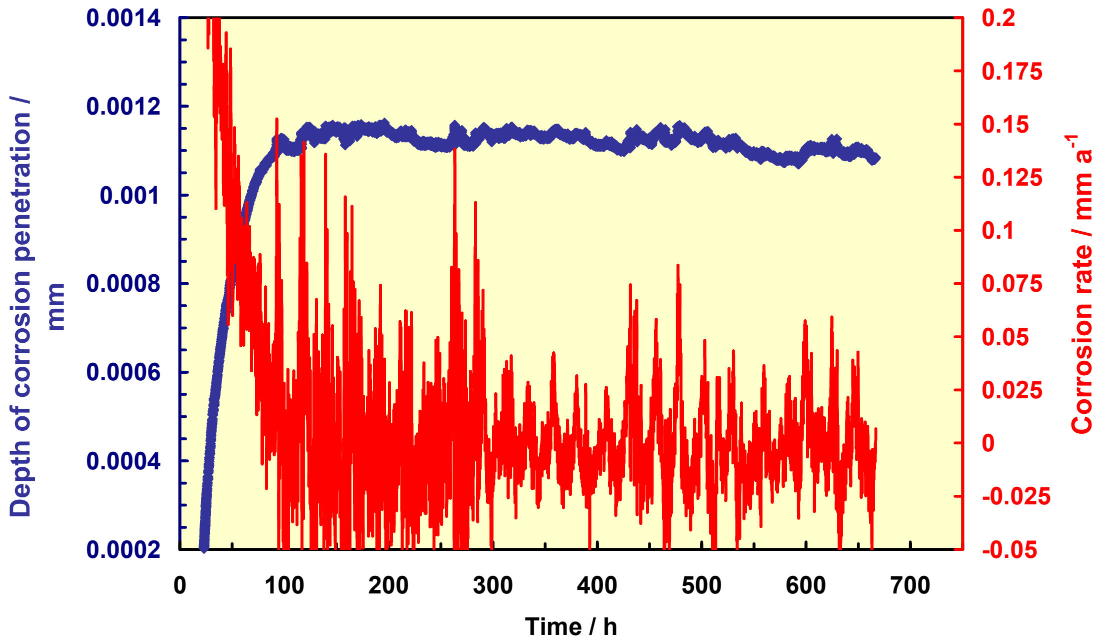

Figure 17a. Corrosion depth and instantaneous corrosion rate of Cu in compacted bentonite (degree of compaction 4.5 MPa) during a 1-month test.

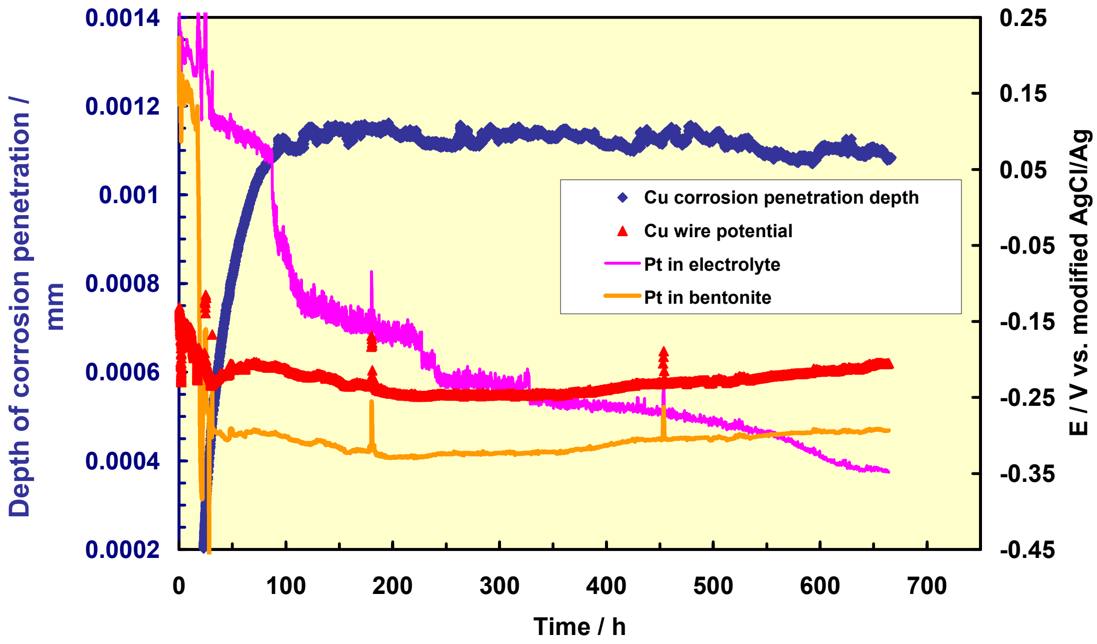

Figure 17b. Corrosion depth of Cu in compacted bentonite (degree of compaction 4.5 MPa) and potentials of Cu corrosion probe, as well as Pt in highly saline groundwater and in bentonite during a 1-month test.

The second test was performed with a bentonite compaction pressure of 10 MPa (Figures 18a and 18b). The test was carried out in such a way that the small Zr vessel in which the Cu probe and the weight loss coupons are located was opened after the first test and the coupons were weighed and re-polished, after which the vessel was closed and the compaction performed. The bentonite and the Cu wire probe inside the vessel were not changed between the measurements. During the polishing of the Cu coupons, however, oxygen was able to penetrate inside the Zr vessel and to the bentonite.

The depth of Cu corrosion penetration stabilises rather quickly after the heat-up period and reaches a level of 0.83–0.89 µm, which is roughly 80% of the level obtained at a 4.5-MPa compaction pressure (Figure 18a). The corrosion depth penetration level seems to be rather stable through the rest of the test, indicating that no further Cu corrosion takes place. This can also be confirmed by corrosion rate calculations, which show a very low average corrosion rate close to zero mm a^{-1}. Again, however, the values of the instantaneous corrosion rate fluctuate considerably as a result of small temperature fluctuations (less than one ^{o}C), which affects the measured resistance of the Cu probe and, accordingly, the differential corrosion rate calculation procedure (see above).

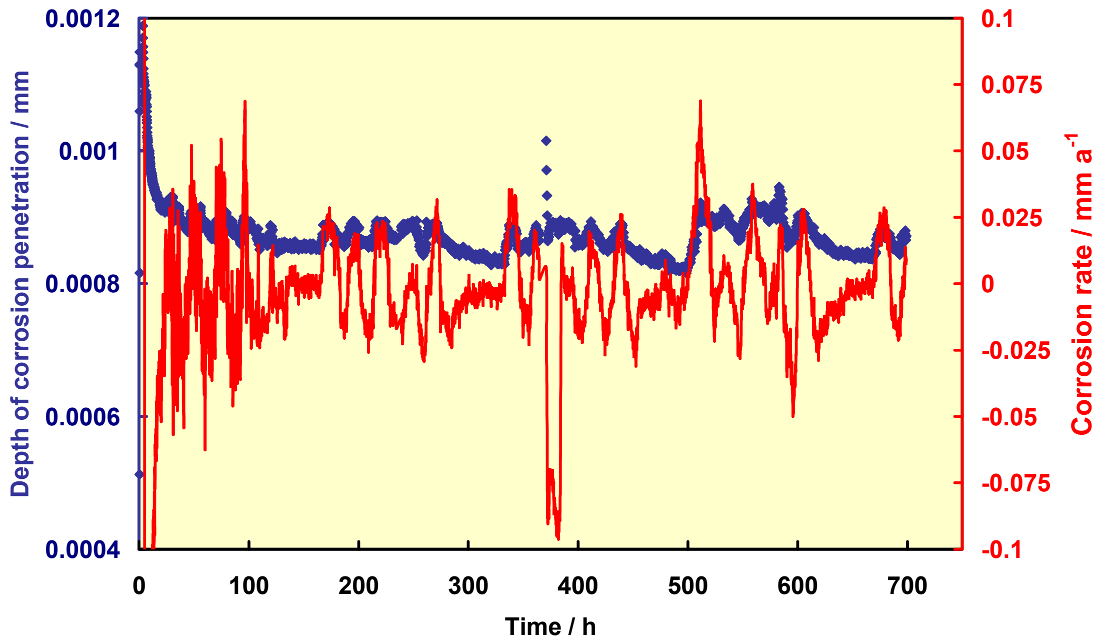

Figure 18a. Corrosion depth and instantaneous corrosion rate of Cu in compacted bentonite (degree of compaction 10 MPa) during a 1-month test.

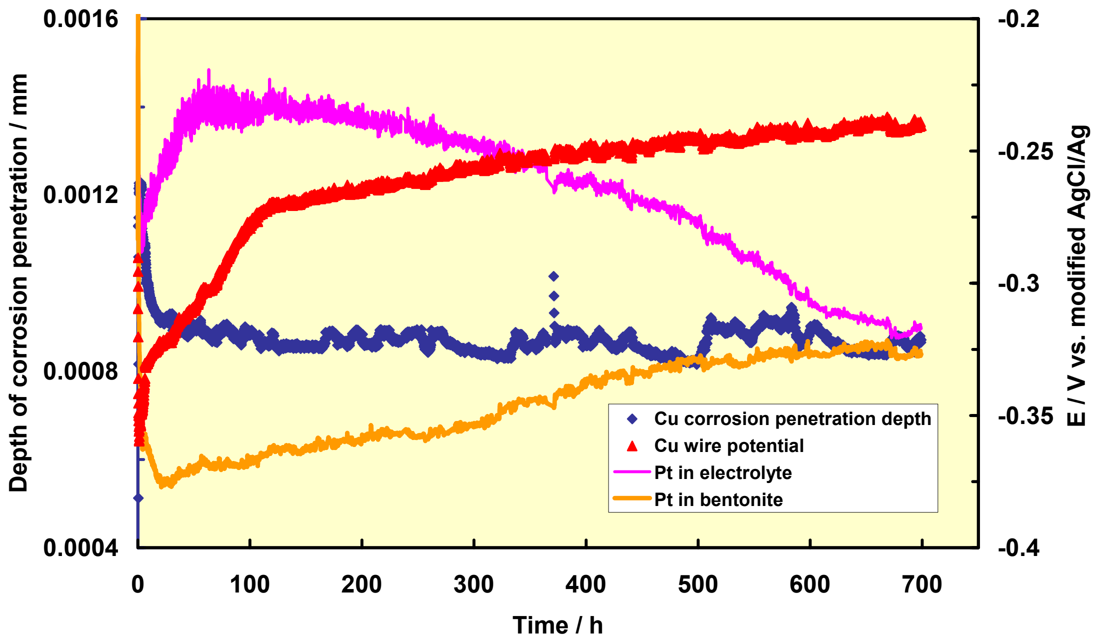

Figure 18b. Corrosion depth of Cu in compacted bentonite (degree of compaction 10 MPa) and potentials of Cu corrosion probe as well as Pt in highly saline groundwater and in bentonite during a 1-month test.

The potential of the Cu probe seems to increase during the course of the whole experiment; between 0–100 h quite rapidly, but after 100 h in a slower fashion, and it reaches the value of ca. -0.24 V vs. modified AgCl/Ag (-0.12 V_{SHE}) at the end of the test

(Figure 18b). This value is almost the same as at the end of the first measurement (see. Figure 17). The total increase in the Cu wire potential, ca. 0.03 V, is relatively small and may be explained by the slower diffusion of copper-containing species away from the wire or some other processes close to the Cu probe, which results in the higher potential of the probe.

The potential of Pt in bentonite shows a fast decrease during the heat-up period and then roughly follows the trend of the Cu probe potential, except at the beginning of the test, between 0–100h, where the rate of increase is smaller (Figure 18b). The final potential, -0.32 V vs. modified AgCl/Ag (-0.20 V_{SHE}) is almost the same as in the first test.

The potential of Pt in the free electrolyte, after a small initial increase, decreases during the course of the whole experiment, reaching a value of -0.32 V vs. modified AgCl/Ag (-0.20 V_{SHE}) at the end of the test (Figure 18b). However, the increase in the potential of Pt in the free electrolyte during and immediately after the heat-up period is much lower than in the first test and the maximum potential is ca. -0.22 V vs. modified AgCl/Ag (-0.10 V_{SHE}) obtained at around 60 h.

The <u>third test</u> (50 MPa compaction at the start) showed essentially no corrosion after the first ca. 200 h of exposure; see Figure 19a. The compaction degree was 50 MPa at the start but decreased slowly to a level of about 18 MPa during the first ca. 150 h of the test run. The reason for the decrease is suspected to be the leaching out of the wetted bentonite from the compaction test vessel. Figure 19b shows the potentials of the Cu wire probe and Pt and Ir electrodes in the bentonite, as well as the potential of the Pt electrode in the highly saline groundwater. The potentials were roughly at the same level as in the earlier tests. In this run an Ir electrode (oxidised in air to produce a surface layer of IrO_2) was added to the bentonite-containing vessel. The potential of the Ir/IrO_2 electrode should follow the changes in pH. In this run the change in potential was about $\Delta E \approx -0.06$ V, which roughly corresponds to an increase of pH by one pH unit. Such an increase might be due to the reaction of the consumption of oxygen during the first stages of the test in the cathodic coupled reaction $0.5O_2 + H_2O + 2e^- = 2OH^-$, which generates hydroxyl ions at the surface of the copper electrode. The essential stability of the potential of the Ir probe during the last stages of the experiment could be taken as indirect proof that the cathodic reaction has practically stopped.

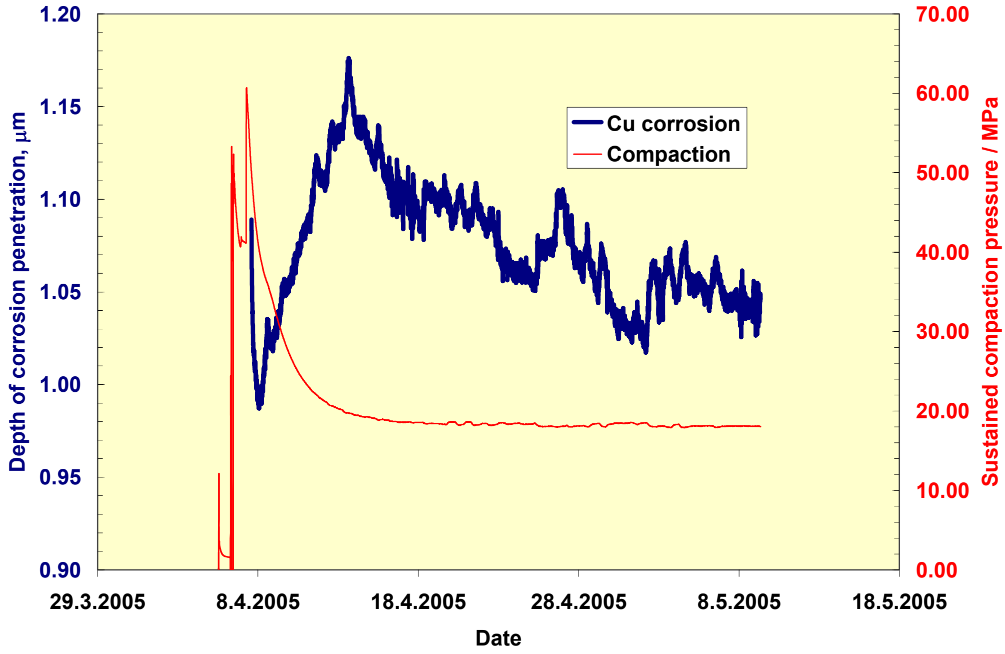

Figure 19a. Corrosion depth of Cu in compacted bentonite (degree of compaction from 50 MPa at the beginning to 18 MPa at the end) during a 1-month test.

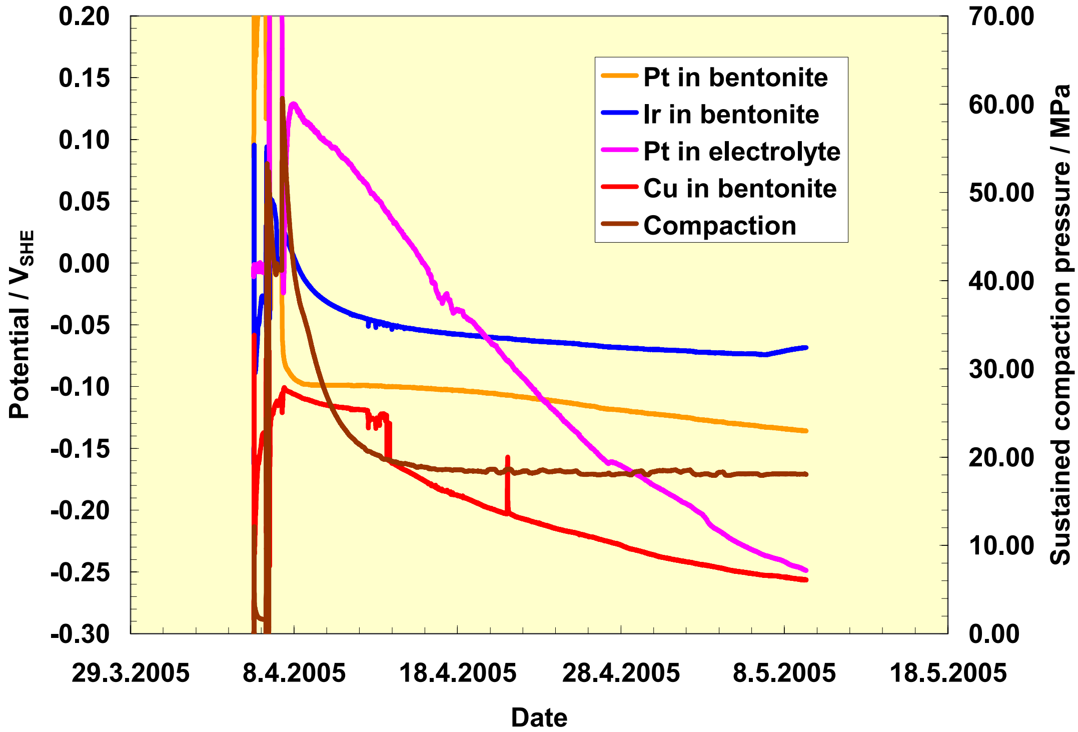

Figure 19b. The potentials of Cu corrosion probe and Pt and Ir in bentonite, as well as Pt in highly saline groundwater during a 1-month test.

Two weight loss samples were exposed to the environment in the compacted bentonite in all tests. The weight loss of the specimens showed an average of 1.0 mg (0.9 mg and 1.1 mg) in the first test (compaction pressure 4.5 MPa), 0.35 mg (0.4 mg and 0.3 mg) in the second test (compaction pressure 10 MPa), and 0.43 mg (0.5 mg and 0.35 mg) in the third test (compaction pressure 18…50 MPa). The general corrosion rate can be calculated from the weight loss using the formula

$$\Delta = \frac{\Delta m \cdot 3650}{\rho \cdot A \cdot t}[mm/a] \quad (4)$$

where Δm = weight loss [g], ρ = density [g cm^{-3}], A = surface area [cm^2], and t = exposure time [days]. The density of copper is 8.94 g cm^{-3}. The calculated corrosion rate is 0.0047 mm a^{-1}, i.e 4.7 µm a^{-1}, in the first test, 1.4 µm a^{-1} in the second test, and 1.7 µm a^{-1} in the third test. This corrosion rate is clearly higher than the 0.5 µm a^{-1} that corresponds to the corrosion of 50 mm of copper in 100,000 years. However, the most likely explanation for this result is that the corrosion takes place mainly at the beginning of the test, as shown by the on-line corrosion probe data (see Figures. 17–19). In other words, the weight loss method does not seem to be a reliable tool for the estimation of the corrosion rate of Cu in compacted bentonite.

The surfaces of the weight loss coupons were visually examined for the presence of pitting corrosion. No indications of pitting were found.

CONCLUSIONS

The general corrosion of copper in compacted bentonite wetted with highly saline groundwater (as measured with the on-line wire resistance corrosion probe developed in this project) virtually stops within ca. 100 h of the immersion of the compacted bentonite. No detectable corrosion was found during the latter part of the test periods, which were of a maximum of 700 h.

During the initial transient phase oxygen trapped in the bentonite is presumably consumed and subsequent corrosion occurs. The small fluctuations observed in the readings of the on-line corrosion probe after the transient period are interpreted as being due entirely to temperature fluctuations of ca. 0.5°C in the measurement system, the mean corrosion rate being essentially close to zero.

The corrosion results from traditional weight loss coupons, which give an integral measure of corrosion throughout the whole exposure, may be misleading and give corrosion rates that are too high.

It has been demonstrated that the corrosion and redox potentials of metallic materials in contact with wetted bentonite are influenced by a number of redox agents in the bentonite, as well as surface reactions on the copper electrode, and are thus difficult to predict or even interpret. The potential of the typical redox electrode, a Pt electrode, can thus not be used as a reliable indicator for the presence of oxygen in an environment influenced by bentonite and dissolved Cu species.

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2.2.2 Combined creep and corrosion of canister copper[8]

INTRODUCTION

The copper overpack of the canister is a release barrier, mainly to protect the canister against corrosion. Heating up to about 100–120°C can be expected in the repository, and this may result in the slow creep of copper under the accommodation stresses of the canister. On the other hand, the groundwater environment of the flooded repository may introduce some potential for simultaneous corrosion. Even if either of the two damage processes alone were not to seriously challenge the integrity and life of the protective overpack, this does not exclude damage caused by combined action in case of some synergistic mechanisms, for example by locally reduced creep strength caused by vacancies produced by corrosion. Additionally, the previous studies on the creep of copper have been short-term only, and the required long-span extrapolation greatly exceeds the limits usually accepted for conventional creep design or materials acceptance. Further complications may appear from materials scatter, defects, welds, and other features of metallurgical and geometric variation, and from the fact that current materials models and experimental experience concentrate on ranges of temperature, stress, and time other than those expected in the repository.

To partially compensate for the inevitably shorter testing time in the laboratory than in repository service, accelerated testing is widely used. However, such testing could also introduce bias if the damage mechanisms in the specimens do not correspond to those in the repository. For example, creep is accelerated by elevated stress and temperature, but an excessive increase in these quantities can easily lead to unrealistic predicted (extrapolated) creep life. Test acceleration without undue elevation of temperature or (reference) stress can be introduced by increasing multiaxiality into the stress state, and this has been shown to help in producing typical creep mechanisms (damage) within a much shorter time than is the case with any other known method (e.g. Auerkari et al. 2003). Comparable acceleration is less meaningful for corrosion, which, as a stand-alone mechanism, is also considered elsewhere, but is here of interest as a potential compounding factor for creep damage.

In theory, multiaxiality effects could also occur in the canister if the copper overpack includes weld defects. In the event that such defects are open to the surface, metal surfaces could be simultaneously subjected to corrosion in the groundwater environment, provided that such mechanisms are active.

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[8] By Pertti Auerkari, VTT.

The investigation particularly aimed

- to determine extended creep behaviour for both representative base metals and welds (friction stir) using uniaxial and multiaxial techniques for modestly accelerated testing (150–175°C for more than 10,000 h) for improved reliability of creep damage and life assessment
- to produce improved materials models for the creep of copper, using uniaxial and multiaxial experiments, finite element analysis, and microstructural evaluation of the specimens (including welds)
- to develop and verify equipment for testing the effects of combined creep and corrosion under a pressurised simulated groundwater environment in a model copper vessel and assess the effects of combined mechanisms according to the test results
- to utilise the combined information from experiments, materials analyses, and modelling for life assessment under combined creep and corrosion.

CREEP STRENGTH OF WELDED COPPER

The parent (cylinder) material and friction stir welds were tested mainly for creep rupture strength, the parent materials also with recorded creep strain. Cross-weld creep strength with specimens extracted in the axial direction of the canister appears to show no significant difference to earlier results (Andersson et al. 2005) for similar parent material of axially extracted cross-weld specimens. However, the radial cross-weld specimens exhibited considerable reduction in creep strength and time to failure (Figure 20). This strength reduction is mainly attributed to relatively small-scale weld defects (Figures 21–23), and related small grains and impurities (Figure 24). The defects are lack of fusion-type defects at the root of the FSW weld, parallel to the cylindrical (axial-tangential) surface of the joint. The stress levels of the radial cross-weld specimens in Figure 20 are nominal stresses for Ø 8–10-mm uniaxial specimens. Assuming that the actual stress is simply elevated by the loss of cross-section related to the defect (net section stress by a correction factor of 0.80–0.85), the corresponding data points in Figure 20 for actual stress would approach the level of short-term parent material and axial cross-weld tests of FSW welds. However, the pronounced downward slope for radial cross-weld creep tests would remain, suggesting significantly decreasing creep strength in longer-term testing. The width of the weld defect in the failed radial cross-weld specimens was similar (about 1.5 mm), but the positions of the defect varied somewhat between the specimens. This variation can be at least partially accounted for in the estimated net section stresses, although naturally it is as yet unknown for the remaining running test specimen. It has been suggested that the actual stresses in the overpack are no more than about 40 MPa (Raiko 2005).

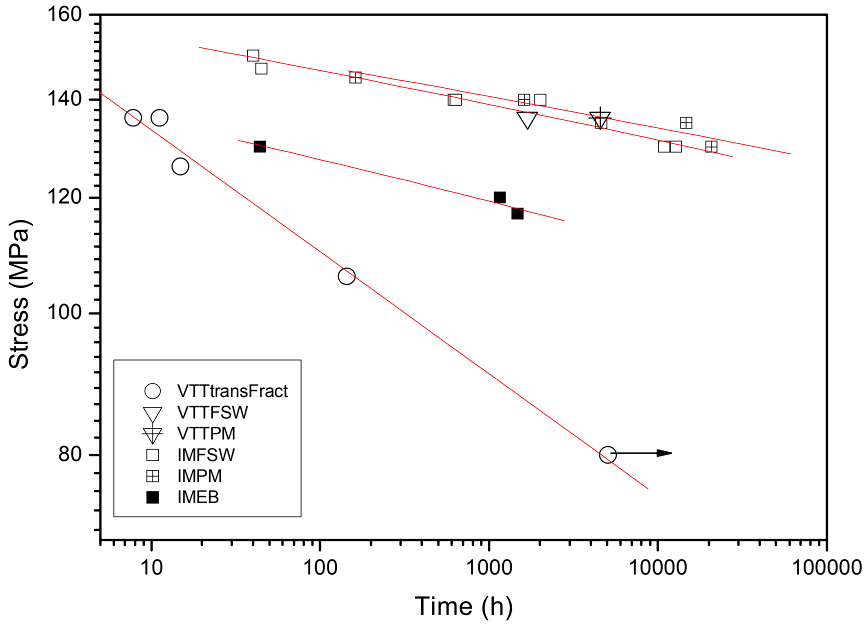


Figure 20. Creep strength at 175 ℃ for parent material and axial cross-weld specimens (upper lines with data from this work and from Lindblom et al. (1995), and radial cross-weld specimens (open circles, this work).

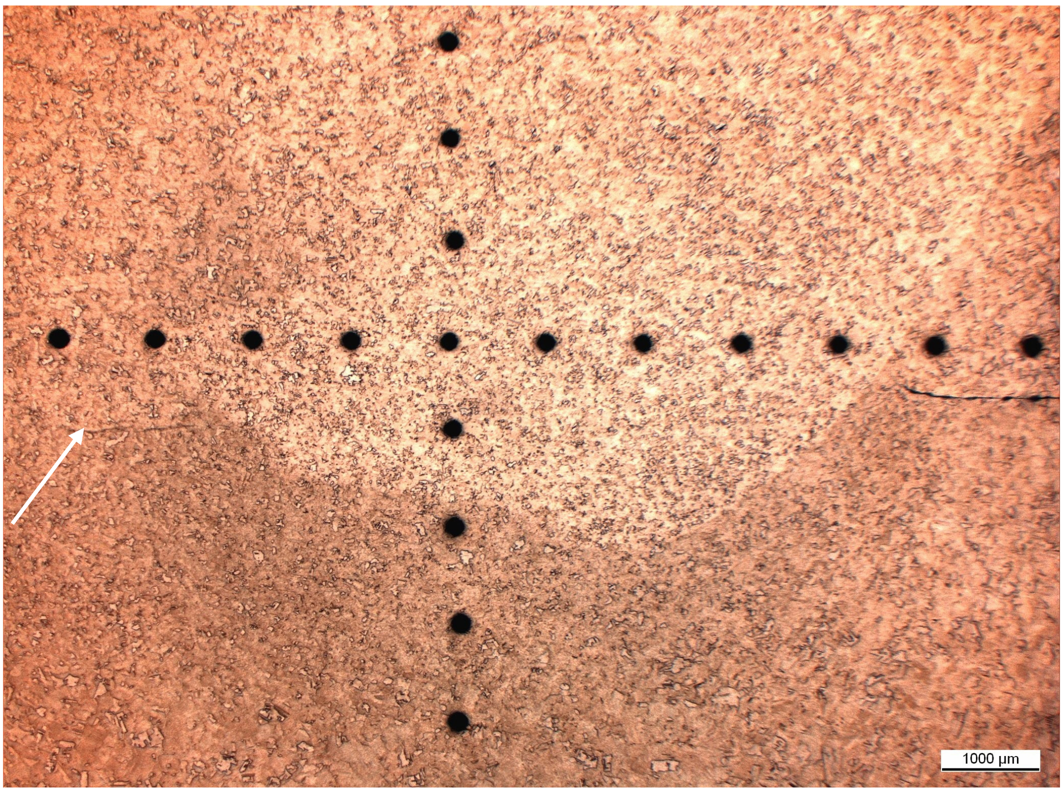


Figure 21. Weld defect (arrow) at the root of the FSW weld along the axial plane of the cylinder-cover joint. Tip of the joint gap is on the right below the row of hardness indentations.

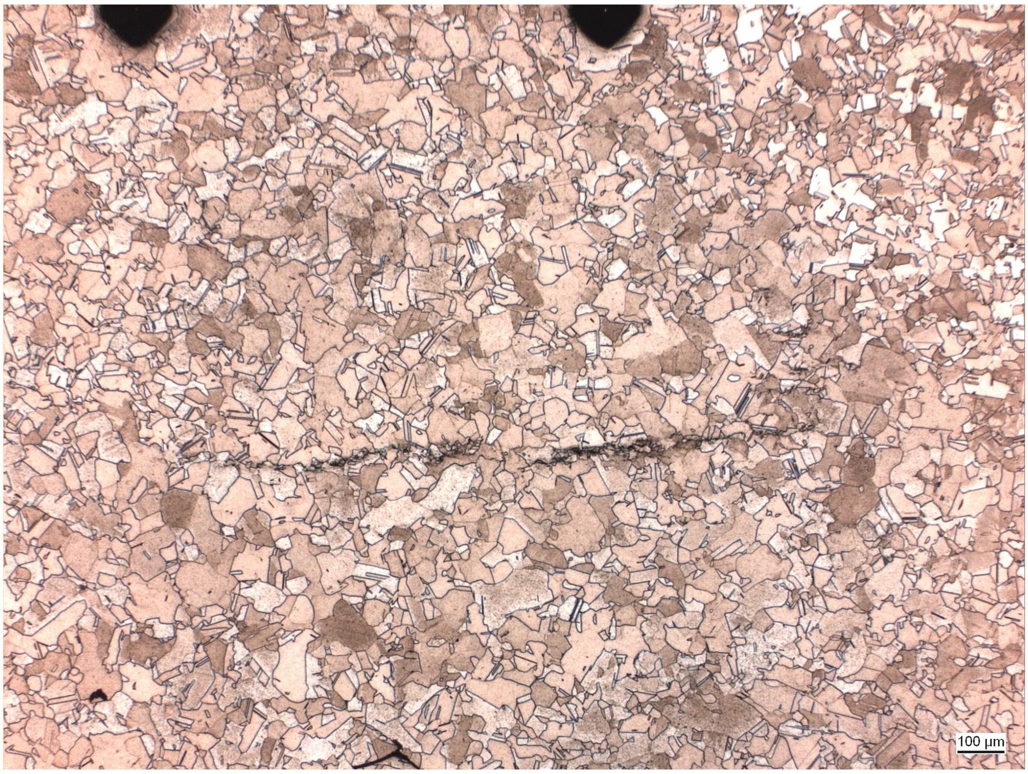


Figure 22. Detail of the weld defect of Figure 21.

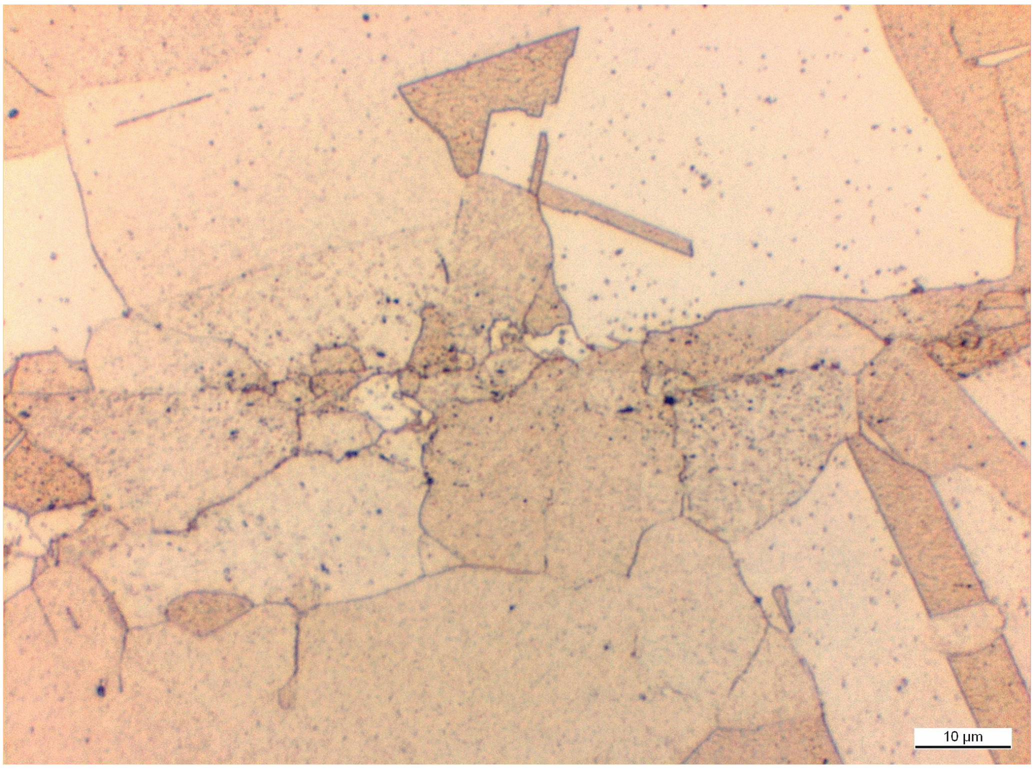


Figure 23. Detail of the weld defect of Figure 22. Note small grain size and apparent inclusions along the defect zone.

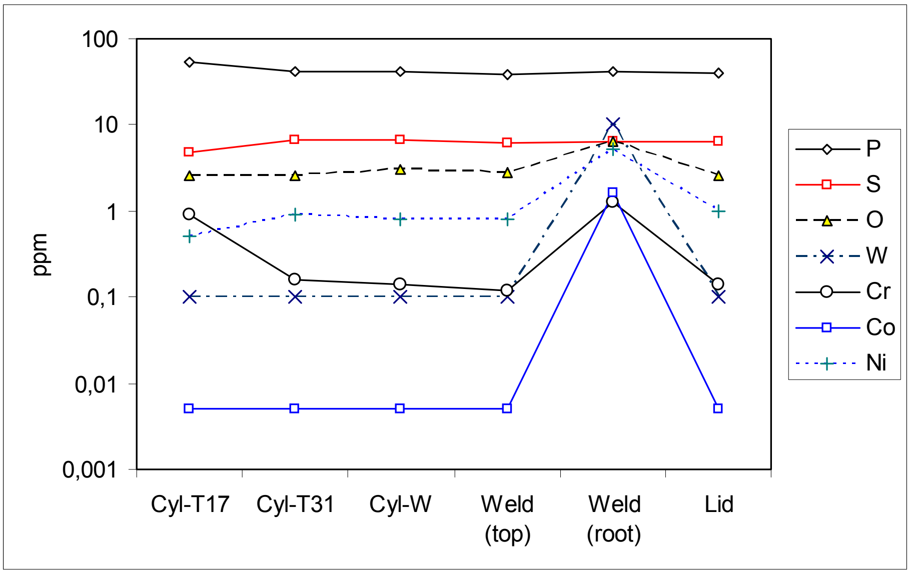


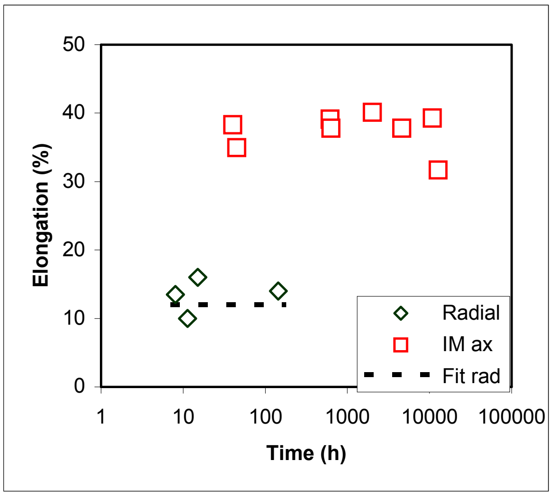
Figure 24. Chemical composition of parent materials (previously tested cylinder sections T17 and T31), welded cylinder W (= T31), and cover lid, and weld metal from the top and root sections.

None of the weld defects were observable in usual non-destructive testing (visual, dye penetrant, and ultrasonic inspection) before creep testing. After creep testing, however, the defects appeared on the failure surfaces as distinct through-thickness flat zones about 1.5 mm wide (Figure 25).

a) b)

Figure 25. Defect zones (arrows) in the fracture surfaces after creep testing of radial cross-weld specimens: a) VT3 (126 MPa/175 °C/15 h); b) VT4 (106 MPa/175 °C/144 h). Scale bar 1 mm.

Reduced creep strength is also reflected in creep rupture strain (elongation), which lies well below the level expected for parent material or axial cross-weld specimens (Figure 26a). The reduction in ductility even in short-term testing is thought to be related to the combined effect of increased net section stress, stress concentration, and metallurgical discontinuities at the weld defect, which also result in a rather jerky creep flow (Figure 26b). Discontinuities such as a zone of small grains and inclusions/impurities appear similar to those before testing (Figure 27, cf. Figure 23).



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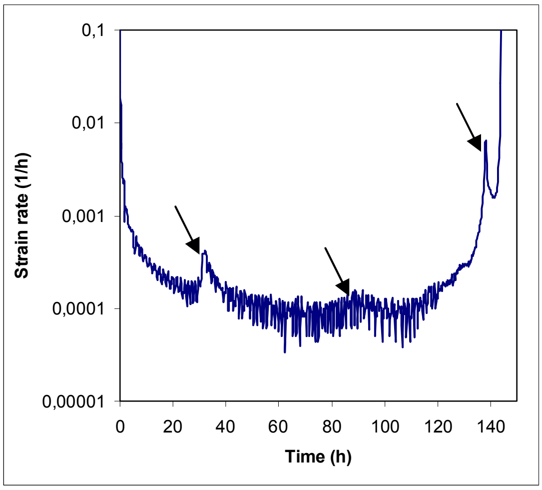

b

Figure 26. a) Creep rupture strain in radial and axial cross-weld specimens; the trend line for the latter refers to parent material behaviour from Lindblom et al. (1995); b) jerky creep strain in specimen VT4 (126 MPa/175 ℃), apparently resulting from discontinuous defect growth.

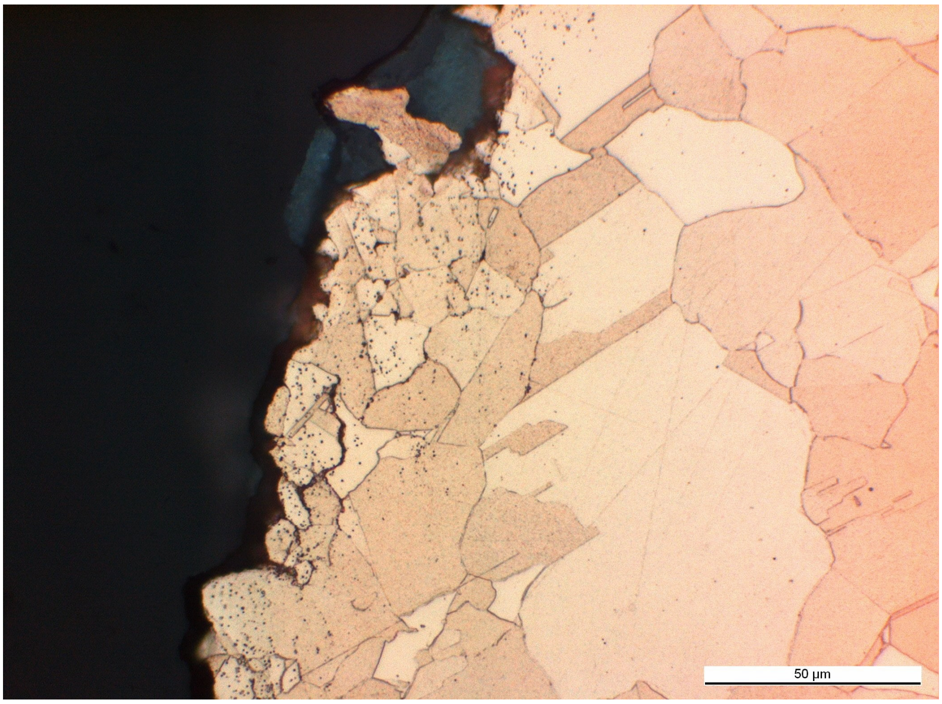

Figure 27. Cross-section of the failed specimen VT2 (136 MPa/175C) at the weld defect; note small grains and inclusions next to the fracture surface.

Periodically interrupted creep testing of compact tension specimens, incorporating an in situ notch from the FSW welded joint gap, was performed up to 5,000 h without any observed damage indication other than strain (load line displacement). This testing continues for modelling and extended life assessment purposes, as well as uniaxial creep testing of parent material and welds. In uniaxial creep testing of parent material, the longest (continuing) testing time exceeded 36,000 h, and for axial cross-welds 15,000 h. These test materials will be used to readdress the earlier observations on apparent thermal degradation and changes in the grain boundary microstructures at relatively low testing temperatures (Auerkari et al. 2003, 2005a, b).

Weld defects at the root of the FSW weld are not expected to have a significant safety impact because of the position and orientation of the defects deep inside the component. The conceivable effects of reduced creep strength would more likely require weld defects at or near the outer surface of the weld. If such a defect were open to the surface, it would also be subjected to the simultaneous effects of the groundwater environment and potential combined effects of creep and corrosion. However, no such defects were seen in the weld tested in this work, and the cross-weld creep strength in the axial direction appears to be comparable to that in the parent metal and previously tested FSW welds.

COMBINED CREEP AND CORROSION IN GROUNDWATER

A tubular copper specimen (Ø 47.5 x 9.5 mm) with artificial internal (axial-radial) defects was tested under internal pressure using oxygen-free simulated groundwater as the pressurising medium. The testing arrangement is shown in Figure 28, and the chemical composition of the medium in Table 3. This medium represents a groundwater (Olkiluoto bentonite equilibrated brine) with a relatively high salt content. The defects extended across both parent material and the weld, in diametrically opposing tube positions, to maximum depths of 1.8 and 1.5 mm. The nominal mid-wall hoop stress was 39.6 MPa and the von Mises skeletal stress (n = 6) 33.8 MPa at an overpressure of 20 MPa, with corresponding stress intensity (K) values (Yoon et al. 2004) of 5.4 and 4.8 $MPa \cdot m^{0.5}$ respectively at the two defects. The anoxic condition of the test environment was monitored on-line with an AgCl/Ag reference electrode (LDR) during testing. The oxygen-free condition was confirmed up to the first 100 h, until the electrode was grounded as a result of chloride-rich water penetrating the electrode lead-through. The test was discontinued after 795 h of testing because the pressure line was blocked by salt deposits at the furnace to ambient interface. The testing medium was cloudy, with a dark mud-like residue that was found to contain iron (0.11 mg/g) and copper (0.23 mg/g). Clearly, the testing conditions were sufficient to dissolve both steel and copper components of the test system. After testing, no macroscopic deformation of the specimen was observed, so that at least the intended loading well below the FEA predicted limit load (Figure 29) was achieved. The predicted and actual limit loads will be compared after further step pressure testing of the specimen. The results are used to tune the loading conditions for longer-term testing, which is necessary for assessing life under the combined effects of creep and corrosion.

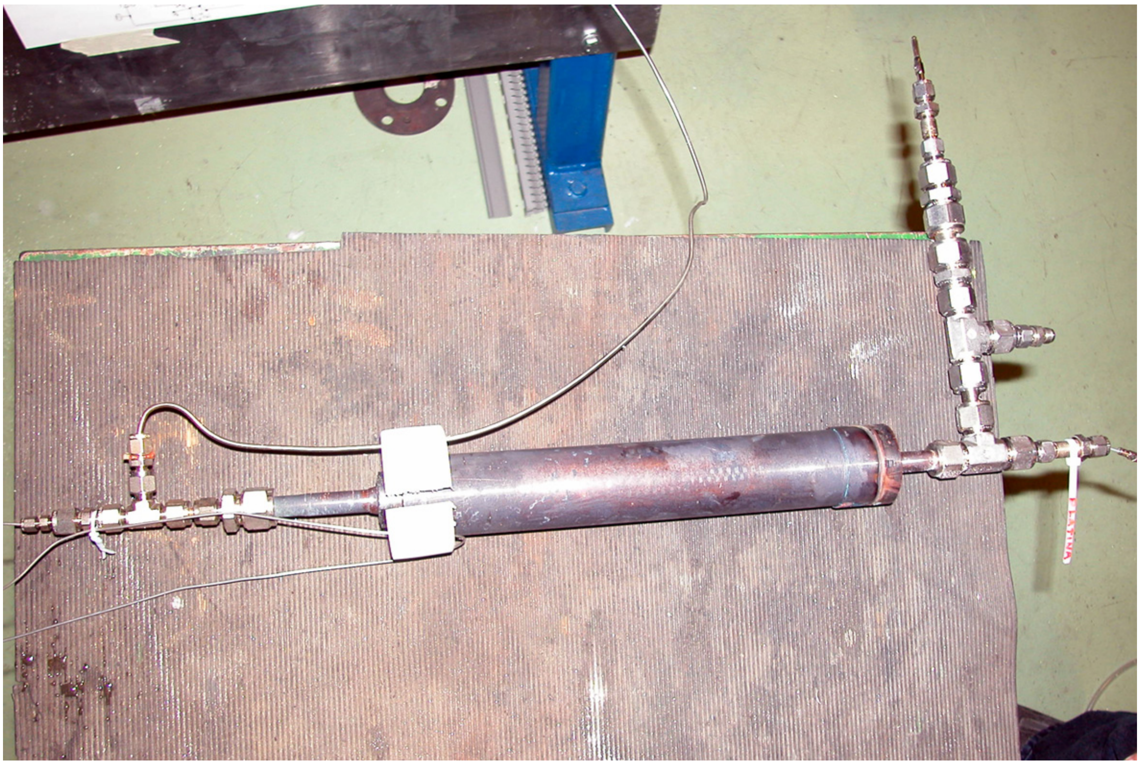

Figure 28. Combined creep and corrosion testing by internal pressure using simulated groundwater (795 h at 175°C/ 200 bar overpressure).

Table 3. Composition of the groundwater (mg/l) used in the test.

Species	Cl^-	SO_4^{2-}	Mg^{2+}	Ca^{2+}	Na^{2+}	K^-	HCO_3^-
mg/l	53800	1200	700	9900	22700	190	4.8

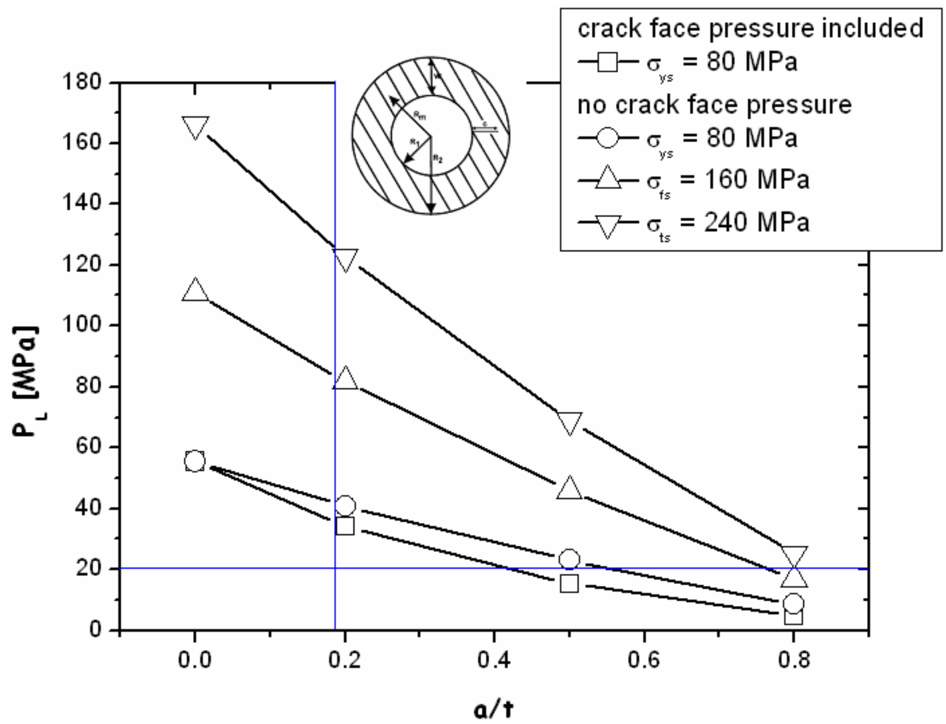

Figure 29. Predicted limit pressure as a function of crack depth to wall thickness at 175 °C (FE analysis). At 20 MPa test pressure and 1.8 mm defect depth, the first test is predicted to remain well below the limit load.

CONCLUSIONS

The long-term creep performance of oxygen-free phosphorus-doped (OFP) copper for the overpack of waste canisters was considered in a series of experimental studies. The principal aim was to assess creep behaviour and the potential combined creep and corrosion effects of this copper material under the foreseen repository conditions. The expected service life of about 100,000 years continues to pose technical challenges for life prediction, because of the difference of about four orders of magnitude between the service life and conventional range of testing times. Extensive extrapolation in time may result in non-conservative life prediction, i.e. predicting a longer life than what is sustainable in reality. Further complexity can be expected from loading at relatively low temperatures and variability in terms of e.g. grain size, welding, and defects.

Greatly reduced creep life was observed when testing across the weld root in the radial cross-weld direction, in comparison with the cross-weld performance in the axial direction of the overpack cylinder. Metallographic investigation suggests that the reduction is associated with small-scale discontinuous lack of fusion defects in the root region of friction stir welds. Weld defects at the root of the weld are not expected to have much safety-related impact because of the position and orientation of the defects deep inside the component. The conceivable effects of reduced creep strength would more probably require weld defects at or near the outer surface of the weld. If such a defect were open to the surface, it would also be subjected to the simultaneous effects of the groundwater environment and potential combined effects of creep and corrosion. However, no such defects were seen in the weld tested in this work, and the cross-weld creep strength in the axial direction appears to be comparable to that of the parent metal and previously tested friction stir welds.

The longest creep tests exceeded 36,000 h for parent material and 15,000 h for cross-weld testing, to support creep modelling for life prediction and for accelerated multiaxial testing. The significance of previously observed stress-enhanced microstructural changes at relatively low temperatures remains poorly understood.

Tubular copper specimen with internal axial-radial defects within both the parent material and the friction stir weld was tested under internal pressure, using oxygen-free simulated groundwater (OL brine) as the pressurising medium. The anoxic condition of the test environment was monitored on-line during the testing. The test was interrupted after pressure line blockage by salt deposits at about 800 h of testing, requiring modification for further experiments. After testing, no macroscopic deformation of the specimen was observed, confirming loading below the FEA predicted limit load. The observed copper content in the testing medium after testing was approximately in accordance with the expected rates of copper dissolution resulting from general corrosion. Further and longer-term testing is necessary for assessing life under the combined effects of creep and corrosion.

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2.2.3 Interaction between copper corrosion products and uranium[9]

INTRODUCTION

Before the selection of the Olkiluoto site for the construction of the spent fuel repository in Finland, the occurrence of sulphide in groundwater in a reduced environment was considered the major threat to the integrity of the copper canister (e.g. Ahonen 1995 and references therein). Later on it was shown that copper corrosion products after sulphidation can retain radionuclides (at least U), even in oxidising conditions (Marcos & Ahonen 1999, Marcos 2002). The selection of the Olkiluoto site emphasised salinity (Vieno 2000) as a new major threat to the performance of the technical barriers. King et al. (2002) estimated that the corrosion of copper in repository conditions (saline and reduced) is quite improbable but not impossible. They pointed out that one corrosion product in such conditions is most probably cuprous chloride, CuCl, a mineral phase named nantokite. The role of nantokite as a possible sink for radionuclides is not known.

This work is the result of a review of copper and uranium mineralisations or deposits. The aim was to search for the simultaneous occurrence of uranium and copper in natural saline and reducing conditions in order to gain knowledge of the interaction of copper corrosion products and uranium in such an environment.

Special attention was paid to the mode of occurrence of copper in saline environments in order to check the conditions in which nantokite occurs. The occurrence of uranium in saline environments may or may not be linked to the occurrence of copper. In this report, attention is paid to the first case and the same examples are mentioned for the last case.

[9] By Nuria Marcos, TKK.

COPPER IN SALINE ENVIRONMENTS

Some of the native copper deposits in the Keweenaw Peninsula region of Lake Superior, USA, have been in contact with connate brine waters (Crisman 1982) in which the chloride concentration varies between 45 and 180 g/L. The conditions were oxidant and the products of the alteration of native copper included a variety of copper minerals (Table 4).

Table 4. Mineral products of alteration of native copper.

Copper oxides	Cuprite Tenorite	Cu_2O CuO
Copper chlorides	Atacamite Nantokite	$Cu_2(OH)_3Cl$ $CuCl$
Copper silicates	Chrysocolla Plancheite	$CuSiO_3$•$2H_2O$ $Cu_8Si_8O_{22}(OH)_4$•$2H_2O$
Copper carbonates	Malachite Azurite	$Cu_2CO_3(OH)_2$ $Cu_3(CO_3)_2(OH)_2$

Later, in a study of copper mineral phases in the mining wastes/tailings of the Keweenaw Peninsula (Jeong 2003), no copper chloride minerals were reported but oxides (cuprite and tenorite), carbonates (malachite), silicates (chrysocolla), and sulphides (chalcopyrite, covellite) were.

In the deepest level (500 m) of the Corocoro Mine (Bolivia), the chloride concentration was around 70 g/L (Entwistle and Gouin 1955). The main ore minerals were native copper and chalcocite and the corrosion products mentioned are azurite, malachite, and cuprite near the surface (see also Marcos 1989 and references therein).

Nantokite and other copper chloride minerals were first found in Nantoko in the Atacama Province, Chile, where sulphide deposits are abundant.

Nantokite is also mentioned in the USA in the Algomath mine, Michigan, in Bisbee, Arizona, in Balmat, New York, in the UK in the Levant Mine, Cornwall, in Mexico in the Ojuela and Piedra mines, in the Czech Republic in the Nový Martin Gallery in Bohemia and in Australia in New South Wales and Queensland. Unfortunately, nothing has been reported about the groundwater chemistry of these sites.

In the field of archaeology, nantokite may precipitate under a former cuprite layer over metallic copper or a copper alloy (McNeil and Little 1992). A biofilm containing acid-producing bacteria could help produce the required conditions by increasing acidity in anodic areas and reducing copper ion transport from the surface, resulting in a higher local copper concentration (Mohr and McNeil 1992).

Copper occurs in sedimentary and saline environments, but mainly as sulphide, along with other heavy metals. Rose (1989) reviewed the low-temperature chemistry of copper and associated elements in sedimentary environments. The role of chloride is mainly as a complexant of copper, forming mostly cuprous complexes, as is also pointed in King et al. (2002). Thus, the cuprous chloride complexes are the most important complexes for transporting Cu to form copper deposits in sedimentary basins, and especially where these complexes encounter reducing conditions over a wide pH range. In a concentration of chloride of around 35 g/L the cuprous complexes may be stable to pH values as high as 10. Some examples of sediment-hosted stratiform copper deposits are listed in Table 5.

Table 5. Examples of sediment-hosted stratiform copper deposits (after Rose 1989).

Deposit	Major metals	Minor
Kamoto, Zaire	Cu, Co	Ag, Au
Mufulira, Zambia	Cu	Co, Zn
Redstone River, Canada	Cu	Ag
White Pine, USA	Cu, Ag	Pb, Zn, Cd
Spar Lake, USA	Cu, Ag	-
Corocoro, Bolivia,	Cu	Ag
Kupferschiefer, Poland	Cu, Ag, Pb, Zn	Co, Au, Pt

Only a few stratiform copper deposits contain appreciable amounts of Co, Pb, Zn, U, or other metals, as indicated in Table 5. Rose (1989) examined the solubility of these other metals, but not of uranium, and there is no mention of the mode of occurrence of uranium in these deposits.

Experiences in the field of archaeology

First Bresle et al. (1983) and later King (1995) made an extensive study of the corrosion processes that affected a bronze cannon found within clays at a shallow depth in the Baltic Sea. These studies are not reviewed further in this report because no copper chloride minerals were found associated with the case.

Scott (1990) reviewed the chemical problems related to the study of bronze disease. Bronze disease is defined as the process of interaction of chloride-containing species within the bronze patina with moisture and air, often accompanied by corrosion of the copper alloy itself. Cuprous chloride forms during the corrosion process in burial. It is not usually exposed to the naked eye but present as a corrosion product, often close to the surface of the residual metal. One of the problems in studying the nature of the corrosion products is the light-sensitivity of e.g. cuprous chloride. It should be kept in a vacuum desiccator to prevent chemical changes. Thus, cuprous chloride, or nantokite, is seen as a relatively stable and insoluble compound if unexposed to water and light.

In the same way that cuprite is thought to be the first copper corrosion product for the canister in the evolution of the spent fuel repository, Organ (1963), Scott (1990), and McNeil and Little (1992) mentioned that cuprite is also the first product of copper corrosion for bronzes in saline waters. Cuprite has a high electrical conductivity and permits the transport of copper ions (North and Pryor 1970) through the cuprite layers, allowing the copper ions to dissolve in water and reprecipitate. If the water chemistry resembles that of seawater and not much dissolved O_2 is available, then cuprous chloride, nantokite, precipitates under the cuprite layer (Scott 1990).

CU-U MINERALIZATIONS

The source of chloride for Cu and Cu-U mineralisations

In a review of metal-evaporite association Warren (2000) showed that most subsurface evaporites ultimately dissolve and, through their ongoing dissolution and alteration, create conditions suitable for metal enrichment and entrapment. In a typical basinal brine (TDS 200 g/L), copper mobilisation is prevented if the oxidation state in the brine is maintained near that of the hematite-magnetite buffer, as the solubility of Cu in such brine ~ 0.07 mg/L (Sverjensky 1989; Warren 2000). Redox conditions in the brine of $\log f_{O2} > -46$ at 125°C are necessary to mobilise copper as cuprous chloride complexes. These authors also state that acidification is an essential step in the acquisition of metals during the dissolution of aquifer minerals and that chloride-rich, high-salinity basinal fluids may became acidified by pyrite oxidation, by the decomposition of organic matter, or by the precipitation of Mg silicates, such as smectite or chlorite. Eugster (1985, 1989) argued that not all metalliferous brines are acidic and that metals can be carried as hydroxy or carbonate complexes up to pH ~ 11.

The intimate association of U and Cu in some sandstone-hosted U deposits and volcanic-hosted Cu deposits indicates that these metals can be moved and deposited together in such ore-forming systems, despite the fact that major economic concentrations of Pb, Zn, and U tend to occur in separate sediment-hosted deposits rather than directly with Cu (Kirkham 1989).

USEFULNESS OF POSSIBLE LAB EXPERIMENTS WITH NANTOKITE AND URANIUM

Because no natural occurrences were found of the intimate association of nantokite or other Cu chloride minerals and uranium in saline and reducing conditions, a laboratory experiment could in principle be performed in strictly reduced conditions with the pure mineral phase nantokite and uranium. But the usefulness of such an arrangement is questioned since:

1. No natural occurrences have been observed either on the interactions of nantokite and uranium or on their coeval existence in the same mineral paragenesis for saline and reducing condition, so will nantokite form at all?
2. In the repository bentonite will be in close contact with the copper canister, i.e. it should be known in the first place what the bentonite-copper interaction products are in order to confirm the relevant corrosion products of copper.

There are ongoing laboratory experiments with copper and bentonite in conditions simulating the ones at Olkiluoto (Aalto et al. 2002), but the final copper corrosion products will not be reported until the end of 2011. Kinnunen et al. (2003) evaluated the corrosion rates of copper in contact with bentonite in Olkiluoto-type saline groundwater. Corrosion occurs indeed, but the nature of the corrosion products is not reported. The corrosion of copper in anoxic saline water (Bojinov & Mäkelä 2003) has been studied in recent laboratory experiments. Copper is corroded and gets into solution, although in aging the experiment the back deposition of copper is observed. The corrosion products of copper, if any, are not mentioned in this study.

With respect to the interaction of uranium with saline waters, complexing between uranium and chloride ions in aqueous media has received scant attention. However, knowledge on the interactions of brines with radioactive wastes is important for understanding and predicting uranium mobility in the natural environment. Uranyl-chloride complexes are thought to be very weak and the only species currently accepted are the UO_2Cl^+ and $UO_2Cl_2^0$ complexes (Bailey et al. 1998). Thus, it seems that the coexistence of copper chloride and uranyl chloride complexes is not very probable. In fact, not a single mineral has been found in nature containing Cu, U, and Cl in its lattice, whilst hydrated copper uranyl silicates (e.g. cuprosklodowskite), phosphates (torbernite), and arsenates (metazeunerite) are relatively abundant.

CONCLUSIONS

Studies of copper corrosion under repository conditions have been going on since 1977, but not much attention has been paid to the nature of the corrosion products and even less to the possible interaction of radionuclides with copper corrosion products.

This study attempted to clarify the interactions of copper corrosion products, especially nantokite, with uranium through a literature review in Cu and Cu-U occurrences in saline and reducing environments. It was found that in such environments uranium occurs mainly as pitchblende or uraninite and copper as copper sulphides. Furthermore, not a single mineral compound containing Cu, U, and Cl is mentioned in the literature.

The competitiveness of the compounds involved in the release from technical barriers may hamper the formation of copper chloride compounds that would otherwise form, i.e. without the bentonite barrier. In any further experiments dealing with copper corrosion, more attention should be paid to the nature of the copper corrosion products in order to get a better idea of the overall evolution of the repository system.

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2.2.4 Freeze-thaw effects on bentonite and bentonite mixtures[10]

A literature study was conducted on the influence of freeze-thaw on the permeability of bentonite and bentonite mixtures (Saarelainen 2002). In the following section an abstract of the study is given.

ABSTRACT

Bentonite buffers and backfills, made using bentonite and bentonite mixtures, are applied

- to retard the access flow of groundwater to the vicinity of the copper canister. Thus, the corrosion of the canister starts after a greater delay.
- to mechanically isolate the canister from the environment.
- to slow down advection and diffusion between the capsule and its environment.

One of the assumed environmental risks is the possibility of frost penetration down to the level of the waste deposit after climate cooling. Thus, a study concerning the consequences of the possible freezing and thawing of sealing materials such as

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[10] By Seppo Saarelainen, VTT.

bentonite and bentonite mixtures was seen as necessary. The research results reported in the literature and concerning structural changes and changes in the hydraulic conductivity of frost-susceptible, fine-grained soils after freezing and thawing were referred to in this literature study. The frost susceptibility and hydraulic properties of bentonite products, bentonite mixtures, and bentonite clay in an unfrozen state and after freeze-thaw were also studied. The laboratory and field testing device and methods applied in the reported investigations were described. Finally, a proposal for a further testing programme was presented for the evaluation of the hydraulic stability of the planned protection materials in reference to freezing and thawing. According to the reports referred to, it is obvious that the risk of shrinkage in the clay material is characteristic for frost-susceptible liner materials, which leads to greater hydraulic conductivity after thaw. For this reason, the freezing properties of the planned protection materials should be investigated. Besides the hydraulic conductivity, the unfrozen moisture content in freezing temperatures and compression and frost-heaving characteristics of the material should also be determined in the laboratory. It was also concluded that if the buffer material were to shrink as a result of freeze-thaw, then the swelling pressure would be reduced.

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2.2.5 Coupled behaviour of bentonite buffer[11]

INTRODUCTION

Present plans for geological spent fuel disposal involve the use of an engineered barrier made of compacted bentonite clay. In addition to giving mechanical support, the purpose of the barrier is to insulate the waste canisters from flowing groundwater and, consequently, to reduce the release and transport of material from the repository. The heat released by the waste, flow of liquid and gas in the porous network, construction gaps and fractures, phase changes, and deformation of solid material constitute a very complicated thermo-hydro-mechanical (THM) system. The major problem is to understand the evolution of the behaviour of the system over long periods of time. Because of the large size of the repository and its surroundings and, especially, because

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[11] By Petri Jussila, TKK.

of the long time periods involved, the problem has to be investigated by means of modelling and numerical simulation.

Since 1991 the relevant coupled processes have been investigated in the series of DECOVALEX (2005) research projects (acronym for International co-operative project for the DEvelopment of COupled models and their VALidation against EXperiments in nuclear waste isolation). Finland has been involved in the series since the beginning with the STUK (Radiation and Nuclear Safety Authority) as the co-ordinator of the Finnish contributions.

In Task 1 of the previous Decovalex III project (1999–2003), coupled analysis of the bentonite and rock of the Febex "in situ" experiment was performed by ten modelling teams from Europe, North America, and Japan (Alonso et al. 2005). In Task A of the current Decovalex-THMC project (2004–2007) five international teams are investigating the influence of near field-coupled THM phenomena on Performance Assessment.

The ongoing FEBEX (Full-scale Engineered Barriers Experiment in Crystalline Host Rock) project has the objective of demonstrating the feasibility of actually manufacturing and assembling an engineered barrier system based on the Spanish disposal concept for a crystalline host rock and developing methodologies and models for the assessment of the thermo-hydro-mechanical and thermo-hydro-geochemical behaviour of the engineered barrier system. The project consists of an "in situ" full-scale test at the Grimsel Test Site underground laboratory in Switzerland, using a cylindrical heater to generate the heat radiated from a waste container, engineered bentonite blocks, and measuring equipment. In addition, a ¼-scale "mock-up" test and a series of complementary laboratory tests are being run in Madrid; modelling work is also taking place (Enresa 2000; Grimsel 2005).

The objective of Task A of Decovalex-THMC is to assess the implications of coupled THM processes in the near field of a typical repository on its long-term performance. The aim is to analyse the coupled behaviour of a hypothetical repository based on a Canadian concept in which high level-waste containers surrounded by a bentonite buffer are emplaced in parallel horizontal tunnels at a depth of 657 m in Canadian granitic bedrock. The first subtasks involve a preliminary coupled analysis of the whole repository and the development of models for bentonite and for rock damage. The preliminary analysis has been divided into two periods. The first part is the post-excavational simulation of open horizontal deposition tunnels for 30 years. The second part is the post-emplacement simulation of a repository filled with heat-producing waste containers surrounded by the bentonite buffer during a period up to 1000 years.

The modelling work presented here relies on the general model for a porous medium derived by the author at Helsinki University of Technology (TKK). Before the current KYT project the model had been applied to the simplified TH analysis of the Febex in situ experiment in Task 1 of Decovalex III (Alonso et al. 2005, Jussila 2005). During the participation in KYT in 2004–2005, the model was extended to incorporate the essentials of the fully coupled THM behaviour of a swelling elastic medium in the limit of small deformations (Jussila & Ruokolainen 2005). The model was calibrated to several laboratory experiments for Febex bentonite and applied to the simulation of the Febex mock-up and in situ experiments. In addition, from the beginning of 2004 the model was used in the participation in Task A of the Decovalex-THMC project. Detailed analysis will be presented in the Doctoral Thesis of Jussila (2006b).

The numerical calculations are performed with ELMER (CSC 2005), which is a program for solving coupled partial differential equations generated by multiphysics problems in continuum mechanics. A particular form of the program was created in cooperation with the experts of CSC to solve the current problems.

THE MODEL

A fundamental part of the work was the derivation of the THM model for an unsaturated swelling porous medium. A rigorous derivation of the general theory is presented in (Jussila 2005; Jussila & Ruokolainen 2005).

The modelling approach is based on thermomechanics and mixture theory. The model describes the behaviour of the mixture via proper choices of free energy and dissipation function. The free energy of the system is chosen to take into account the individual non-dissipative behaviours of the constituents and their mutual interactions, in particular, adsorption and swelling. The resulting thermodynamically consistent macroscopic model can be fitted to suction and swelling experiments and applied to the THM simulation of porous media by the finite element method.

The conservation equations to be solved are those for solid mass, air mass, water mass, linear momentum, and energy. The respective primary variables are porosity, gaseous pressure, degree of liquid saturation, solid phase displacement, and temperature. The constitutive relations needed are derived to be particular forms of the Clausius-Clapeyron equation, stress-strain relation, gaseous phase state equations, Darcy's law for liquid and gas, Fick's law for vapour and air, Fourier's law for heat conduction, and the internal energy differences of the mixture components.

THE FEBEX ANALYSES

The basic use of the current model has been its calibration against small scale laboratory experiments and the simulation of larger-scale Febex mock-up and in situ experiments. The simulation of the laboratory experiments and the basic choice for the model parameters are presented by Jussila and Ruokolainen (2005).

Small-scale HM and THM laboratory experiments

A large number of suction-controlled oedometer tests has been performed for the Almerian (Febex) bentonite; see e.g. Enresa (2000) or Pintado (2000). In the tests the volume change of a sample is measured, while the suction and vertical load are controlled. Four of the tests performed at the Technical University of Catalonia (UPC) were chosen for the calibration of the HM part of the current model. The best fit was achieved for those experiments involving the smallest deformations. The trajectories have a hypothetical counterpart in a buffer surrounding a waste canister in a geological repository. For example, Experiment No 3 (Figure 30) represents a point near rock wetting under an increasing load. For fully coupled THM calibration one of the experiments introduced by Villar et al. (1996) was chosen.

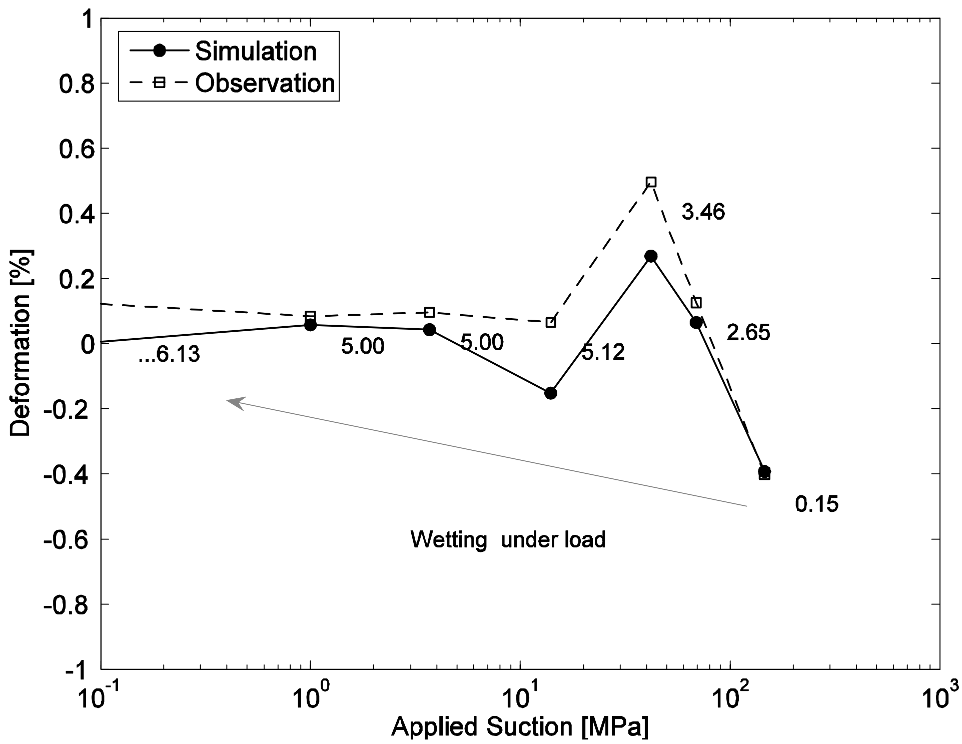
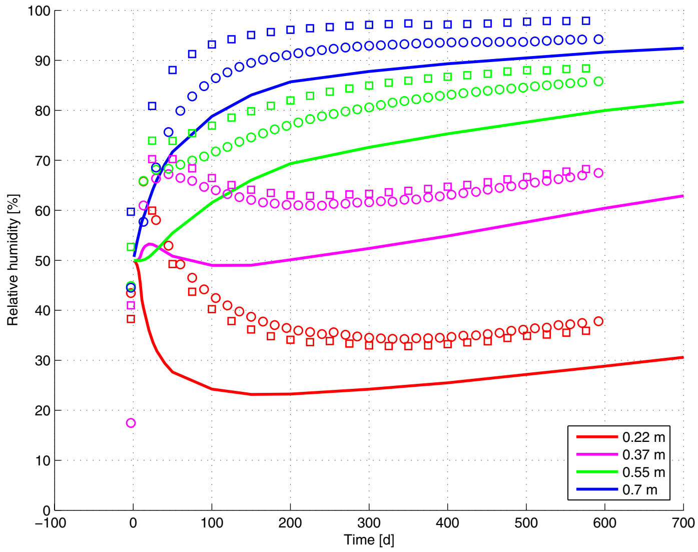
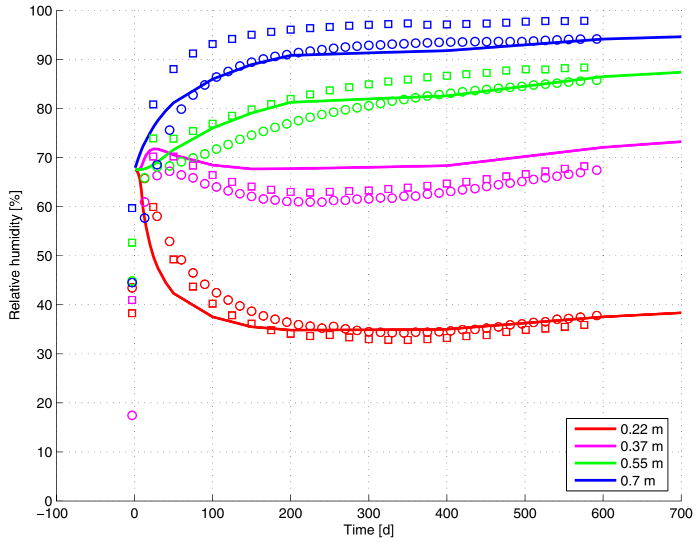


Figure 30. Hydromechanical laboratory experiment No 3 performed by UPC (Pintado 2000) and simulated by Jussila and Ruokolainen (2005). In the experiment the swelling of the sample was restricted by increasing the external load while the water content was increased by decreasing the suction.

The Febex mock-up and in situ experiments

Buffers of the mock-up and in situ experiments were simulated in 2D axially symmetric domains. A major difference between the observed and the simulated results for the mock-up (Figure 31) and in-situ experiments (Figure 33) occurs in the early stages of the moisture evolution.





Figures 31 and 32. Febex mock-up test. Simulated (curves) and measured (symbols) evolutions of relative humidity at four material points in the buffer for the basic case of the actual initial water content (above) and by assuming the initial water content to be considerably higher (below). The observed rapid increase in the humidity at the beginning of the experiment does not occur in the simulation. By assuming the initial water content to be significantly higher, the simulated result is very close to the observation after the transient phenomenon. The observed phenomenon thereby contributes to the construction gaps characteristic of the Febex buffer.

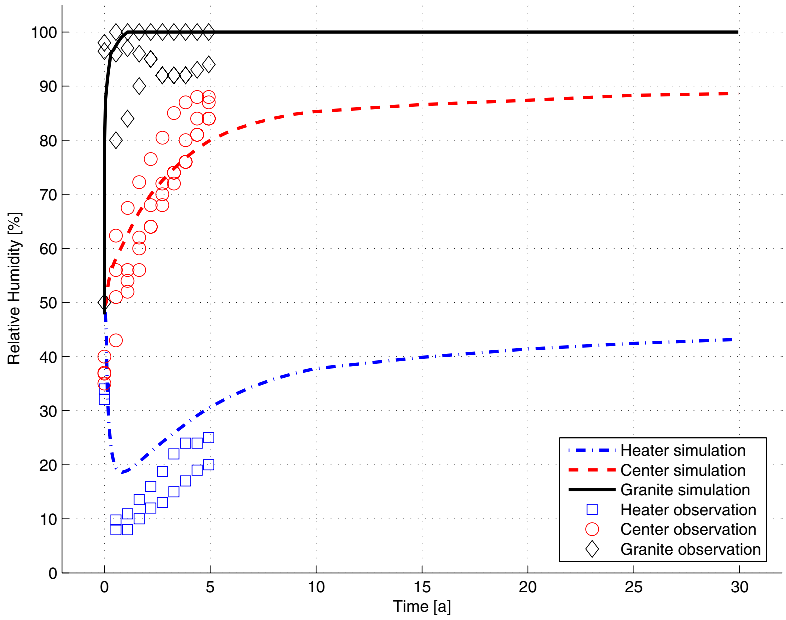


Figure 33. Febex in situ test. Simulated and measured evolution of relative humidity at three material points in the buffer. The model predicts a steady hydration state with a fairly dry inner buffer. In other words, in this particular experiment of prevailing high temperature difference the buffer will never get fully saturated.

The measured relative humidities increase much faster during the first ten days and stay at elevated levels. In Figure 32 the mock-up simulation is performed with the same parameter values but assuming the initial moisture content to be significantly higher than the actual average. It can be seen that the measured moisture behaviour after the transient phenomenon is very close to this latter simulation. In the in situ measurements the same transient phenomenon is not as regular as in the mock-up data. The transient phenomenon is not observed in the current simulations, in various simulations performed by other teams with different codes, or in the measured small-scale laboratory data for Febex or MX-80 bentonite.

For the in situ experiment there are experimental data for 5 years. The simulation with the basic set of parameter values predicts that the inner portions of the in situ buffer will remain fairly dry and a steady state will be achieved in about 30 years (Figure 33). The prediction is that for this particular exercise of prevailing high temperature difference the buffer will never become fully saturated.

DECOVALEX-THMC TASK A

The fully coupled THM development of a hypothetical repository consisting of adjacent horizontal tunnels at a depth of 657 m was simulated in two parts. The first part is the post-excavational simulation of open horizontal deposition tunnels for 30 years. The second part is the post-emplacement simulation of the repository filled with heat-producing waste containers surrounded by the bentonite buffer during a period up to 1000 years. A more detailed analysis will be provided by Jussila (2006a, b) and by Chijimatsu et al. (2006).

Some intermediate calculations were performed with 2D and 3D geometries reaching from the ground surface y = 657 m to the symmetrical depth of y = –657 m. All the final results presented here, however, are calculated with a cut geometry reaching 300 m above and below the repository level in order to spare calculation time.

The rock mass is modelled as an elastic continuum. To verify the mechanical part of the numerical Elmer program a case comparable to the available analytical solution (Jussila 1997) for 2D steady state plane stress around a circular cavity in an infinite continuum was formed. Comparison showed that the numerical radial displacements deviate only slightly from the analytical solutions because of the different boundary conditions (Jussila 2006a).

Task A1 – The post-excavational THM analysis of rock at 0–30 years

The coupled THM system was applied to the modelling of rock by neglecting the swelling interaction. The initial condition for the rock was chosen in such a way that an undisturbed system does not theoretically evolve from the initial state. The chosen initial conditions are constant porosity of the rock at the depth of the repository, hydrostatic linearly depth-dependent pore pressure, full saturation, zero displacement, linearly depth-dependent in situ stresses, and linearly depth-dependent temperature.

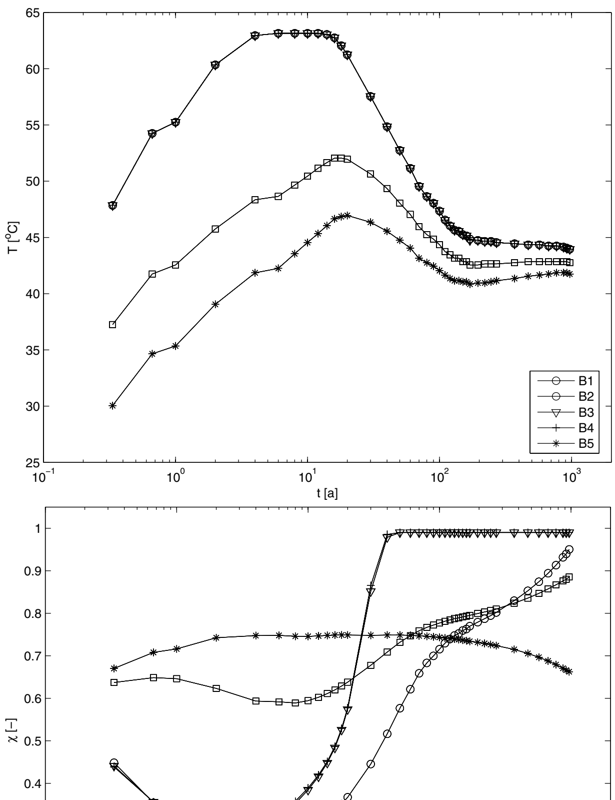
The rock material is assumed to be elastic and the stresses obtain their final values at the first time step. According to the simulations the vertical displacement, however, does evolve during the whole post-excavational period of 30 years because of the evolving pore pressure. Because of the excavation, the pore pressure around the tunnel is reduced and the whole rock mass moves downwards. The pressure field and the vertical displacement are still evolving and a steady state has not been reached at 30 years. The pore pressure drop and the ventilation of the tunnel reduce the saturation degree up to dozens of metres from the tunnel. The resulting initial condition for the post-emplacement THM analysis would slow down the commencement of the bentonite resaturation.

Task A1 – The post-emplacement THM analysis of the container, bentonite, and rock at 30–1000 years

The post-emplacement analysis involved the container, buffer, and rock, time-dependent heat production by the waste, and a time range of 30–1000 years.

Originally, it was planned to use the fully coupled post-excavational results as an initial condition for the post-emplacement problem. Unfortunately, this was not numerically successful. Furthermore, with the swelling model of the buffer included, convergence was achieved only for a couple of relatively small time steps. Consequently, to achieve numerical post-emplacement results up to 1000 years all three materials were assumed to appear at the same time at 30 years and the swelling of the buffer had to be discarded.

The time evolution of temperature and saturation degree at selected points of the buffer are illustrated in Figures 34 and 35, respectively.



Figures 34 and 35. Decovalex-THMC, Task A1. Post-emplacement analysis of rock and bentonite. Simulated evolution of temperature (above) and saturation degree (below) at selected material points in the buffer after emplacement. The maximum temperature occurs at 8 years after emplacement and the temperature difference decreases with time as the temperature of the surrounding rock mass is increased. The buffer along the vertical axis is fully wetted in 50 years, while the buffer along the horizontal axis remains dry. This unexpected latter result is due to the combined effect of various invalid assumptions in the modelling approach.

The temperature results seem to be realistic. The maximum temperature of 63°C is obtained at 8 years after emplacement and the temperature gradients tend to decrease along with time as the temperature of the surrounding rock mass increases.

The results for saturation degree are partially unusual and unexpected. Points along the vertical axis seem to exhibit traditional behaviour. The container surface (B2 and B3) initially dries, followed by full resaturation in 50 years. This is slow but compatible with the previous experience with the current model. On the other hand, the results along the horizontal axis are unusual. The full resaturation of the container surface (point B1) takes more than 1000 years, while the point at the rock surface (B4) remains unsaturated and even dries further over time. While the possibility of numerical errors cannot be discarded, there are numerous possible explanations for this behaviour based on the modelling approach and the computational set-up:

i) The temperature gradient prevents the resaturation, which by itself is not a sufficient explanation, though.

ii) The initial condition used, assuming all the materials appear at the same time, is not realistic. The in situ stresses deform the tunnel and the buffer in such a way that the volume of the material along the x-axis increases. This results in the opening of the pores and apparent drying of the rock and the buffer which seems to prevail.

iii) A major driving force in the model for the liquid transport is the suction, which depends on the porosity. The initial deformation of the tunnel surroundings leads to increased porosity along the x-axis and decreased porosity along the y-axis. As a consequence, the suction ability of the material along the x-axis is relatively decreased, which prevents the resaturation of the same area.

iv) The choice of the reference state for the adsorption function results in inaccurate resaturation of areas of increased porosity. Consequently, the rock material along the x-axis stays unsaturated.

v) The saturation degree as a state variable was assumed to be a continuous variable at the rock-buffer interface, which, by definition, is an error. Because of this the buffer along the x-axis also stays unsaturated. If a full saturation prevailed at this interface this would not be a problem. For the current unsaturated situation the correct boundary condition should be a continuous suction.

Task A2 – Calibration of MX-80 bentonite models

In the first part of Task A2 the bentonite models are calibrated by means of small-scale laboratory tests performed by SKB (Börgesson & Hernelind 1999) and a larger-scale CEA mock-up experiment. Detailed results of the teams' work will be presented by Börgesson et al. (2006).

The CEA mock-up experiment is well documented and proved to be very valuable and interesting. The applied temperature gradient is too intensive, however. Because of this the sample has encountered very intensive water redistribution and, consequently, very intensive local swelling and shrinking. The current model is not capable of reproducing such large deformations. Instead, for the time being, the whole experiment has been simulated by neglecting the swelling behaviour in the model. Figure 36 illustrates the simulated and observed evolution of the relative humidity of three material points inside the bentonite sample. In Phase 1 of the experiment an increasing temperature difference is applied across the sample. In Phase 2 the temperature difference is kept constant, while the sample is wetted from its other end. As can be noted, the transient phenomenon encountered in the early stages of the Febex experiments does not occur in this experiment.

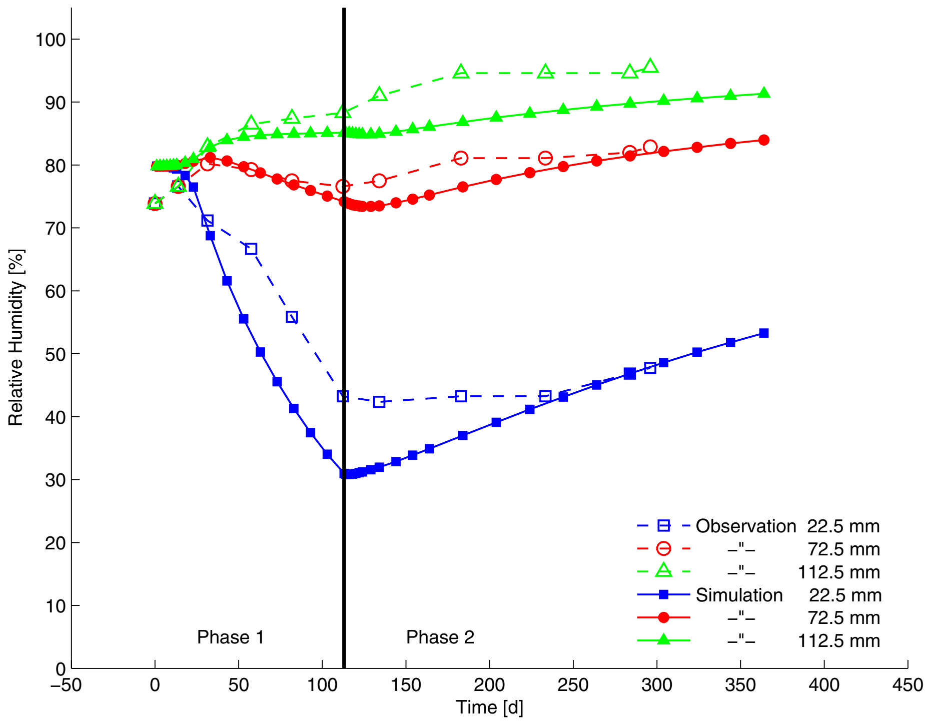


Figure 36. Decovalex-THMC, Task A2. Simulated and observed relative humidity evolutions of three material points of Cell 2 of the CEA mock-up test for MX-80 bentonite. In Phase 1 the temperature difference in the sample is increased at constant water content, which makes the water move towards the cold end. In Phase 2 the high temperature difference achieved is kept constant, while the sample is wetted from one end. Note that the rapid increase observed at the beginning of the Febex mock-up test (Figure 34) is not observed in the CEA mock-up test.

CONCLUSIONS

Coupled continuum mechanical modelling of the repository and the surrounding bedrock gives a fundamental insight into the geological behaviour of the spent fuel repository. The simulation performed in Task A of the Decovalex-THMC project involving adjacent deposition tunnels, heat-producing waste containers, bentonite buffer, and hundreds of metres of bedrock has been a major effort. The traditional assessment of the long-term safety of disposal and the release of radionuclides from the repository is based on static isothermal and fully saturated conditions in the repository and the bedrock. According to the results of the current project, this kind of static condition will not be encountered in the considered time span of the first 1000 years of disposal. The excavation of the repository causes the pore pressure of the bedrock to collapse and the groundwater to flow towards the repository, yielding a global settlement of the surrounding bedrock. When the heat release of the waste is taken into account, the moisture field in the repository and its surroundings is affected in a complicated and coupled way by the model parameters. Especially, the values of the relative permeabilities of liquid and gas in the buffer and in the rock have a crucial effect on the evolution of the moisture field and on the buffer resaturation time.

In Task A1 the simulated unusual moisture field behaviour in certain parts of the domain is probably the combined result of various invalid assumptions in the modelling approach and in the computational set-up, which will have to be corrected. A major problem encountered during the phase Task A2 was the unsuitability of the given experimental data for the calibration of the MX-80 bentonite models. The main problem is that clay is inherently plastic and exhibits large irreversible deformations. The current model is capable of describing small reversible deformations only. Furthermore, the behaviour of swelling material depends on its initial conditions, i.e., on the initial porosity and moisture content. The experimental set-ups allow large local deformations and the initial conditions of the various experiments to deviate from each other and from the given simulation target. The suggestion is that every experiment to be used for the calibration of swelling material behaviour should have the same initial conditions and should provide as much information as possible on the behaviour of the system in the close vicinity of these initial conditions.

Experimental data on the evolution of the Febex mock-up (and in situ) moisture content exhibit a transient phenomenon not encountered in smaller-scale experiments. Nor is it reproduced in the current simulations or in the corresponding simulations found in the literature. This phenomenon is the fast wetting of the internal measuring points at the beginning of the experiment. The explanation is assumed to stem from the brick wall-like structure of the Febex buffer, which consists of thousands of compacted bentonite blocks with construction gaps between them. Temperature gradient-induced vapour flow occurs preferably in these gaps. After the condensation of the vapour and the consequential sealing of these gaps the moisture contents of these areas stay at a higher level than inside the blocks. After the transient phenomenon the observed evolution is qualitatively the same as in the other experiments and in the simulations. The simulation of a geometry involving the construction gaps would give more confidence regarding this hypothesis.

The evolution of the thermo-hydro-mechanical field is affected in a complicated way by the interaction of excavation, pore pressure evolution, local and global rock deformation, the intrinsic permeabilities of rock and buffer, the relative permeabilities of liquid and gas, and the heat production of the waste. As the simulation results depend significantly on the couplings of the modelling assumptions, the long-term behaviour of an actual repository with its own local properties is very difficult to predict. In addition, the engineered structure of the bentonite buffer seems to have a significant effect on the evolution of the moisture field in the early stages of the disposal. However, this effect is not significant for the long-term behaviour.

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2.3 Bedrock and groundwater

The studies on bedrock and groundwater are grouped into eight basic components. The strength of crystalline rock in long-term loading was studied mostly as experimental work in order to understand brittle rock failure. The time-dependent fracturing of rock mass is a modelling exercise within the international DECOVALEX THMC project supported by related experimental work. Bedrock fracturing and hydrogeology aims at obtaining an improved link between bedrock structure and groundwater flow modelling. Geochemical signals of ice ages in bedrock aim at a methodology for the better interpretation and understanding of paleohydrogeological signs in crystalline bedrock. Gas studies in the Olkiluoto area deal with gas or groundwater anomalies found recently in sediments in the Olkiluoto sea area. Permafrost studies at the Lupin Mine in Canada were conducted in international collaboration. Postglacial bedrock movements were studied in order to summarise the current knowledge of the postglacial deformation of the mosaic-like bedrock in Finland. The most important conclusions drawn from the natural analogue studies at the Palmottu U deposit in SW Finland were summarised with a view to safety case considerations.

2.3.1 Strength of crystalline rock in long-term loading [12]

INTRODUCTION

In the design of an underground repository in hard rock for the disposal of spent nuclear fuel two different time spans should be considered in rock mechanical analysis. The first time period covers the construction, operation, and closure of the repository and its possible reopening and the recovery of disposal canisters. For this period, the stability of the excavation is of paramount importance. During the construction of the repository the safety of personnel is the key issue. During the later phases, the operation, closure, and possible reopening of the repository, any rock failure is a potential source of major problems with remotely operated or even autonomously operating equipment.

The second time period extends from the final closure of the repository to 100,000 years in the future, when the activity of the waste will have decreased to a level comparable with uranium ores mined today. During this period at least a partial glaciation cycle is expected, which will have a major effect on the rock stress at the level of the repository. On the other hand, the requirements for stability are not as strict as for previous phases, because there is no evident need to enter the repository after a few hundred years after closure. Thus some rock breakage can be tolerated under the loading of glaciation, as long as no new continuous and extensive fracture zones are formed.

The use of rock reinforcement in the repository is restricted because the existence of different materials inside or in the vicinity of the repository complicates the chemistry of the deposition process. Both the number of different materials and the amount of any

[12] By Juha Antikainen, TKK.

“foreign” material should be kept to a minimum. Therefore, just adding steel and concrete would not necessarily improve the performance of the repository, but possibly have the opposite effect. Thus the need for rational rock mechanical design and a better understanding of long-term loading conditions and also the long-term strength of the rock is highly significant.

ROCK MECHANICAL CONDITIONS IN A REPOSITORY

In this work the rock mechanical conditions around a hypothetical hard rock repository in a sub-Arctic geographical region were reviewed, using the literature as the main source. The rock mechanical conditions in and around a repository vary with time. The in situ state of the rock is disturbed by the excavation of the repository. The groundwater pressure drops, the temperature of the rock changes as a result of the ventilation, and, most importantly, the rock stress field is strongly affected in the vicinity of the repository. Whether this secondary stress field is capable of producing the failure of intact rock depends on the in situ stress field, the geometry of the excavations, and the properties of the intact rock and rock mass.

During the operation of the repository the changes in conditions are small and slow, consisting mainly of gradual changes in rock temperature and groundwater pressure. When the repository is partially and finally completely closed the conditions will change. The spent fuel produces heat, the groundwater pressure increases, and the filling materials produce internal pressure inside the deposition holes and, to some extent, also in other filled excavations. Internal pressure in the excavations increases the strength of the adjacent rock by increasing the minimum principal stress σ_3. The swelling pressure of filling materials varies. For bentonite filling the swelling pressure can be in the same order of magnitude as the in situ rock stress. This effect is rather local, however, because the volume of excavations is very small compared to the volume of the rock mass in the area of the repository. The major source of short-term (up to a few hundred years) stress changes in the rock mass is the thermal expansion of rock resulting from the heat produced by the spent nuclear fuel (Öhman et al. 2004).

The actual rock mechanical conditions within a specific site can be found out by means of geotechnical investigations and the changes can be forecast tens or even hundreds of years from the present with adequate accuracy. This is not the case when a 100,000-year time period is considered. The climate changes preceding and during the next glaciation period are quite impossible to predict in detail. Some general features are generally accepted, however.

Glaciation is likely to begin with a long periglacial period thousands or tens of thousands of years long, with a cold and dry climate. Permafrost up to 0.5–1 km deep will develop.

As the glaciation advances and possibly later retreats, the thickness of the permafrost will decrease and the groundwater pressure and hydraulic gradients increase. Nevertheless, the hydraulic jacking or rafting of megablocks is neither predicted in modelling (Chan et al. 2005) nor has it been observed within crystalline rocks (Talbot 1999).

The glacial ice sheet can be up to 3 km thick. The weight of the ice increases the vertical stress and, because of the elasticity of the rock material, the horizontal stress also increases in a proportion defined by Poisson's ratio:

$$\Delta\sigma_h = \frac{\nu}{1-\nu}\Delta\sigma_v \tag{5}$$

where: $\Delta\sigma_h$ = change of horizontal stress, $\Delta\sigma_v$ = change of vertical stress, and ν = Poisson's ratio.

Assuming an ice thickness of 3 km and a Poisson's ratio of 0.23, the increase in horizontal stress is about 8 MPa (La Pointe & Hermanson 2002). This is only the instant effect on the weight of the ice. Because of the bending of the Earth's outer crust as a result of the weight of the ice sheet, the increase in the horizontal stress can be up to twice the increase in the vertical stress. The stress change resulting from crust bending is slow; a 10,000–20,000-year period is required for its full effect (Talbot 1999).

The stress changes induce strains in intact rock and shear displacements along existing fractures. In good-quality rock mass the shear displacements induced are a few centimetres over the entire glaciation cycle (Hutri & Antikainen 2002, Chan et al. 2005). Additionally, the shear displacements associated with earthquakes typically take place along existing fractures or larger-scale discontinuities (La Pointe & Hermanson 2002). The larger the geometrical extent (area) of a fracture, the larger the shear displacement that will take place under a given stress change. Thus it is obvious that the deposition holes and, preferably, also the deposition tunnels must not intersect with any larger discontinuity.

ROCK STRENGTH CRITERIA

As described above, the fractures, if present, have a major effect on rock mass properties. In sparsely jointed rock under high stress, the strength of intact rock can also be the weakest link. The typical behaviour of brittle rock under uniaxial compressive loading in laboratory tests is presented in Figure 37. Testing is usually carried out with a relatively high loading rate (0.5–1.0 MPa/s), as defined by ISRM (ISRM 1981). With this test all the stress levels σ_{ci}, σ_{cd}, and σ_f can be obtained. When the crack initiation stress σ_{ci} is exceeded, stable crack growth starts. Stable crack growth refers to the situation where an increase in loading is required for further cracking. When the crack damage stress σ_{cd} is exceeded, the cracks will grow under constant load, and, the higher the stress is, the sooner the failure of the rock will take place. Thus the peak strength of rock σ_f is, in fact, strongly dependent on the time and loading rate. For a very long period of time $\sigma_f = \sigma_{cd}$, indicating that the crack damage stress σ_{cd} is the long-term strength of the rock (Martin & Chandler 1994). Under certain geometry and loading combinations, even lower long-term strength has been observed (Diederichs 2003).

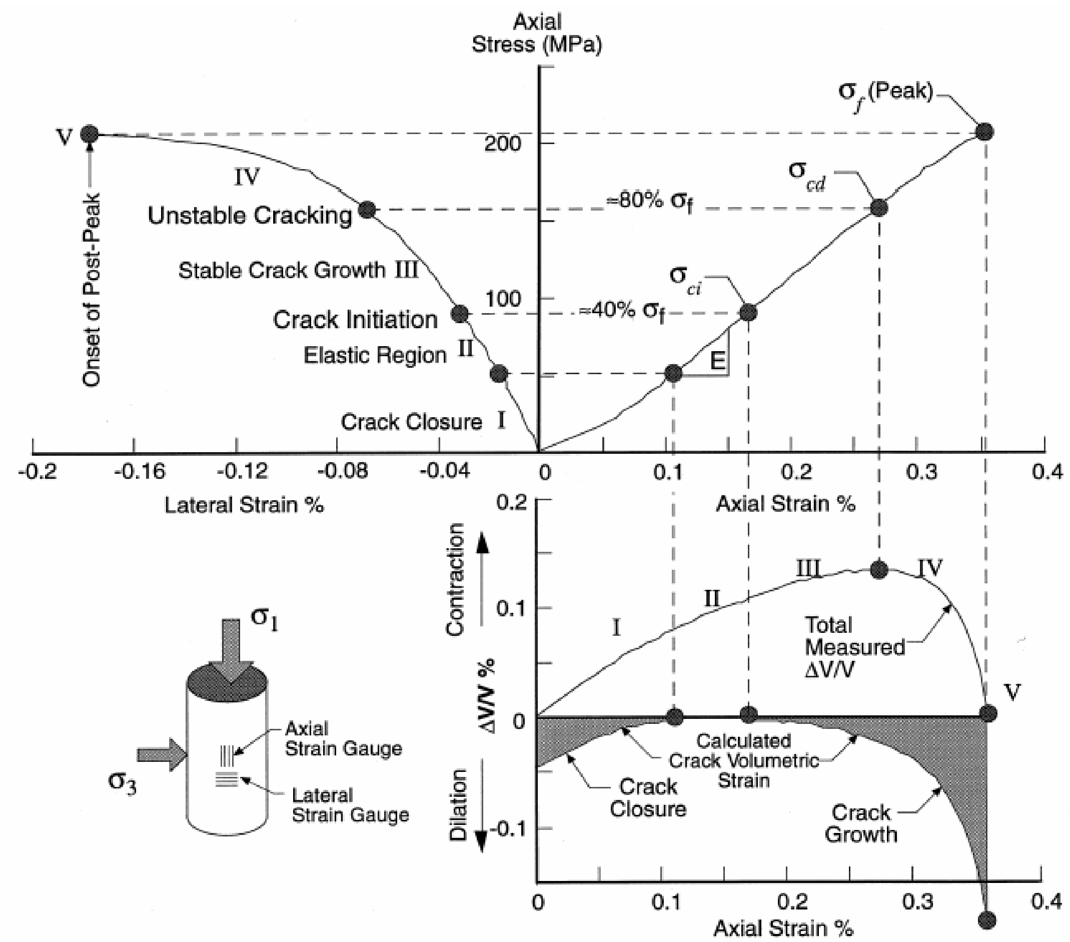

Figure 37. Stress-strain diagram obtained from a single uniaxial compressive test for Lac du Bonnet granite showing the definition of crack initiation (σ_{ci}), crack damage (σ_{cd}), and peak strength (σ_f). The axial and lateral strains are measured and the volumetric strain and crack volumetric strain are calculated using the measured strains (Martin & Chandler 1994).

The Hoek-Brown and the Mohr-Coulomb strength criteria are the most widely used criteria in rock mechanics. The Mohr-Coulomb criterion's advantage is its mathematical simplicity:

$$\tau = c + \sigma_n \tan \phi \qquad (6)$$

where τ = shear strength, c = cohesion, σ_n = normal stress, and ϕ = friction angle. The Hoek-Brown criterion generally produces better strength estimates with large variations in minimum principal stress σ_3, and the properties of intact rock and rock mass can be better taken into account:

$$\sigma_1' = \sigma_3' + \sigma_{ci}\left(m_b \frac{\sigma_3'}{\sigma_{ci}} + s\right)^a \tag{7}$$

where σ_1' = maximum effective principal stress, σ_3' = minimum effective principal stress, σ_{ci} = uniaxial compressive strength of intact rock material, m_b, s, a = Hoek & Brown parameters for rock mass, m_i = parameter m value for intact rock.

These material models are not able to reproduce all the failure modes observed in brittle rock. Modelling the formation of thin slabs (spalling) in rock mass under high stress and low confiment requires either major manipulation of material model parameters or the use of completely different material models. The slabbing phenomenon is especially important in the underground disposal of spent nuclear fuel. The repositories will be located in sparsely jointed rock and probably at a depth of 300–700 m. Thus, besides local minor failures associated with the few joints present, the major mode of possible rock failure is likely to be spalling. Minor spalling is not an important problem in mining or tunnelling, because thin slabs can easily be retained with support. In temporary mining openings the slabs are just scaled down periodically. In repositories the formation of slabs should be avoided, both for safety during operation and so as to prevent the formation of a large Excavation-Damaged Zone (EDZ).

A CWFS (Cohesion Weakening Friction Strengthening) model is suggested for the modelling of slabbing phenomena (Hajiabdolmajid et al. 2002a, Hajiabdolmajid et al. 2002b). In the CWFS model the cohesion and friction are not constant, but are a function of strain:

$$\tau = c(\varepsilon) + \sigma_n(\varepsilon) \tan \phi \tag{8}$$

When intact rock is subjected to increasing loads, the cohesion of the intact rock is instantaneously mobilised, but the friction component is not mobilised before permanent damage has taken place in the form of the growth of microcracks. The required stress level is the crack initiation stress σ_{ci} (Figure 38). In the post-peak area the residual value of cohesion c_r is realised at the strain value ε_c^p and the maximum friction at the strain value ε_f^p. The strain ε_c^p needed for minimum cohesion is basically a material property, but the strain ε_f^p needed for the full development of friction depends on the geometry and loading rate. Thus the same rock material can show either brittle or ductile behaviour (Hajiabdolmajid et al. 2002a).

The brittleness can be defined with the brittleness index I_B if the strains ε_c^p and ε_f^p can be found or estimated:

$$I_B = \frac{\varepsilon_f^p - \varepsilon_c^p}{\varepsilon_c^p} \tag{9}$$

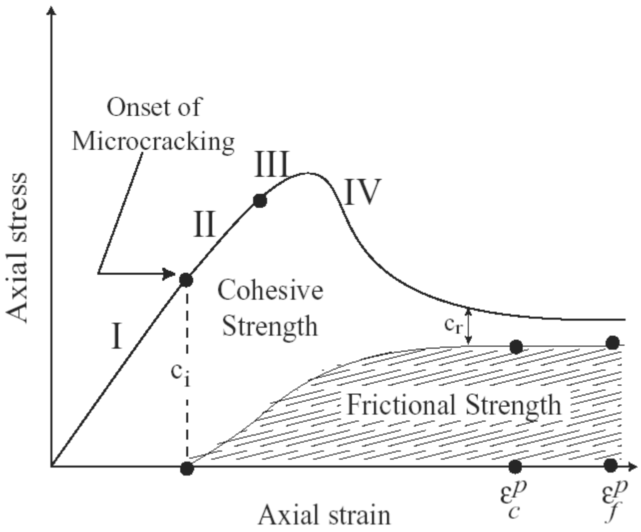

Figure 38. Mobilisation of the strength components at different levels of strain (Hajiabdolmajid et al. 2002b).

If the friction mobilises instantaneously, the brittleness index I_B will get a negative value -1. A positive I_B will manifest brittle behaviour. In this case there will be a local minimum in the stress-strain curve (Figure 39). Until now the I_B values have been acquired only with back analysis of actual spalling in excavations (Hajiabdolmajid & Kaiser 2003), because with the normal testing geometry the friction is mobilised instantaneously (Hajiabdolmajid et al. 2002b).

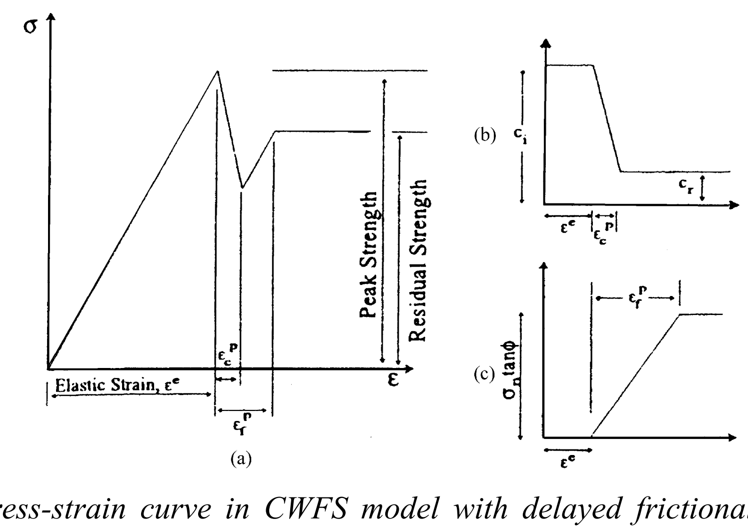

Figure 39. Stress-strain curve in CWFS model with delayed frictional strengthening (Hajiabdolmajid et al. 2002b).

LABORATORY TESTS

The goal of the laboratory tests was to find out if the brittle behaviour and/or the spalling phenomena can be reproduced in the laboratory. The rock type tested was grey granite, well known in the dimension stone industry as Kuru Grey. Kuru Grey is a fine-grained, isotropic, and homogenous rock type. The test material was from the Niemenkylä quarry. All 22 specimens tested originated from a single block (Figure 40).

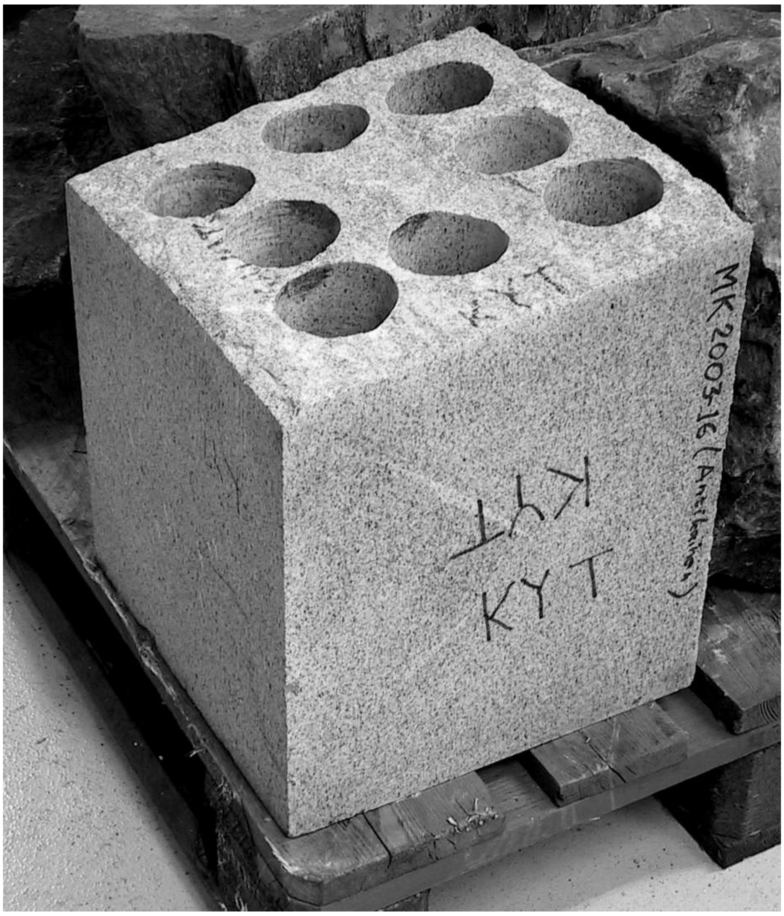

Figure 40. All specimens were drilled from a single block of Kuru Grey granite. The diameter of the holes is 69 mm and the diameter of the core 62 mm.

Although the properties of Kuru Grey granite are well known, a limited number of normal tests was performed just to confirm that the sample block represents typical Kuru Grey. These tests were carried out according to the ISRM Suggested Methods (ISRM 1981) and the results are presented in Tables 6–7. The results are well within previously published values (Salminen & Viitala 1985, Mesimäki & Jauhiainen 1997).

Table 6. Standard compressive test results.

Specimen N:o	Length L mm	Diameter D mm	L/D	Weight g	Density kg/m^3	Confining pressure MPa	Compressive strength MPa	E GPa	Poisson's ratio
1	162.23	62.09	2.61	1292.4	2631	0	274.5	67	0.26
2	162.22	62.11	2.61	1292.7	2630	0	276.3	67	0.26
3	162.30	62.13	2.61	1294.9	2632	0	277.7	67	0.25
4	162.31	62.11	2.61	1294.4	2632	10	440.4		
				average density	2631	kg/m^3			

Table 7. Indirect tensile test (Brazil test) results.

Specimen N:o	Length L mm	Diameter D mm	L/D	Weight g	Density kg/m^3	Tensile strength MPa
B-1	32.25	62.11	0.52	255.48	2614	11.3
B-2	31.75	62.12	0.51	252.15	2621	13.3
B-3	31.87	62.12	0.51	253.64	2626	9.5

The measurement of the brittleness index I_B is not possible with normal testing geometry, where the length/diameter (L/D) ratio is, according to the ISRM recommendation, 2.5–3.0. Thus an L/D ratio of 0.5 was adapted. The test control practice also had to be modified. The normal practice in the uniaxial testing of strong, brittle rock samples is to use the circumferential displacement as the control variable, which usually gives the best test stability at the failure (Brady & Brown 1993). The disadvantage of this practice is that the actual loading rate will slow down by as much as several orders of magnitude when the peak strength is approached. The reason for this is the dilation of the specimen, which increases the apparent volume of the specimen very rapidly near and after the peak strength. Thus the uniaxial testing of short specimens was performed under axial displacement control (Figure 41), while circumferential control was applied in triaxial testing (Figure 42). The strengths of short specimens are presented in Table 8.

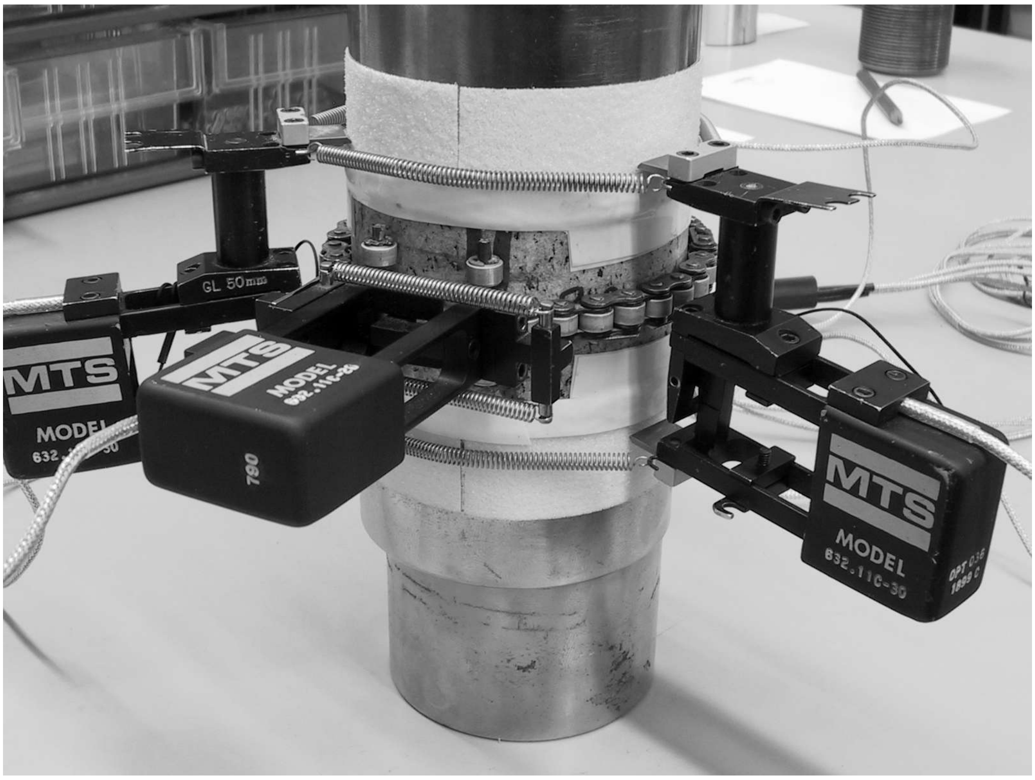

Figure 41. Test set up for the uniaxial testing of a short (L/D =0.5) specimen.

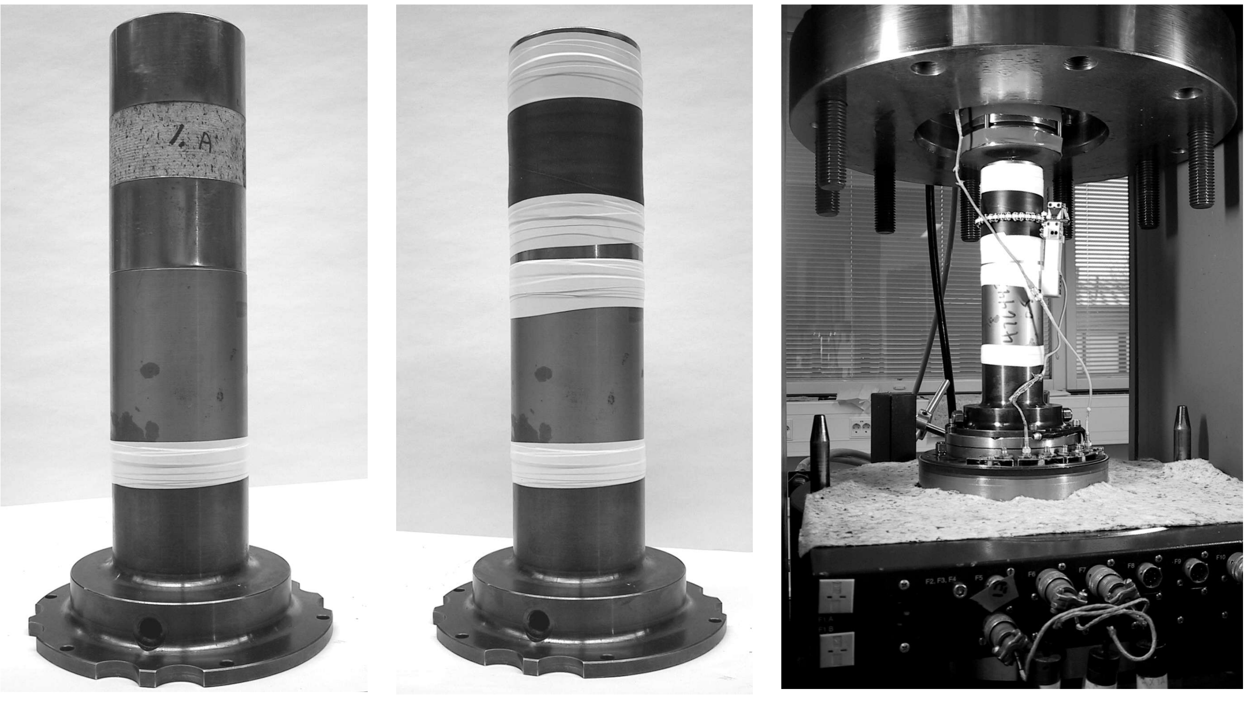

Figure 42. Set-up for triaxial test. Left: specimen and endcaps, centre: rubber sealing sleeve installed, right: instrumented specimen ready for testing.

Table 8. Test results for short (L/D = 0.5) specimens.

Specimen N:o	Length L mm	Diameter D mm	L/D	Weight g	Density kg/m^3	Confining pressure MPa	Peak strength MPa
5A	30.06	62.14	0.48	239.41	2626	0	386.8
9A	31.39	62.24	0.50	250.79	2625	0	388.5
9B	31.53	62.22	0.51	251.81	2627	0	354.9
2A	31.60	62.11	0.51	251.04	2622	0	359.4
6B	31.09	62.12	0.50	247.43	2626	0.5	410.3
1B	31.40	62.08	0.51	249.55	2625	1	393.8
2B	31.68	62.10	0.51	251.94	2626	2	469.8
3B	31.09	62.11	0.50	247.33	2626	3	489.8
6A	31.04	62.11	0.50	247.06	2627	5	519.0
7B	31.27	62.12	0.50	248.76	2625	5	493.0
4A	31.54	62.12	0.51	250.84	2625	7	567.3
7A	31.85	62.14	0.51	253.52	2625	15	680.1
3A	31.71	62.06	0.51	251.76	2624	10	614.0
8B	31.10	62.12	0.50	247.46	2626	20	732.1
8A	31.31	62.13	0.50	249.13	2625	20	735.0
				average density	2625	kg/m^3	

When all the strengths are plotted in the σ_1–σ_3 plane, both the effect of specimen L/D ratio and the confining pressure are obvious (Figure 43). The reason for the greater strength of short specimens is the existence of a triaxial stress state in the centre of the specimen resulting from the geometry and friction between the specimen and steel endcaps.

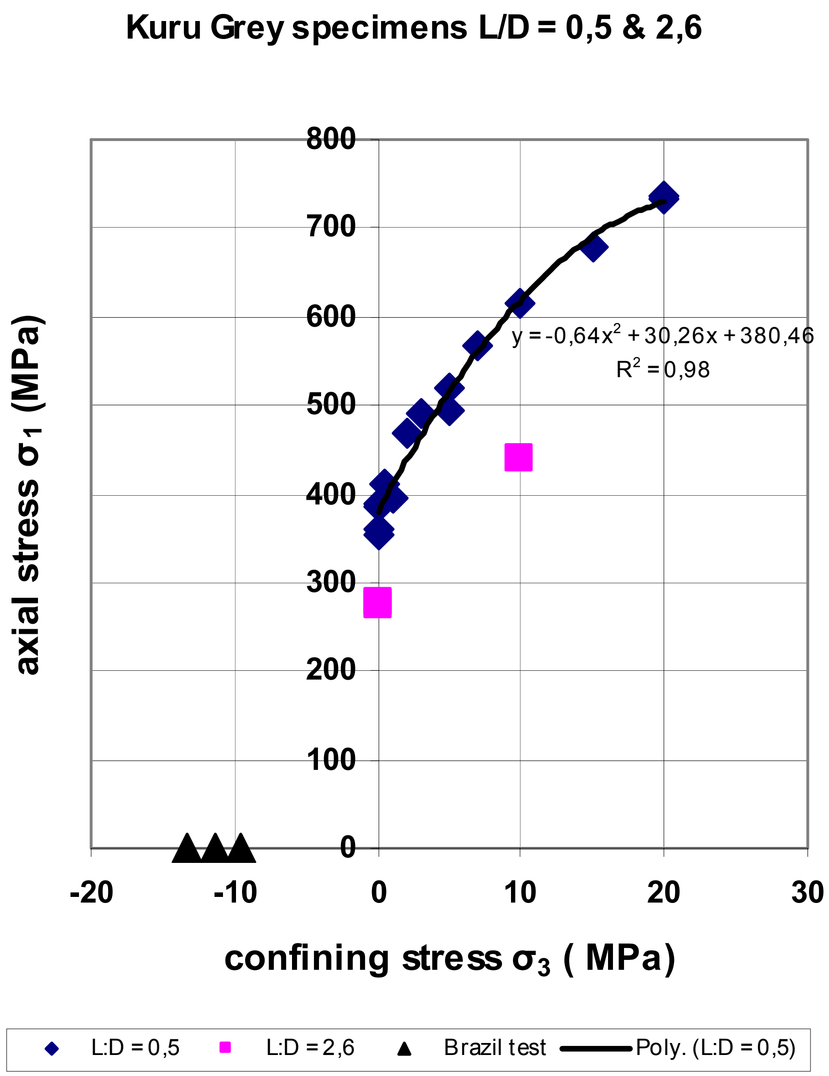

Figure 43. Peak strength values plotted in the σ_1-σ_3 space. Note the different scales of the axis.

If the stress state in a brittle specimen is completely uniform, the stress-strain curve would have the shape presented in Figure 39. In practice, and especially when testing short specimens, the conditions promote brittle failure only in a limited volume and near the specimen surface. In testing this become evident when a series of fractures develop gradually from the surface towards the centre of the specimen (Figure 44). The reason for the asymmetry is the change in local stiffness in a specimen after local fracturing, which leads to asymmetric axial loading.

The gradual, zonal failure of a specimen produces a stress-strain curve which has upward portions in the post-peak area (Figure 45). This temporary increase in strength is real, because the test was carried out with a steadily increasing axial strain controlled by extensometers attached to the steel endcaps (Figure 41).

Figure 44. Gradual failure of specimens 5A (left) and 9B (right).

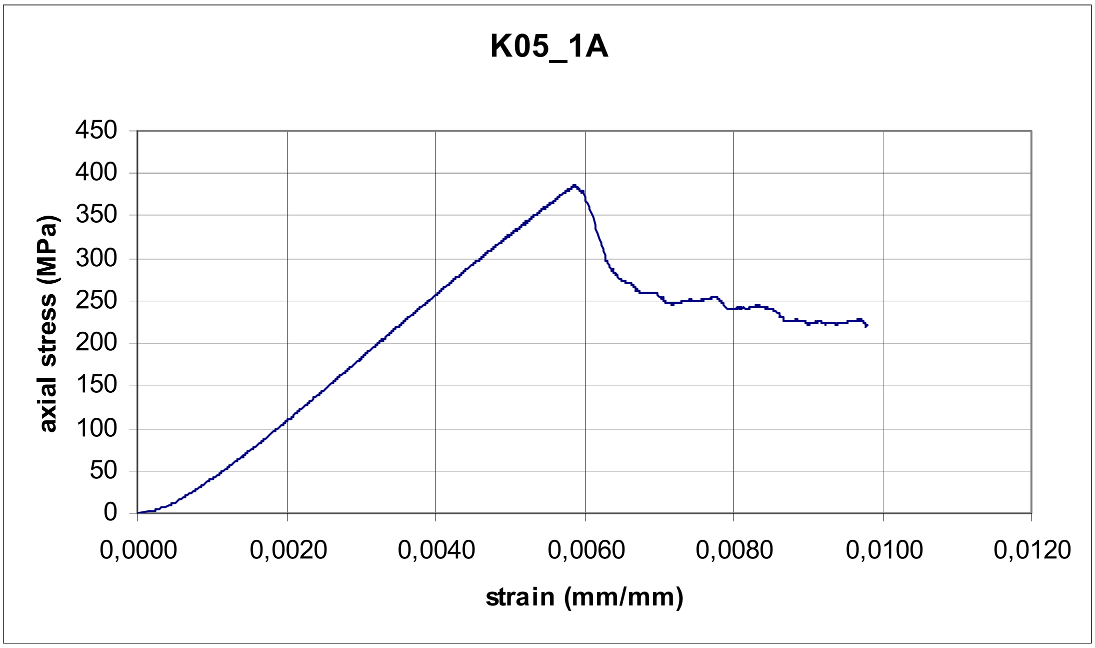

Figure 45. Stress-strain graph of uniaxial testing of short sample 5A.

Because the brittle failure takes place only in a small portion of the specimen at any given time, the test results are actually only the sum of overlapping events. With detailed inspection of the results, single events can be separated and the brittleness index I_B calculated (Figure 46). The brittleness index for Kuru Grey is about 2.5.

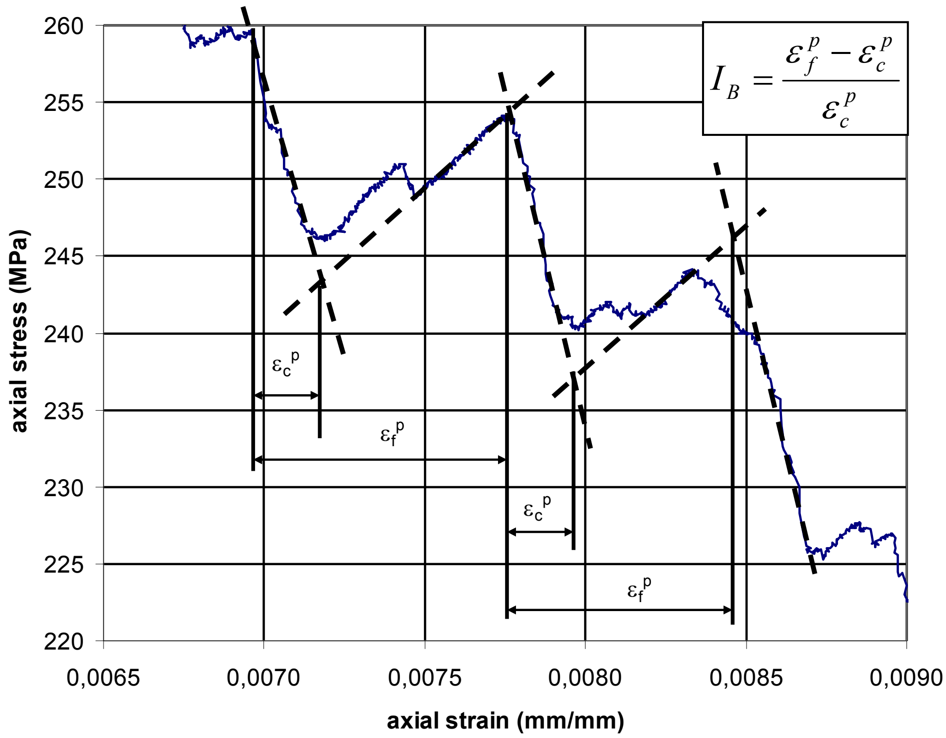

Figure 46. Partial magnification of Figure 45, calculation of brittleness index I_B.

CONCLUSIONS

The rock mechanical design of a hard rock repository for spent nuclear fuel is a demanding task, because a high level of safety against rock failure is needed, while the amount of rock reinforcement should be minimised. Because the repository is located in competent rock at a remarkable depth, the failure of intact rock can be the critical factor. Thus an understanding of brittle rock failure under low confinement is a key factor when the stability of the repository during operation is being considered.

The changes in rock mechanical conditions in underground disposal site can be predicted, with adequate accuracy, for the time from the present up to the possible recovery of disposal canisters before the next glaciation. The understanding of the loading during a complete glaciation cycle should be further enhanced.

The brittle behaviour of tested rock and spalling-type rock failure were realised using special testing geometry and axial strain as a control variable. The test results are best utilised as case studies when evaluating the applicability of different modelling methods for the prediction of brittle failure in rock.

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2.3.2 Time-dependent fracturing of rock mass[13]

INTRODUCTION

One of the fundamental functions of the bedrock in the geological disposal of spent nuclear fuel is to provide the repository with a safety barrier which is mechanically stable in both the short and long term. This means that the functions of the buffer and the canister should not be altered significantly by deformations of the rock for long periods of storage. Another safety function of the bedrock is to retard the transport of radionuclides to the biosphere, which, from a mechanical viewpoint, means that the retention properties of the rock may not be seriously degraded by large movements along fractures and fracture zones, or by the extensive formation of new fractures in the near-field and far-field of the repository (Figure 47).

Fracture propagation and the failure of a rock mass can be a sudden and catastrophic event, as in a rockburst or in movements related to excavation. It can also take place very slowly. In the latter case, slow sub-critical crack propagation is thought to play an important role in long-term rock stability.

Studies of brittle rocks have shown a strong connection between time-dependent behaviour and fracture phenomena. Creep tests conducted on brittle rocks consistently show an association with both acoustic emissions and decreasing P-wave velocities, both strong indicators of fracturing as a function of time. Time-dependent deformation is often neglected because the amount of creep is very small in common rock engineering problems. However, in typical repository conditions creep fracturing may lead to delayed failure when long time spans are considered.

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[13] By Mikael Rinne, Fracom Oy.

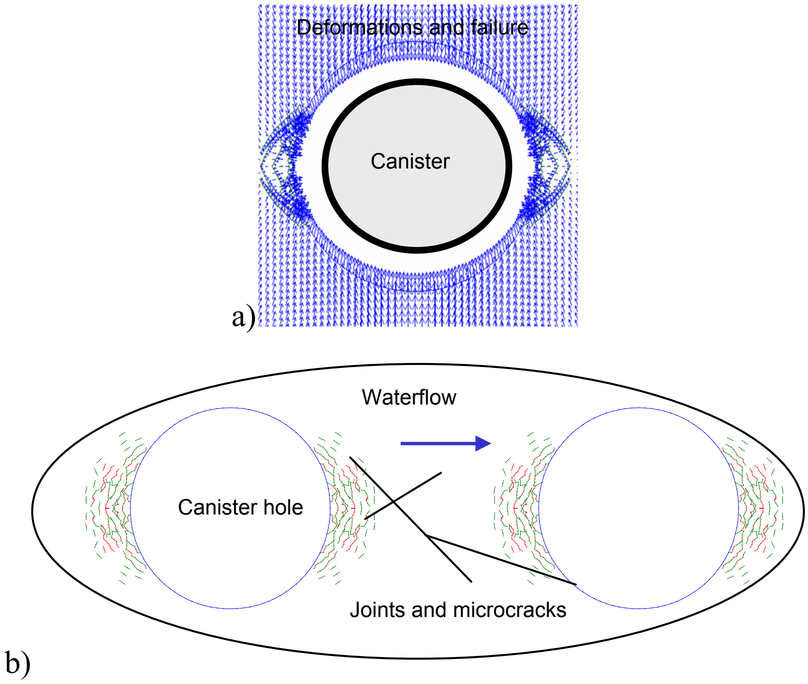

Figure 47. Safety functions of the bedrock for a series of waste canisters: a) to provide a mechanically stable environment for the engineered barrier system (EBS); b) to prevent the transport of radionuclides by groundwater flow (figures not in scale).

It is often observed in the laboratory that fracture initiation starts from microcrack formation at a stress level of 0.3–0.6 of the ultimate uniaxial rock strength. Slowly growing microcracks may coalesce and with time form macro-fractures. Induced stresses close to the repository in deep rock mass can be high enough to initiate this time-dependent fracturing. If insufficient jointing is present, the stresses generated by thermal loading from spent fuel and ice loading (glaciations) can also enhance the creep deformation and lead to delayed rupture.

Recent developments in fracture mechanics theories, laboratory testing methods, and in computational techniques have made it possible to explicitly predict rock mass failure by numerical methods. Although some simulations of creep effects have been presented in the literature, fracture mechanics have not yet been applied to long-term rock mass response, so-called subcritical crack growth, and creep phenomena.

The CREEP project used fracture mechanics principles to predict the time-dependent behaviour of fractured rock mass. The study is focused on the Excavation Disturbed Zone (EDZ) around a rock tunnel. The project participated in the DECOVALEX THMC projects (c.f. Chapter 2.1.1).

THEORETICAL STUDIES AND THE CONCEPTUAL CREEP MODEL

The work was initiated with a state-of-the-art literature review of time-dependent fracturing focused on applications for nuclear waste disposal. The literature study also includes the response of chemical factors in repository conditions. The literature research is an ongoing process and it will continue throughout the DECOVALEX project.

The theoretical studies aim to formulate a conceptual model for time-dependent mechanical effects and two approaches have been investigated more carefully. The first one is based on time-dependent strength reduction resulting from the degradation of cohesive and frictional strength. This approach utilises the Mohr-Coulomb criterion by considering the time-dependent cohesion reduction and mobilisation of the friction angle. It was concluded that this concept could be used to model the time-dependent loosening of rock matrix and to model the delayed slip of rock joints.

The second approach is based on subcritical crack growth principles. This approach utilises the relation between stress intensity at the crack tip and the crack growth rate (Figure 48). The sub-critical crack theory was chosen as the platform for further studies. One major advantage of this approach is that there are well-established methods to monitor and to characterise subcritical crack growth in the laboratory. In addition, the chemical effects can also be considered by including them in the subcritical parameters A and n (See Equation 10).

An analytical solution was established for the relation between crack length and time. The concept is based on the formula presented by (Charles 1958), who stated that most experimental data can be fitted with an expression for the subcritical velocity of the form:

$$v = AK^{n} \tag{10}$$

where v is the crack velocity, A is a constant, K is the stress intensity factor and n the stress corrosion or crack propagation factor.

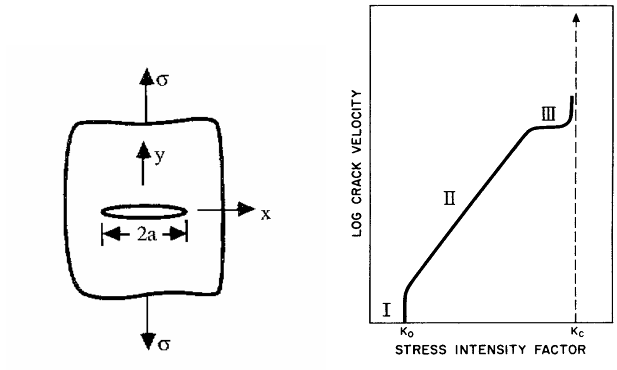

Figure 48. Left: An infinite plate containing a crack under uniaxial tensile loading. Right: Schematic crack velocity/normalised stress intensity factor diagram for subcritical crack growth in rock undergoing stress corrosion. K_0 is the stress corrosion limit. Crack velocity in region II is controlled by the stress corrosion reactions (Atkinson & Meredith 1987).

Assuming that the maximum propagation velocity v_{max}, occurs when $K = K_c$ (K_c = critical stress intensity at failure), and by replacing the stress intensity factor K with

$$K = \sigma\sqrt{\pi \times a}\ , \tag{11}$$

σ is the far-field stress and a is the crack half-length, we get:

$$v = \frac{da}{dt} = A\left(\frac{\sigma\sqrt{\pi \times a}}{K_c}\right)^n \tag{12}$$

by integration the constant crack length is replaced by an effective crack length and we get an expression for the crack length at a certain time by:

$$a(t) = \left\{a_0^{-n/2+1} + (-n/2+1)\left(A\left(\frac{\sigma\sqrt{\pi}}{K_c}\right)^n \times t\right)\right\}^{\frac{1}{-n/2+1}} \tag{13}$$

The derivation of the analytical solution has been described in (Rinne 2005a).

IMPLEMENTATION OF THE CONCEPTUAL MODEL INTO A NUMERICAL METHOD

FRACOD^{2D} is a two-dimensional fracture mechanics code (DDM/BEM) used to analyse the stress-induced rock fractures and deformation of pre-existing joints at discrete locations around rock excavations. It predicts the explicit fracturing process, including fracture initiation and fracture propagation, fracture closure, sliding, and opening. The code was invented in the early 1990s and since then it has been continuously improved and applied in several rock engineering projects (Shen et al. 2005).

The advantage of FRACOD^{2D} for fracture creep modelling is that it takes into account existing joints in the rock mass. The method can also introduce new fractures in the model when the intact rock strength has been exceeded. The model will predict the locus and the direction of creep deformation and creep fracturing on the macro scale. Macro-scale fractures here means fractures larger than the average grain size of the rock matrix.

Another advantage of using FRACOD^{2D} for time-dependent analysis is the type of input parameters the code is using. All the parameters applied are directly measured by laboratory tests and they all have a physically sound meaning, derived from Linear Elastic Fracture Mechanics (LEFM) and the sub-critical crack growth theory.

The conceptual model for rock creep has been implemented into the code according to the plan described in (Shen 2005). As a first step, improvements were made in order to save the stress/displacement history. As a result, the code is now capable of monitoring the stress/displacement history at any point and following the loading path. This aspect has already been used in the DECOVALEX project to model the failure of rock samples subjected to uniaxial and triaxial loads and to compare the stress/strain response with laboratory experiments (Figure 54).

Major code modification has been directed at coupling a time marching algorithm and the creep-fracturing concept in the code (Figure 49).

The improved FRACOD^{2D} code will be used for the analysis of the creep-fracturing of the rock mass using site-specific properties and rock conditions at Äspö HRL. The modelling work has been initiated and is scheduled for completion during the year 2006.

Prior to real data from creep laboratory tests, the accuracy of the numerical approximation was studied and compared with results from the analytical solutions. The subcritical crack parameters applied in the calculations were rough estimates and the results can only be used for the purpose of code testing. The code testing was successful and a detailed description is reported in Fracom's Progress report (Rinne 2005b).

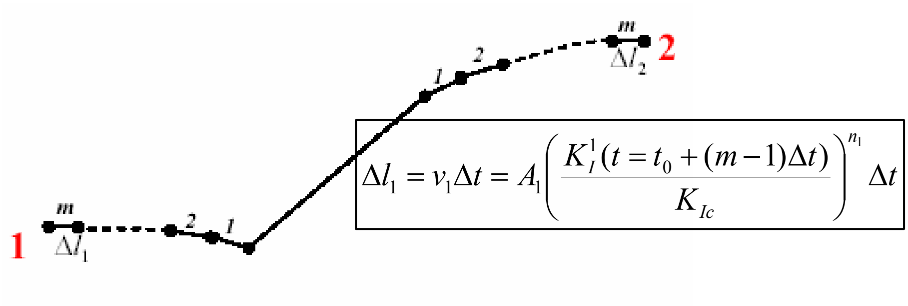

Figure 49. After the length and the direction of the subcritical crack growth have been determined, FRACOD^{2D} will add new tip elements to the original crack tips. This iteration process will be continued until a designed time is reached or an unstable crack growth (i.e. instant failure) is detected.

EXPERIMENTAL STUDIES

Laboratory experiments are necessary in order for it to be possible to calibrate the model for a certain material and for certain initial conditions. Two test programmes have been established, one to define the creep properties of intact rock (Antikainen 2004) and the other for creep-fracturing (Ojala et al. 2004). The experimental work performed was carried out on Äspö diorite. The samples were taken from the APSE pillar experiment site, the reference site for EDZ modelling in DECOVALEX Task B. Experiments at TKK and Geoframes were initiated in August 2005 and some of the tests are still in progress (January 2006). However, preliminary results are available and some of them are presented here. The laboratory work covers the following tests.

The determination and study of the sub-critical crack growth parameters under tensional (mode I) and shear (mode II) loading have been carried out at Geoframes, using:

- Wilkins-[1980] type method with uniaxial tensile tests
- Three point bending tests
- Double torsion tests
- Punch Through Shear tests, (PTS) (Backers et al. 2002).

A sub-critical crack growth curve for shear fracturing (mode II) is presented in Figure 50.

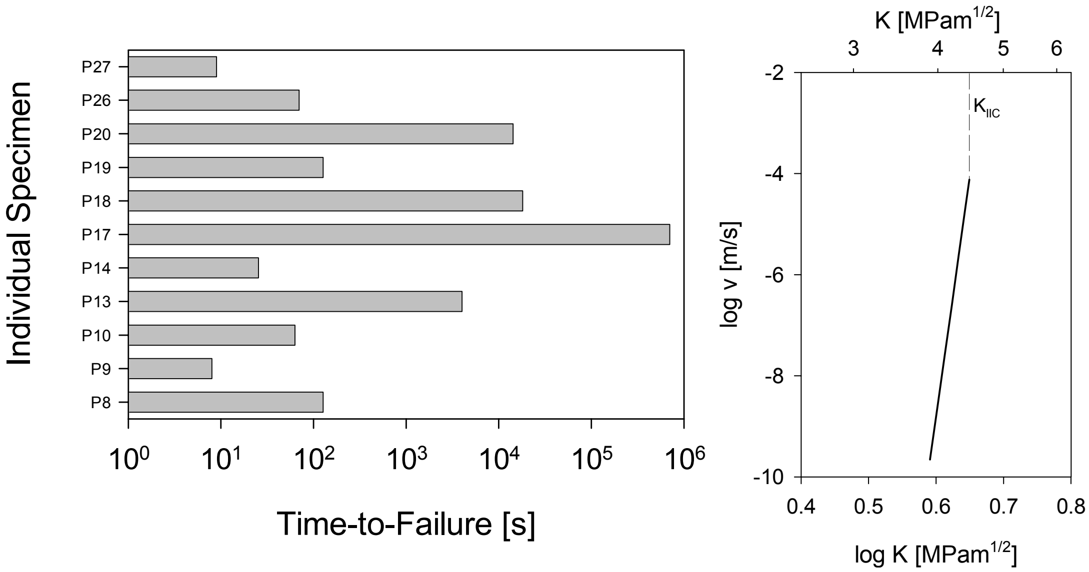

Figure 50. Left; Time-to-failure data from PTS/CP testing under constant load for the individual experiments. Right: Crack velocity, v, vs. stress intensity factor, K, plot for the Mode II sub-critical crack growth equation as derived from PTS/CP testing.

Tests at TKK will clarify the effect of strain rate on the failure to give an estimate of the time to failure and the deformation features of a compressed, initially intact rock sample. The creep behaviour will be modelled in order to test the developed numerical model. The samples have been prepared and tested according to the ISRM Suggested Methods. The tests include:

- Uniaxial compression tests with Acoustic Emission registration (see Table 9)
- Triaxial compression tests
- Brazilian tests (see Figure 51 and Table 9)
- Strain rate stepping tests suggested according to (Lockner 1998).
- Microscopic studies of the damage.

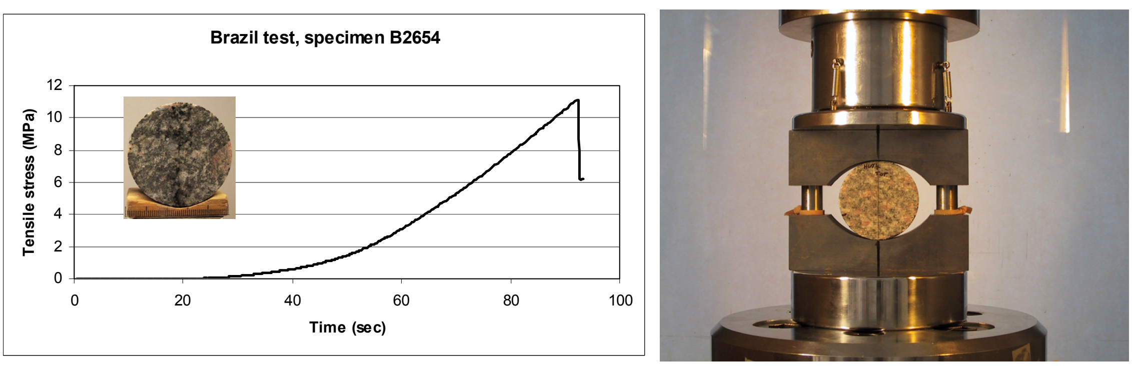

Figure 51. Brazilian tests have been performed to declare the tensile strength of Äspö diorite.

Table 9. The location, dimensions, saturated density, uniaxial compressive strength UCS, Modulus of Elasticity (Young's modulus) E, and Poisson's ratio of uniaxially tested specimens. The indirect tensile strength values at the same locations are presented for the purpose of comparison.

Drillhole N:o	Depth (m)	Length (mm)	Diameter (mm)	Density (kg/m³)	UCS (MPa)	Tensile strength (MPa)	E (GPa)	Poisson's ratio
KF0066A01	26.40	129.3	50.8	2720	185.3	11.1	61.6	0.32
KF0066A01	36.04	129.3	50.8	2673	270.2	17.1	70.1	0.31
KF0066A01	38.95	129.8	50.8	2683	300.0	14.9	69.9	0.31
KF0066A01	49.89	129.8	50.8	2670	257.4	16.6	70.4	0.29
KA3376B01	12.08	130.3	50.8	2743	182.1	13.5	63.0	0.32
KF0069A01	45.77	129.5	50.7	2677	301.6	18.1	71.2	0.32

An abstract of the laboratory work has been accepted for the GeoProc 2006 conference (Nanjing, China May 22–24 2006). Beside laboratory experiments at TKK and GeoFrames, the chemical effects on rock strength have been studied at SP Laboratory, Borås, Sweden (Ann Bäckström, SKB, Figure 53).

DECOVALEX THMC TASK B

The Task B research plan has been documented in (Hudson 2005), and the work is organised into six phases (Figure 52).

The Task B sub-tasks, or Phases, are essentially sequential in nature, but they are being carried out to some extent in parallel because of the significant overlaps between them. The progress of Task B has been comprehensively presented in the Workshop and meeting reports. Here only the Fracom teams' contribution to Task B is presented. The Fracom teams' focus is mainly on Phases 2, 3, and 4, but contributions will also be provided to support the Phase 1 and 5 studies.

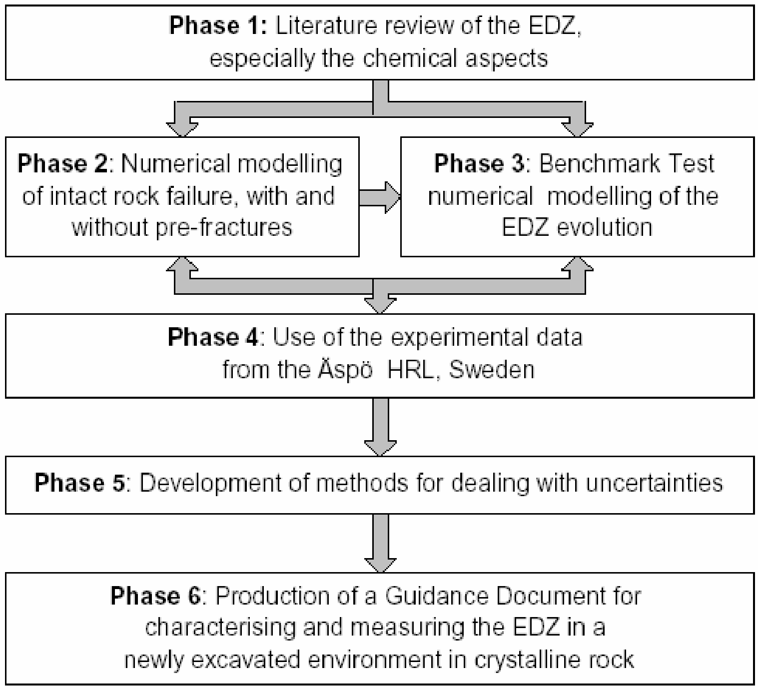

Figure 52. Flowchart of the six phases comprising the DECOVALEX-THMC Task B programme of work.

Task B, Phase 2

Phase 2 models aims to explain the complete failure of intact rock in compression. FRACOD models were set up to numerically describe the initial microcracking, deformation, and failure of intact rock. Both uniaxial and tri-axial laboratory tests and the effect of pre-existing fractures on rock failure have been modelled (Figure 54).

The work was initiated by setting up generic rock failure models. The values of the parameters used in modelling are those for the Äspö diorite as quoted in (Staub et al. 2004). The set of parameters ensures that all research teams will simulate the failure using the same parameters.

The Fracom team simulated sample-specific stress/strain curves representing laboratory tests conducted at TKK (Kuula 2003, Särkkä & Eloranta 2003). Beside the axial strain response on loading, the radial response was also modelled for a number of confining pressures (Figure 55). The fracture initiation, fracture propagation, and peak strength match well with the laboratory results. The preliminary Phase 2 models have been presented at DECOVALEX workshops and Task Force Meetings. A paper related to the study will be presented at the EUROCK'06 Conference (Rinne et al. 2006).

Figure 53. A rock specimen on completion of the complete stress-strain curve testing in uniaxial compression. Äspö diorite rock core sample; SP Laboratory, Borås, Sweden (Hudson 2005).

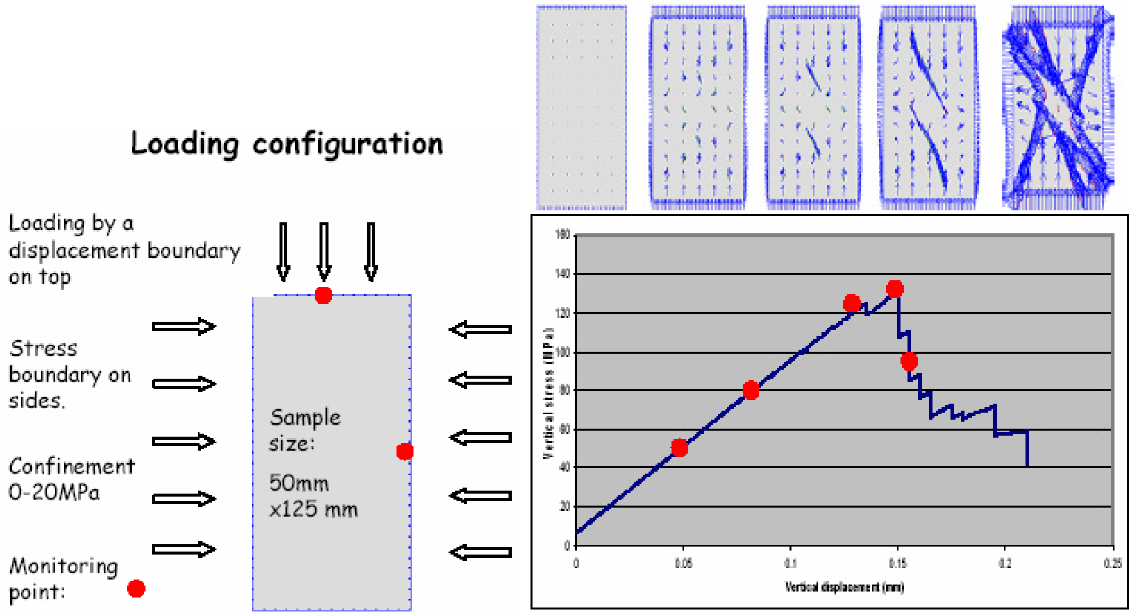

Figure 54. Left. Loading configuration for generic models. Right: Complete stress-strain curve calculated for a generic uniaxial compression test for an intact rock specimen.

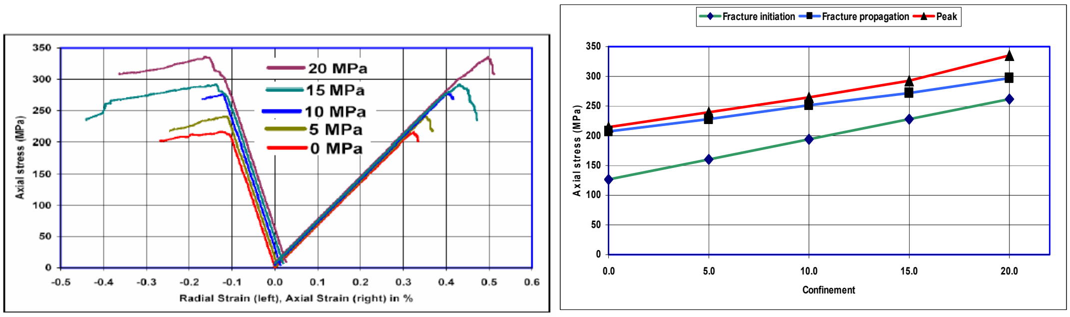

Figure 55. Left; Axial and lateral strain from numerical strength tests. Confining stress of 0 to 20MPa has been applied. Right: Modelled effect of confinement on fracture initiation, fracture propagation and peak strength.

<u>Task B, Phase 3</u>

The main scope of the work of Task B, Phase 3 is the BMT calculation of THMC processes in the EDZ around the emplacement drift of a nuclear waste repository. The purpose is to study the mechanical responses and long-term time effects of EDZ from THMC sources from a pre-emplacement period of 30 years and a post-closure period of up to 100,000 years. The basic layout for EDZ is shown in Figure 56, (Rutqvist et al. 2005). The representation of EDZ consists of 2 types of domain, wall block models with fractures (Figure 57), and a near field model (Figure 58). The final layout of the near field model is under discussion.

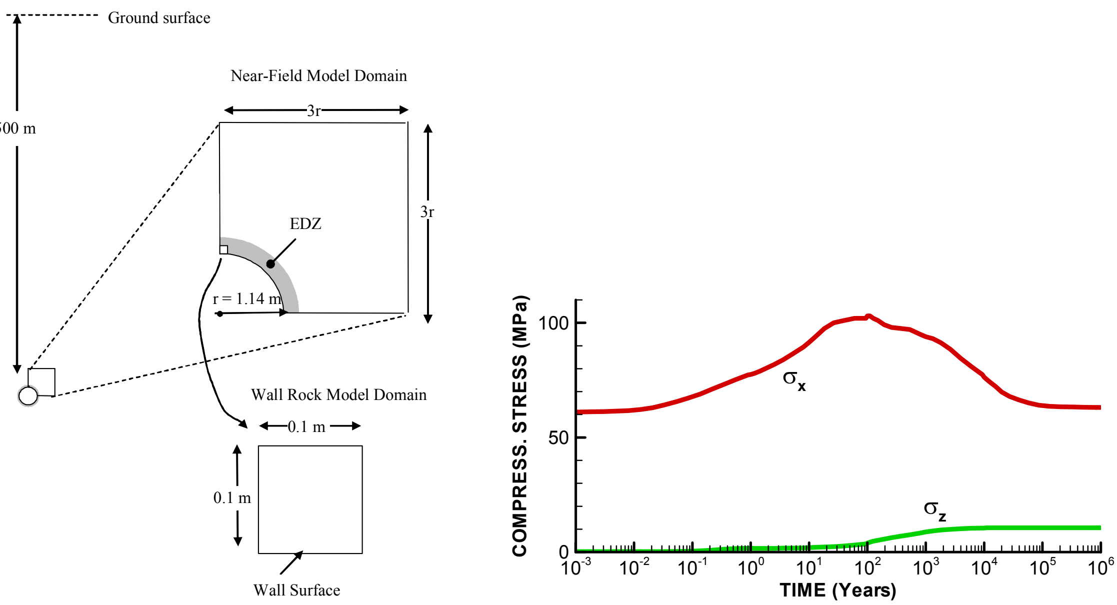

Figure 56. Basic layout for BMT modelling and boundary conditions.

Suggested modelling stages for Phase 3 are as follows:

Stage 1 – Linear thermal-hydro-elastic modelling: model inception with linear elastic properties

Stage 2 – Non-linear, elasto-plastic failure modelling: extend model to include non-linear and elasto-plastic properties for failure analysis

Stage 3 – Time-dependent failure modelling: extend model to include time-dependent changes in mechanical properties for analysis of creep and mechanical degradation

Stage 4 – Chemo-mechanical modelling (optional): extend model to include simplified chemical modelling of time-dependent pressure solution/stress corrosion or other chemo-mechanical effects

Stage 5 – Full THMC modelling (optional): implement chemo-mechanical model developed in Stage 4 to link THC and THM models into a fully coupled THMC model.

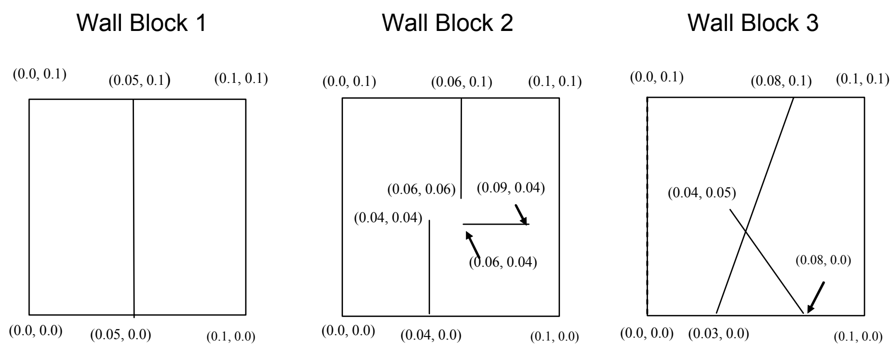

Figure 57. Detailed configuration of wall block model for BMT Task B.

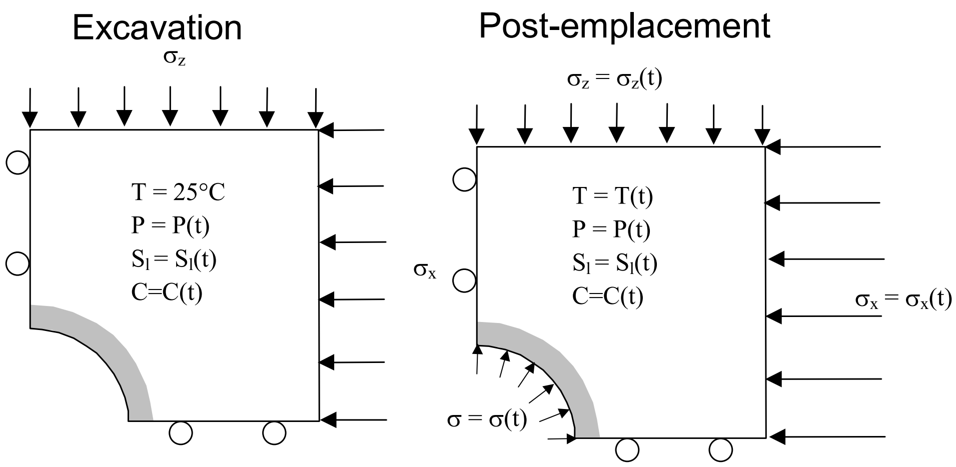

Figure 58. Detailed configuration of near field model for BMT Task B.

The first-year modelling (2005) mainly handled Stage 1 and Stage 2 and they were dealt with simultaneously. Time-dependent failure modelling in Stage 3 will be performed with the new updated code, using sub-critical fracture parameters recently defined by laboratory tests.

Preliminary Phase 3 block models have been reported by (Lee & Rinne 2005) and the central results can be summarised as follows:

1) There was no fracturing for an intact rock block model and Wall block model 1 with given time evolution of boundary stresses until 100,000 years of emplacement.
2) Slight damage near pre-existing fracture tips occurred as a result of the initiation of elastic fractures in Wall block model 2. However, the model remained stable for the whole period of time.
3) Presence of pre-existing fracture clearly affects the distribution of displacement.

A joint DECOVALEX abstract dealing with the modelling of Task B, Phase 3 has been accepted for the GeoProc 2006 conference (Nanjing, China, May 22–24, 2006). A more detailed paper presenting the Fracom teams' models has also been accepted.

CONCLUSIONS

A conceptual model for time-dependent crack growth and creep deformation has been established and it has been successfully implemented in a numerical code.

The numerical approximation demonstrates good accuracy compared with the analytical solution. Accurate time-to-failure calculation calls for accurate definition of the model geometry and loading conditions. Slight inaccuracy in the initial stress intensity at the crack tip leads to significant errors in the time-to-failure calculations. The pre-defined length of time steps also dictates the accuracy of the time-to-failure calculations. A very accurate model leads to long calculation times. However, long-continuing calculations are stable even with a conventional PC.

According to preliminary calculations, the time-dependent model works well in tensional loading. Further work is mainly focused on creep behaviour in compression. The ongoing laboratory tests are mainly being conducted under compression. The outcomes of these tests will be thoroughly analysed, modelled, and reported. Code modification and debugging is an ongoing process.

On the basis of the Fracom teams' work on DECOVALEX Task B, some preliminary conclusions can be drawn. The improved FRACOD code is capable of accurately simulating the failure of core samples from laboratory tests. An important achievement is the capability to model realistically the brittle axial and lateral response during the failure process. The success of modelling Class II behaviour in the post-peak failure region is an important achievement.

Laboratory experiments aimed to develop experimental testing methods for the determination of the subcritical fracture growth parameters under Mode I and Mode II conditions. It has to be concluded that the proposed methods have the potential to determine these parameters. The experimental work has yielded a set of data describing the fracturing behaviour of the Äspö diorite (Table 10).

Table 10. Summary of the parameters determined describing fracture growth.

Mode	**K_C** (fracture toughness) [MPa $^{m1/2}$]	**n** Subcritical crack growth index	**-log A** Subcritical scaling factor
I	2.74 ± 0.05	48	25
II	4.46 ± 0.24	95	66

The saturated samples in this study show about 12% lower fracture toughness compared to the former studies on the Mode II fracture toughness of Äspö Diorite. Although the specimens were not obtained from the same location, there is an indication that K_{IIC} is lowered by the presence of a fluid. This is in analogy with the behaviour of the Mode I fracture toughness in this study. The determined fracture toughness is 2.7, about 15–30% lower compared to earlier tests using dry samples.

A statement that can be made is of a fundamental character. The constant load experiments on the PTS/CP set-up have shown that Mode II time-dependent fracturing behaviour exists under laboratory conditions at ambient pressure conditions. This has never been shown before.

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2.3.3 Bedrock fracturing and hydrogeology[14]

INTRODUCTION

The main units of the crystalline bedrock of the Fennoscandian shield formed more than 1.3 billion years (Gy) ago during the orogenic (mountain-building) processes. Rocks crystallised under high pressure and at high temperatures from molten magma or re-crystallised in the metamorphism of earlier sediments (clays, sands, muds etc.) or volcanic rocks (lavas or tuffs). Characteristic deformation processes associated with orogenies are the formation of fold and fault structures in the rocks. These tectonic processes manifest themselves in the bedrock as deformation structures which, depending on their genetic setting, may exhibit ductile or brittle characteristics. Especially in the latter case, the emplacement of magmatic veins or rock recrystallisation from intruding fluids may have taken place. Large-scale faulting during orogeny leads to the formation of extensive overthrusts of rock masses, which may now appear as major tectonic zones associated with changes of rock type, metamorphic domains, or other rock characteristics.

The bedrock of southern Finland was formed in the Svecofennian orogeny about 1.9–1.8 Gy ago, when the current migmatitic schist belts and associated plutonic rocks formed. The last clearly observable major tectonic feature in the bedrock of southern Finland is the mid-Proterozoic extension and rifting (NW-SE), which led to the deposition of Satakunta sandstone in a graben structure. The extensive Rapakivi granites and associated mafic dykes in southern Finland are thought to be associated with the rifting process. The Precambrian of southern Finland concluded with the intrusion of late Proterozoic (post-Jotnian) diabases 1.27 Gy ago. The last major orogenic event affecting the Scandinavian Peninsula was the formation of Norwegian Caledonides

[14] By Lasse Ahonen, GTK.

about 400 million years ago, but its effects on the Finnish part of the Fennoscandian shield are considered to have been rather small.

For the Finnish bedrock, the span of the last 1500 million years has been mainly a time of erosion and denudation of the originally deep-seated crystalline rocks. The release of the load above has led to the uplift of the crust, in which horizontal fracturing has formed as a result of the pressure release. Continuous plate tectonic movement of the continents has subjected the Fennoscandian shield to horizontal stress, which tends to dissipate the stress continuously in small quakes of the fracture zones. Additionally, the Earth's crust is subjected to diurnal variation of vertical forces applied by the gravity of the Moon and Sun (tidal force). During the last 0.5 million years or more the Fennoscandian shield has been subjected to repeated glaciations in cycles of about one hundred thousand years.

SCOPE OF THE PROJECT

In the Finnish nuclear waste disposal concept bedrock is an important component of the multi-barrier system. From the hydrogeological point of view, a characteristic feature of the Fennoscandian crystalline bedrock is the prominent fracturing on different scales, while the total and effective porosities are very low. Consequently, profound understanding of the mode of bedrock fracturing is a key issue and the starting point for the conceptualisation of the groundwater flow and radionuclide transport processes.

The fundamental aim of the "Bedrock fracturing and hydrogeology" project (see Annex A) is to examine the bedrock "structures" through a hydrogeologist's eyes and thus to support the efforts to model the groundwater flow and radionuclide transport processes adequately. On a more practical level, the project deals with the research activities that are required – and have been applied – to produce input data for the geohydrological models. The approach is based on one hand on a review of earlier structural and hydrogeological site studies (mainly the Palmottu and Outokumpu sites) and on the other hand on new site studies (Kopparnäs). The integration of the research 'tradition' into learning through doing is a guideline.

The geological outlines of the bedrock of south-western Finland are shown in Figure 59. In addition to the distribution of the main rock types, major deformation zones are also indicated. Kopparnäs is the main target of the present study, while Palmottu was a site thoroughly studied in an earlier project. An important aim of the present project is to produce non-site-specific information and expertise to be applied to the site studies of Olkiluoto, which is situated in an analogous geological environment.

Fracturing is a basic structural property of crystalline bedrock, which can be observed on all scales, from major geological deformation zones to microscopic fissures. Fracturing can be understood as nested systems in which a large bedrock block defined by large-scale structures is divided into smaller subsystems. As applied to nuclear waste disposal, this has led to the 'block mosaic' concept, which – from the hydrogeological point of view – may imply the adoption of the concept of a 'hydraulic cage' surrounding the block. Fracture zones/fractures on different scales may have significantly different hydraulic properties, which – if verified – can be accounted for in the hydrogeological models.

The construction of a structural model of the bedrock is an essential part of site characterisation programmes for nuclear waste disposal. The models typically describe the spatial distribution of lithological units and zones of weakness. As a coarse approximation, certain hydraulic properties may be attributed to the structures identified. However, direct measurements of the hydraulic conductivity of the bedrock often seem to indicate that the distribution of water-conducting zones may differ from the distribution of zones otherwise interpreted as broken.

Structural studies of the bedrock include a variety of methods, including air-borne and surface-based mapping and geophysical measurements, examination of the bedrock by (cored) drilling, drill core studies, borehole geophysical measurements and, nowadays increasingly, borehole videosurvey. In the Kopparnäs site study, all these approaches were used and compared with the borehole hydraulic information so far available from a fluid logging test.

LINEAMENT INTERPRETATION

The purpose of the work was to carry out an integrated interpretation of geophysical and topographic lineaments for the Kopparnäs area (Figure 59). In the context of the work, lineaments were considered as indications of possible deformation zones. The work aimed to survey possible water-bearing zones in the crystalline bedrock.

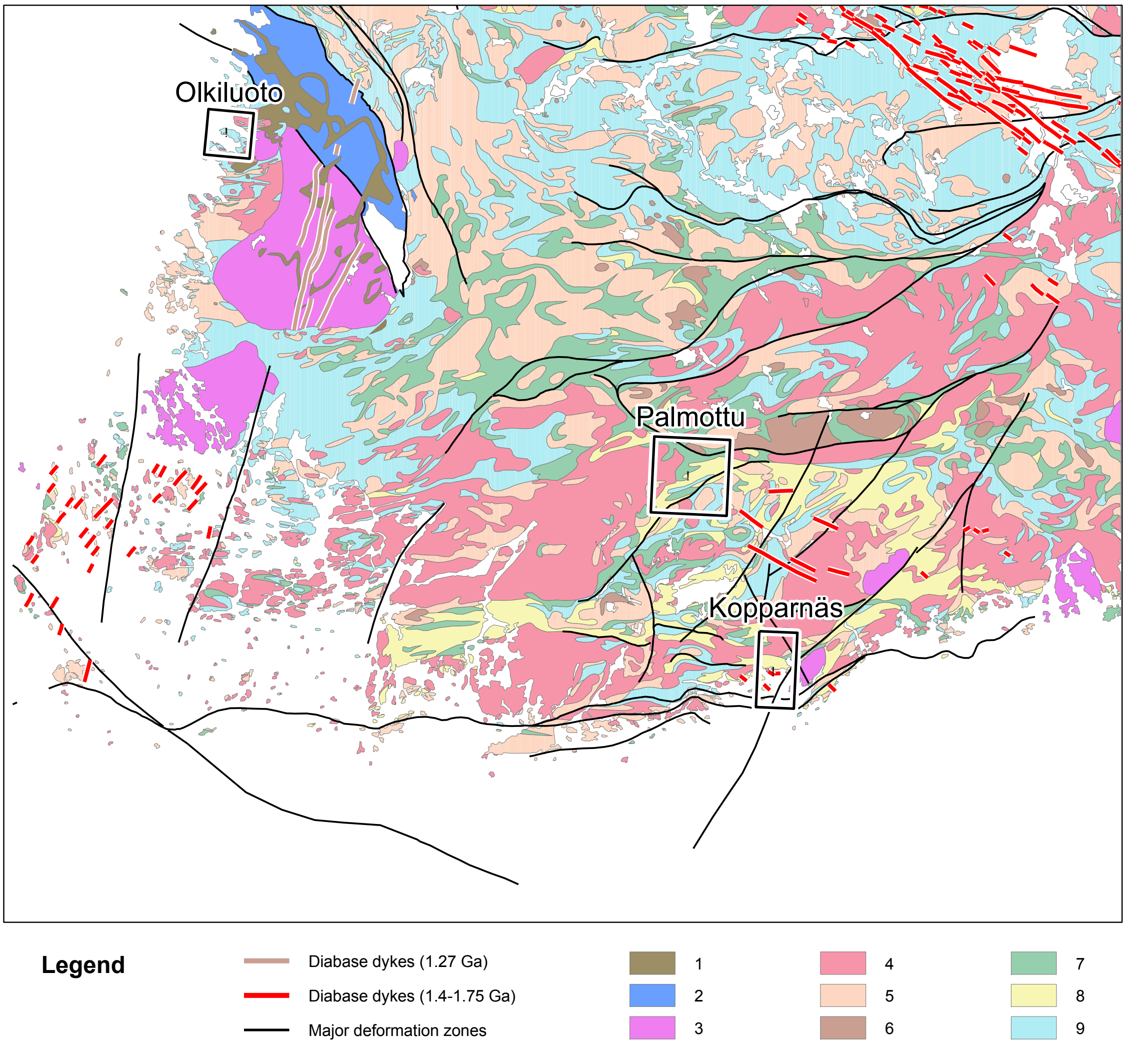

Figure 59. Geological map of southern Finland (modified from Korsman et al. 1997). Rock types: 1) Olivine diabase (1.27 Gy); 2) Satakunta sandstone (about 1.3 Gy); 3) Rapakivi granite (1.57–1.65 Gy); 4) Microcline granite (1.82–1.84 Gy); 5) Granodiorite, tonalite (1.87–1.89 Gy); 6) Gabbro, diorite (1.87–1.89 Gy); 7) Amphibolite (1.88–1.9 Gy); 8) Felsic/intermediate schists/gneisses (1.88–1.9 Gy); 9) Mica schist/gneiss (1.88–1.9 Gy). Lineament models of the sites indicated are given by the following authors: Korhonen 2005 (Kopparnäs), Kuivamäki et al. 1991 (Palmottu), Korhonen et al. 2005 (Olkiluoto).

Data

Aerogeophysical and topographic data were used. The aerogeophysical data included magnetic, dipole-source electromagnetic (EM), apparent resistivity, and radiometric data sets. The topographic data comprised the Digital Elevation Model (DEM) of Finland and aerial stereographic photos. The aerogeophysical data for the research area were measured in 1996 by GTK. The nominal flying altitude was 40 metres and the line spacing 200 metres. The data included measurements of the geomagnetic field intensity (corrected for diurnal variations), the real and imaginary components of the induced EM field at two frequencies (3125 and 14368 Hz), and gamma ray spectrometry. Apparent resistivity estimates calculated from the measured real and imaginary component data were also used.[15]

A topographic data set covering the research area was extracted from the DEM, which covers the whole of Finland. The DEM was created by the National Land Survey of Finland by interpolating elevation contours to a regular grid with a cell size of 25 m × 25 m. The standard error of the DEM for the research area is 1.76–5 m[16]. More detailed topographic data were obtained from aerial stereographic photos (National Land Survey of Finland).

Method

The work proceeded in four stages: (*i*) data processing and map compilation; (*ii*) interpretation of method-specific lineaments; (*iii*) integration of the method-specific lineaments into coordinated lineaments, and (*iv*) integration of the coordinated lineaments into linked lineaments. All the interpretations were carried out utilising GIS software.

All the data were non-uniformly spaced (the aerogeophysical data for methodological reasons and the DEM because it was transformed from the YKJ coordinate system into the KKJ coordinate strip 2), thus requiring interpolation to regular grids (gridding) to facilitate map compilation. The data sets were gridded using the minimum curvature method. Once the data had been gridded, several hill-shaded colour maps were compiled from the grids to act as the basis for lineament interpretations.

Each data set (magnetic, dipole-source EM, apparent resistivity, radiometric, and topographic) was interpreted independently. Lineaments representing possible deformation zones were identified and digitised into vector format and stored in method-specific shapefiles. Each lineament was assigned unique attributes documenting its interpretation (e.g., length, trend, and the uncertainty in the interpretation). The attributes were stored in attribute tables linked with the shapefiles.

Once the method-specific lineaments had been interpreted, they were viewed together on top of the maps visualising the method-specific data sets. Then, those method-specific lineaments that were judged to represent the same deformation zone were

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[15] http://www.gtk.fi/aerogeo/

[16] http://www.maanmittauslaitos.fi

integrated into coordinated lineaments and stored in a shapefile. The coordinated lineaments were assigned unique attributes documenting their interpretation. The attributes were based on the attributes of the method-specific lineaments that were integrated together. The attributes were stored in an attribute table linked with the shapefile for the coordinated lineaments.

Finally, the coordinated lineaments were viewed on top of the maps visualising the method-specific data sets. Then, those coordinated lineaments that were judged to represent the continuation of the same deformation zone were integrated into linked lineaments and stored in a shapefile. The linked lineaments were assigned unique attributes documenting their interpretation. The attributes were based on the attributes of the coordinated lineaments that were integrated together. The attributes were stored in an attribute table linked with the shapefile for the linked lineaments.

Results

The method-specific lineaments comprise 244 topographic, 252 magnetic, 107 dipole-source EM, 125 apparent resistivity, and 94 radiometric lineaments. The integration of the method-specific lineaments into coordinated lineaments produced one shapefile and one attribute table that contain 861 lineaments and their attributes. The integration of the coordinated lineaments into linked lineaments produced one shapefile and one attribute table that contain 502 lineaments and their attributes.

Figure 60 shows the linked lineaments plotted on a map of the research area. A regional major tectonic zone passes the Kopparnäs site (square) in the southeast corner. A major portion of the long lineaments is approximately in the east-west direction. When the Kopparnäs site is studied on a more detailed scale, more short lineaments can be observed, but the general pattern remains the same (Figure 61).

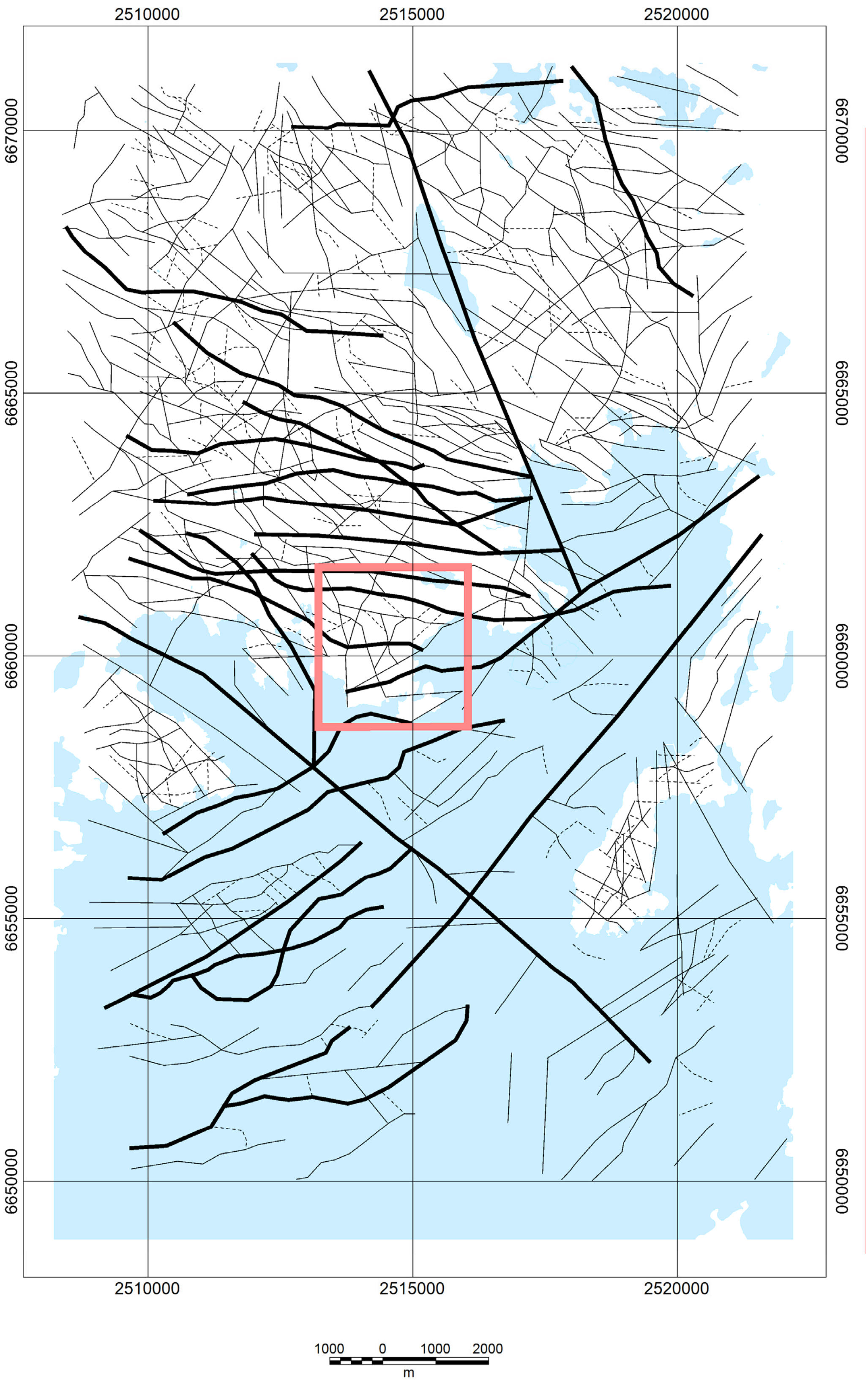

Figure 60. Results of the integrated interpretation of geophysical and topographic lineaments for the Kopparnäs area. Lineaments are classified according to their apparent length: bold line = length > 5 km.

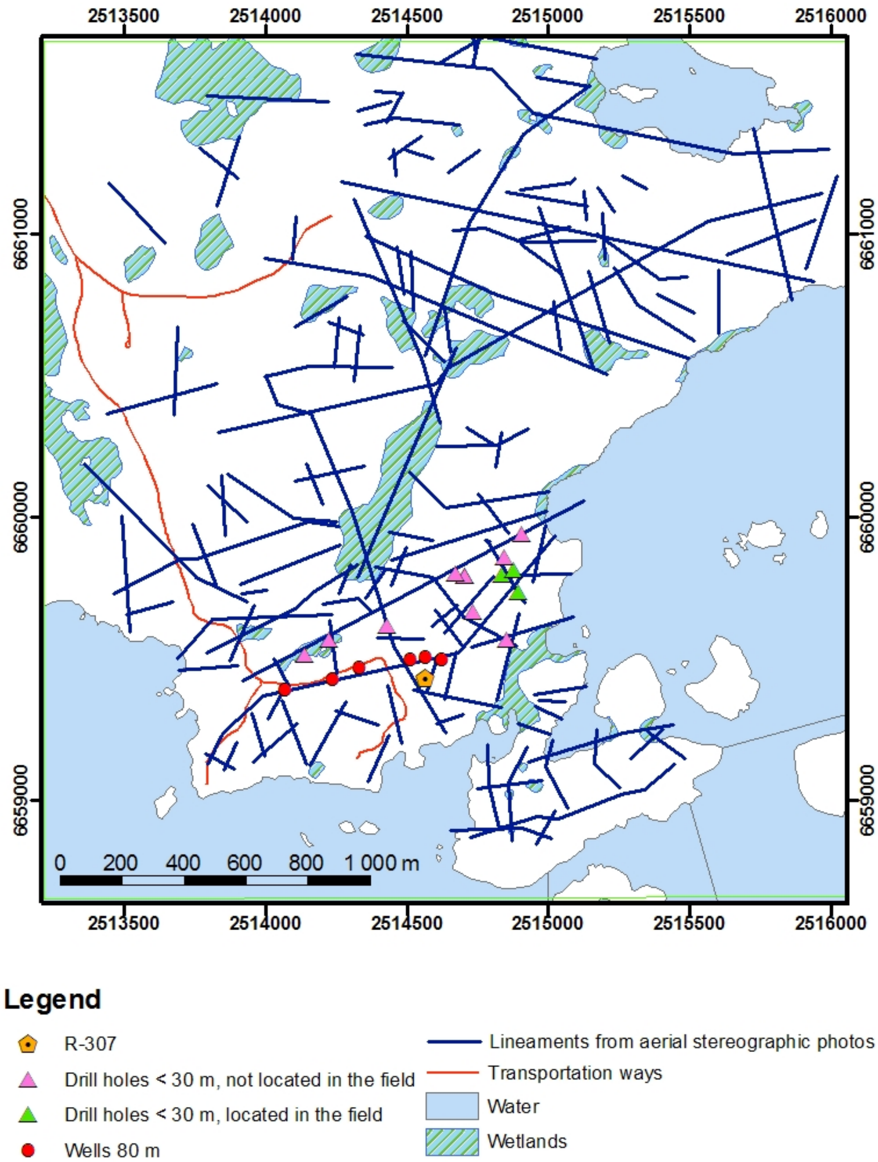

Figure 61. Lineaments at the Kopparnäs site interpreted from aerial stereographic photos. New research borehole are denoted by yellow symbols, old cored boreholes (depth < 30 m) are denoted by triangles (green = found, pink = not found), bored wells denoted by red circles.

DRILLING, DRILL CORE, AND VIDEO SURVEY

On the basis of the lineament interpretation, a 233-m-long new research borehole was drilled at the Kopparnäs site in order to study the geological structure indicated by one of the major lineaments (Jääskeläinen & Korhonen 2005). To keep the core sample as intact as possible, a triple-tube drilling technique was used. The drill core was photographed and studied in detail, with a special emphasis on fracturing, which was classified in categories (Figure 62). The aim was to distinguish open fractures with signs of water contact.

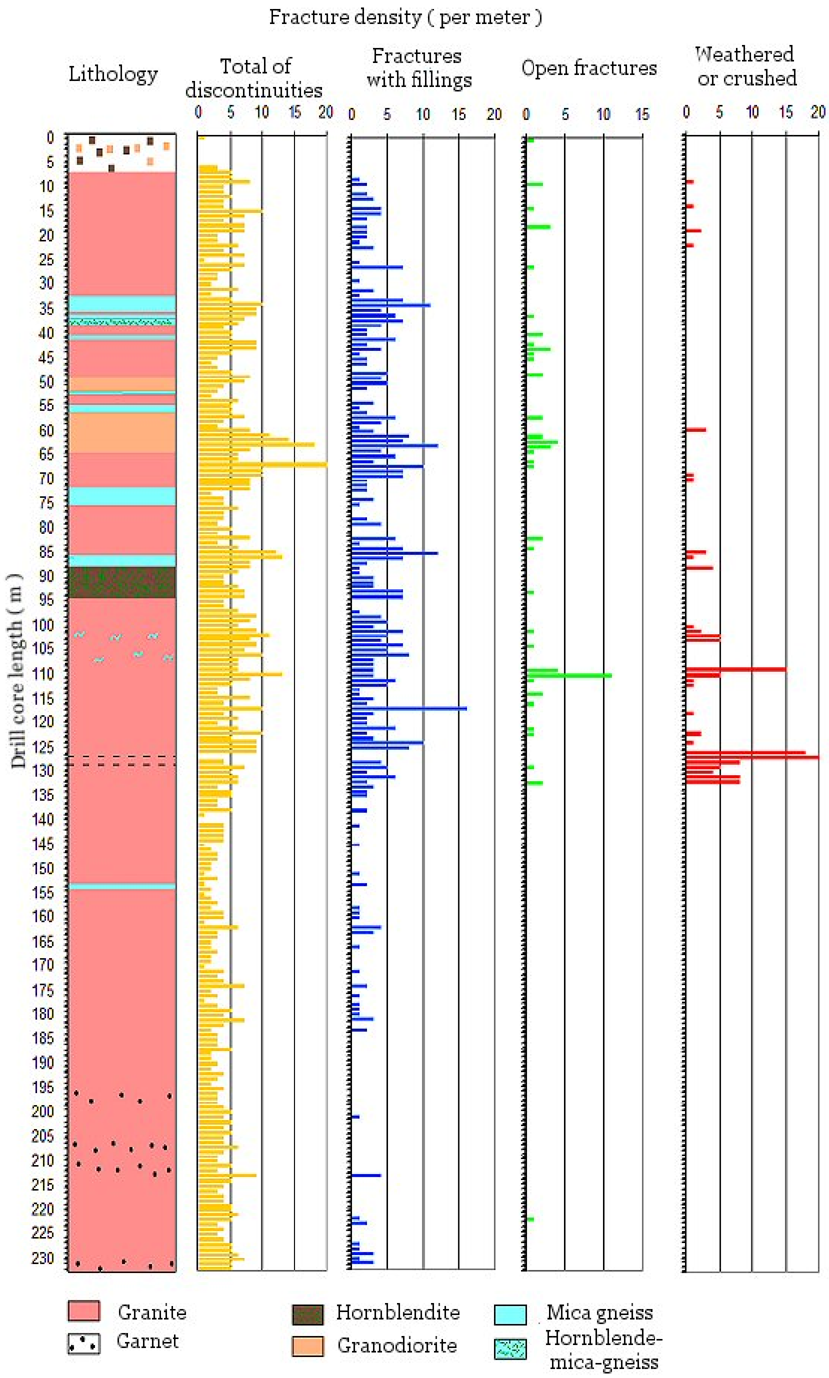

Figure 62. Core log of Kopparnäs borehole R-307 (Jääskeläinen et al. 2005a).

It is evident from the drill core study alone that only a minor portion of all the observed natural fractures are clearly open and water-conducting. On the other hand, the relative amount of open fractures can also be underestimated in a drill core, if the effects of water contact are not seen on the fracture surfaces. In determining the true openness of the fractures, the videosurvey proved to be an incomparable method, because even *in situ* fracture apertures can be estimated. Figures 63 and 64 show examples of a comparison between a drill core sample and the corresponding borehole wall.

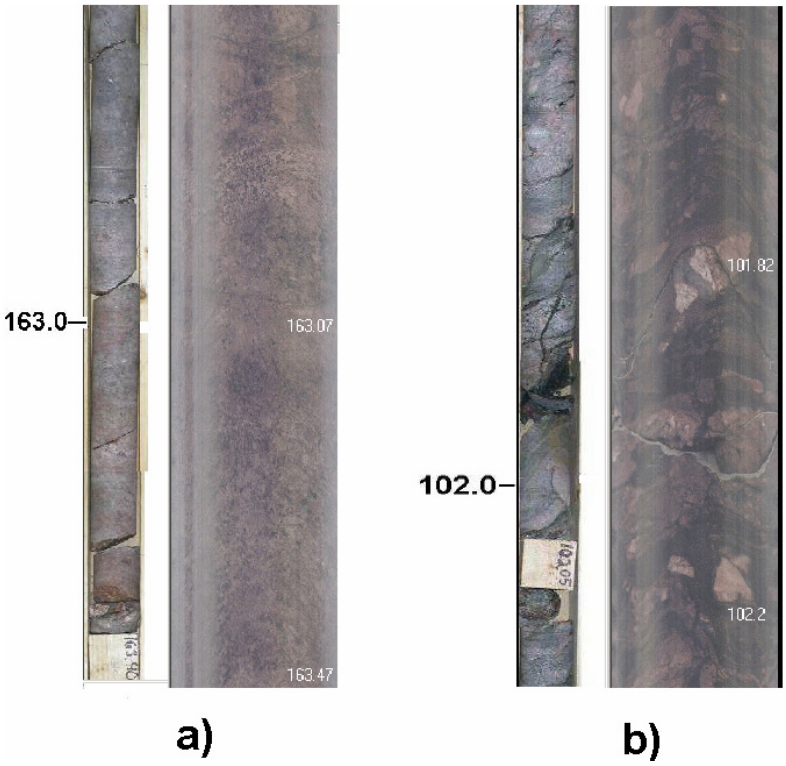

Figure 63. Comparison of drill core and drill-hole wall (picture angle 360^o) at Kopparnäs R-307. A) At a depth of 163 m about 7 fractures/m opened during drilling, while no visible indication of fracturing was observed in the borehole wall. B) Strongly broken core sample and drill-hole wall at 102 m.

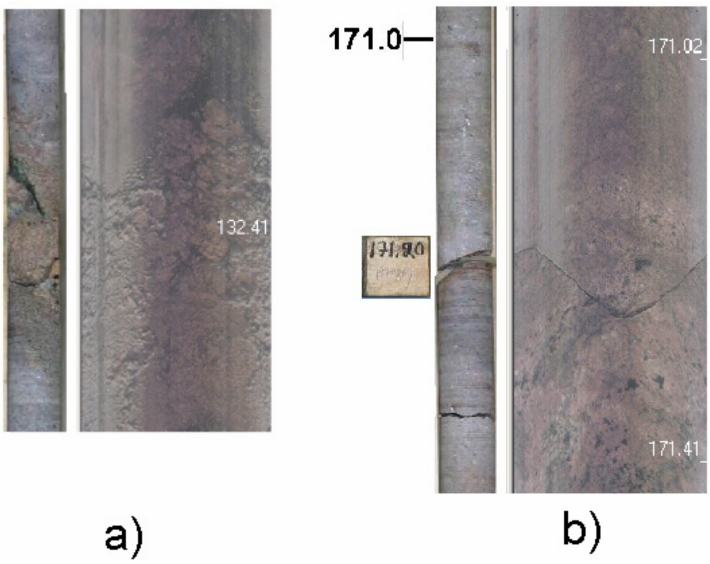

Figure 64. Comparison of drill core and drill-hole wall (picture angle 360^o) at Kopparnäs R-307. A) Strongly altered, porous rock at 132 m, and B) A single fracture in an otherwise intact rock at 171 m.

Kopparnäs borehole R-307 was drilled using water taken from a near-by bored well and labelled with sodium fluorescein (500 mg/m^3). Monitoring of the flushing water indicates that there was a near-surface connection(s) between the drilled borehole and the well. However, even during drilling it became evident that flushing water in the new borehole was rapidly exchanged for saline water. Borehole water sampled half a year after the drilling indicated high salinity, while the flushing water used in drilling had completely disappeared. Water salinity in the borehole shows changes at certain depths (Figure 65). Analysed total salinities at 100 metres and deeper are clearly higher than the present salinity of the Gulf of Finland. A sharp increase in salinity is associated with the crush zone observed at the depth range of 100–130 m. The factors affecting the analysed water composition remain to be resolved, but – based on the total salinity and distribution of main components – a contribution of the ancient Litorina sea stage seems likely.

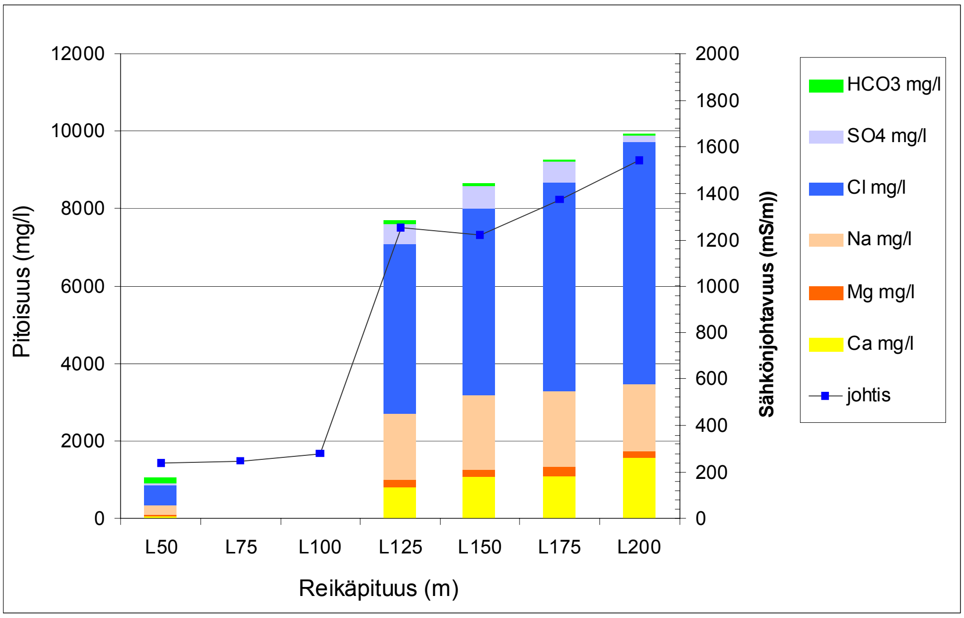

Figure 65. Water composition in Kopparnäs borehole R-307 as a function of depth. The tube sampling technique was used. In addition to the main chemical components, the electrical conductivity (EC) of the water is also shown (mS/m at 25^{o}C).

GEOPHYSICAL BOREHOLE MEASUREMENTS

During the summer and autumn of 2005 geophysical borehole measurements were carried out in the Kopparnäs borehole R-307 (Jääskeläinen et al. 2005b). The aim of the study was to evaluate the feasibility of different methods for studying water conductive fracturing. Another major study conducted was the interpretation of the results of the geophysical borehole logging of the 2500-metre-deep Outokumpu borehole (Tarvainen 2006). The drilling of the 'super-deep' borehole, including geophysical logging, was carried out as a part of the depth conversion programme between Russia and Finland, which did not include interpretation of the results.

A comprehensive set of borehole geophysical data from the Outokumpu deep borehole is available. Different measurements include galvanic logs in different electrode configurations, magnetic susceptibility and field measurements, fluid resistivity and temperature, acoustic logs, radiometric measurements, and borehole calibre. Hydraulic tests and their interpretations (Drill Stem Test, DST) after the drilling of each 500 m were also included in the drilling contract. The length of the test sections varied from about 50 to 80 metres (space between a mechanical packer and concurrent bottom of the borehole). The specific aim of the present study is to compare the results of geophysical measurements with the results of the hydraulic tests performed in the sections.

After a detailed depth correction procedure, the methods were analysed and compared with hydraulic tests to locate possibly water-conducting fracturing. The following methods were concluded to be the most promising with respect to the target: bedrock resistivity logs applying *normal* and *microlaterolog* configurations, acoustic P/S-wave ratio, epithermal neutron log, bedrock density, and borehole calibre. Further on, cut-off values were ascribed to the parameters, defining their potential to indicate water-conducting structures. However, these cut-off values are specific to the rock type. An example of the results is given in Figure 66.

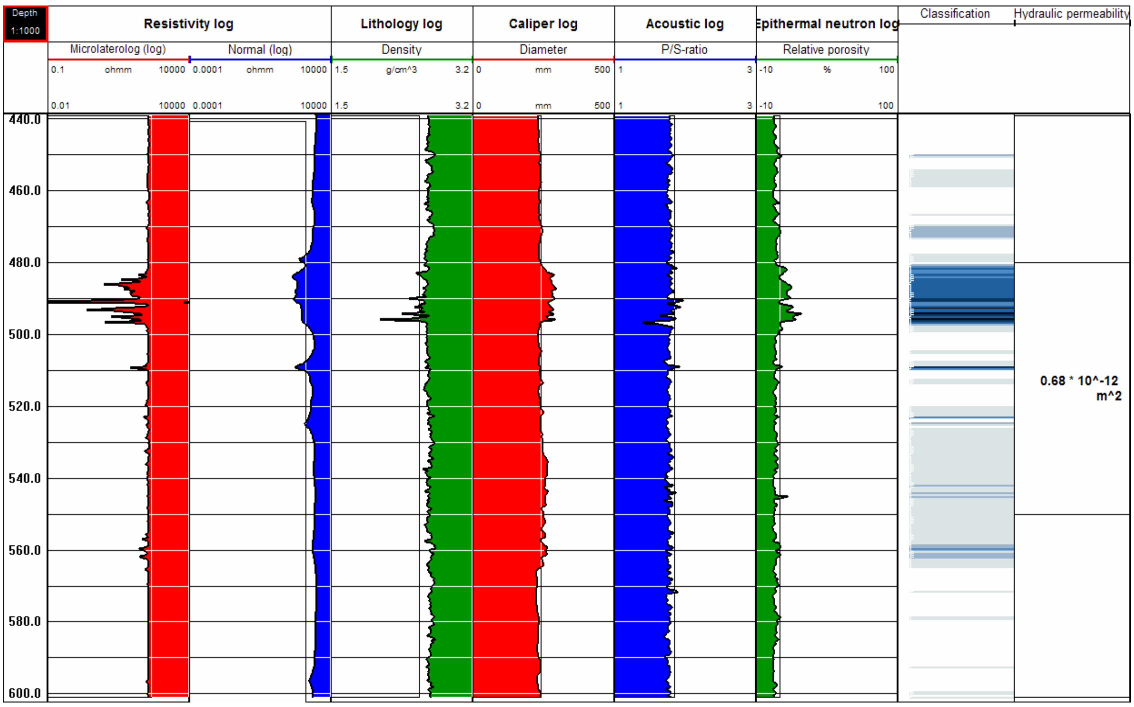

Figure 66. An example of specific borehole geophysical logs in the borehole section 480–550 m, having a known (high) hydraulic conductivity. The given permeability value corresponds to the hydraulic conductivity value $K \approx 0.7 \cdot 10^{-5}$ m/s for the section.

Geophysical borehole measurements at Kopparnäs were taken with the Wellmac Logging system (Malå Geoscience). The equipment includes a measuring probe, a probe with power supply/modem, a surface unit with software, a winch, and a depth-measuring wheel. Two different measuring probes were used. One probe measures galvanic bedrock properties with different configurations and the other probe measures fluid resistivity and temperature simultaneously.

Geophysical borehole logging was combined with a fluid logging experiment. The fluid logging test was motivated by the fact that the borehole water is very saline, while fresh water was available from a nearby well equipped with a submersible pump. The idea was to replace the saline borehole water with fresh water and then to monitor the intrusion of saline water from the bedrock fractures by fluid resistivity logging.

Galvanic bedrock and borehole water properties were measured in June 2005, and the stability of borehole water conditions was checked in September before fluid logging. The fluid logging experiment started with the pumping of water (14.8 m^3) from the bottom of the borehole; it was replaced by fresh water (total 46.8 m^3) fed on the collar. The resistivity of the pumped water varied, but became steady at the end of the pumping, electrical conductivity (EC) being about 200 mS/m (5 ohm-m), which is still high compared to the EC of the well water fed into the borehole (20 mS/m = 50 ohm-m).

The first resistivity logging, carried out immediately after the water pumping stopped, indicated that a major part of the fresh water pumped into the borehole intruded into bedrock fractures at the depths of about 10, 40, and 60 metres, while the sections 100–130 m and at a depth of about 170 m discharged abundantly saline water into the borehole. After about six weeks the original salinity distribution of the borehole was almost recovered.

The fluid resistivity data during different phases of the fluid logging experiment are summarised in Figure 67a. The bedrock resistivity data (Figure 67b) indicate the strongest anomalies at a depth range of about 130–135 m, but other distinct anomalies can be seen at about 35 m, 60 m, 85–96 m, 100 m, 110 m and, less distinctly, at 170 m. A distinct change in the resistivity level at the depth of about 100 m can evidently be attributed to the change in water salinity, while the reference level of self-potential (SP) seems to remain unaffected.

The distinct correlation between SP and hydraulically active fracture zones is one of the interesting findings of the Kopparnäs borehole geophysical study. However, the general feasibility of borehole self-potential anomalies as indicators of water-bearing fractures remains to be verified by other measurements. As generally known, sources of a self-potential field include electrochemical potential gradients resulting from the redox processes and fluid movements. SP effects caused by fluid movement are also known to possess streaming potential. However, problems that are mainly related to the polarisation of electrodes are considered a major drawback of the applicability of the self-potential method.

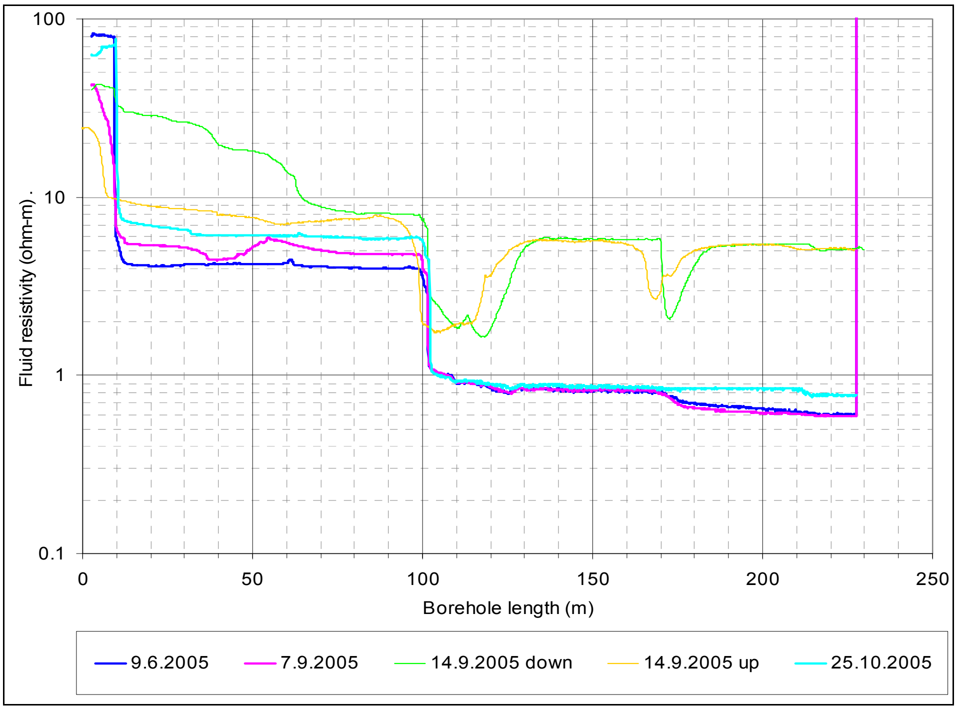

Figure 67a. Fluid resistivity distribution in the Kopparnäs borehole R-307, and resistivity variation after fresh water injection, 14.9.2005 (fluid logging).

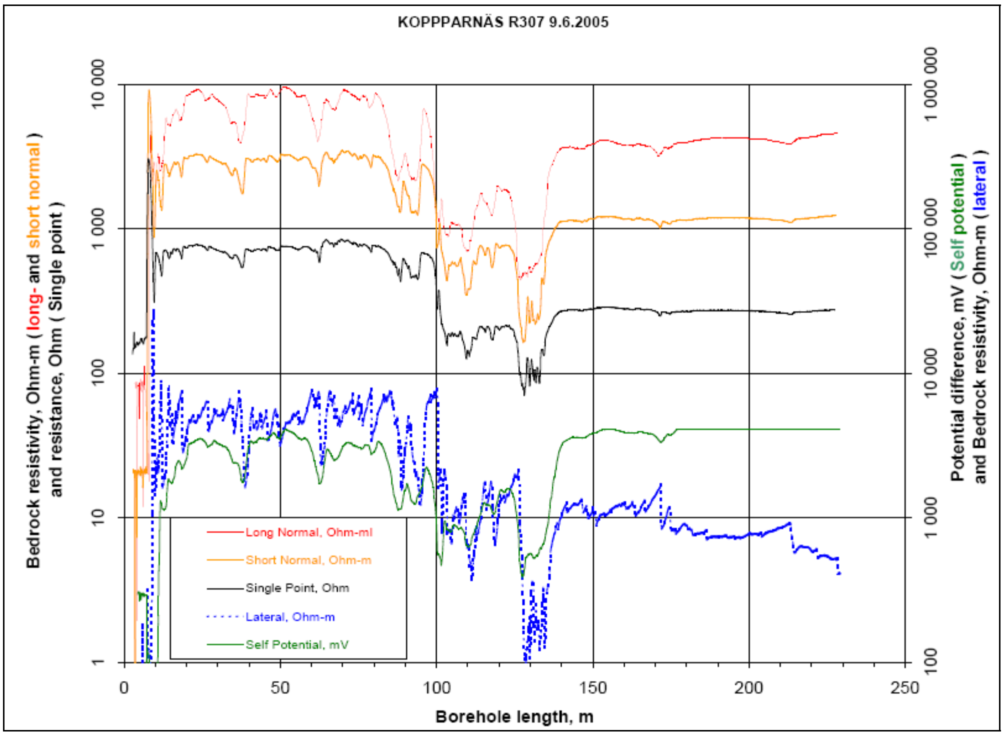

Figure 67b. Bedrock resistivity and self-potential distribution in the borehole R-307.

CONCLUSIONS

The first step in determining possibly water-conducting geological structures is usually based on lineament interpretation. Currently, low-altitude airborne geophysical measurements are available from practically the whole country. The possibility of using different non-interdependent data sets in lineament interpretation is an important advantage. A systematic and transparent methodology for lineament interpretation was applied for the Kopparnäs site, similarly to that used at Olkiluoto by Posiva Oy.

At Kopparnäs, one of the interpreted structures was taken as a target for detailed study, including drilling through the structure, as well as for geophysical and hydrogeological studies of the borehole. In good agreement with the lineament interpretation, a broken water-conducting bedrock section was observed at a depth of about 100 metres. The borehole was surveyed by video imaging and the results were compared with the drill core log. The general observation was that the true open fracturing of the bedrock is clearly less than would normally be inferred from the drill core.

Groundwater sampling of the borehole half a year after the drilling revealed that water below 100 m is saline (TDS about 10 g/L). Saline borehole water is evidently discharging from the main fracture zone at that depth. The salinity corresponds to that of the Litorina sea stage, also observed elsewhere in the Finnish coastal regions (e.g. Hästholmen).

The new borehole proved to be feasible in testing and demonstrating the use of geophysical methods in characterising bedrock hydrogeology. The geophysical borehole logging study was strongly supported by the possibility of replacing the saline borehole water with fresh water and performing a fluid logging test, in which the discharge of saline water from the bedrock into the borehole was monitored. Many hydraulically active zones were discerned in the borehole, and a clear correlation with a different geophysical resistivity log was observed. A very interesting observation was the correlation between the anomalies in the self-potential log and the observed hydraulically active fractures. Additionally, a systematic study of the geophysical borehole logging of the Outokumpu deep borehole was carried out. The results were compared with the results of the available hydraulic tests and, as a major outcome, a methodology for discerning hydraulically active borehole sections on the basis of geophysics was developed.

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2.3.4 Geochemical signals of ice ages in bedrock[17]

INTRODUCTION

The current interest in studying glaciation-induced geochemical traces in bedrock was triggered by the observations of successive U release and accumulation events at Palmottu (Blomqvist et al. 2000; Suksi et al. 2001a, b). Because such behaviour on the part of U generally reflects changes in redox conditions, it was thought that systematic study of the phenomenon could help to answer the following questions:

1) How deep can oxidising glacial melt water penetrate before losing its oxidising power?
2) What routes have melt waters used?
3) Could the behaviour of U be used to conceptualise flow system behaviour?
4) What are the main uncertainties related to interpretation?

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U-series dating has revealed that most of the U accumulations studied were formed during the last glaciation cycle, suggesting respective U mobilisation in the same time scale. The U mobilisation was the result of either current or glacial melt water; in both cases it created U-series disequilibrium signatures that unambiguously characterise the process. This type of information is of prime importance concerning repository safety assessment (SA), because it offers an analogy for possible future changes in similar conditions.

Research studies in the period 2002–2005 were focused on clarifying what kind of relationship, if any, exists between U mobilisation and past hydrogeological changes. Important questions concerning U mobility that were posed were:

(1) Can the impact of current oxidising groundwaters on U be distinguished from that of glacial melt waters?

(2) Can $^{230}Th/^{234}U>1$ in rock be regarded as unequivocal evidence of recent U release?

(3) Can other geochemical indicators be used to indicate U mobilisation conditions? What is the potential of using Fe and Mn oxidation?

(4) Could the duration of oxygen transient be studied in the rock matrix adjacent to the fracture? Can diffusion modelling and mass balance calculations be used for that purpose?

Answers were elicited by: (i) examining data from Palmottu and other study sites in Finland and Sweden; (ii) investigating new samples from Palmottu; (iii) improving U-series modelling capability, and (iv) studying water-rock interaction in the laboratory. A milestone in the research period was the feasibility assessment of the U series disequilibrium method in which both pros and cons were estimated (Rasilainen et al. 2004). The results of the assessment pointed out the key role of the technique.

GEOCHEMICAL SIGNATURES OF U MOBILISATION AND THEIR INTERPRETATION

Oxidising water that penetrates rocks in reducing conditions reacts with the components of the rock-water system as water becomes anoxic. In the rock U is mobilised, U isotopes fractionated, and Fe and Mn oxy-hydroxides formed. The geochemical signatures that may prove the occurrence of the process (i.e. penetration of oxidising water) consist of U-series disequilibria ($^{230}Th/^{234}U>1$ and $^{234}U/^{238}U\neq1$) and the occurrence of Fe and Mn oxy-hydroxides, which can be concluded from Fe_{ox}/Fe_{tot} and Mn_{ox}/Mn_{tot} profiles in the rock matrix adjacent to water-conducting fractures.

Obtaining an unambiguous U-series disequilibrium signature from a U mobilisation event requires careful analysis of the sample material and subsequent data. The most critical stage in the analytical process is sample dissolution, where the disequilibria signature might be distorted. The signature might dilute if the sample contains significant quantities of “old U”, i.e. U in radioactive equilibrium. Fortunately, the possible dilution always changes the time information in the same direction, i.e. to older ages, yielding an upper age limit for the U mobilisation event.

In the case of Fe and Mn oxides, it is worth knowing that both hydrothermal and low-temperature oxides can occur in the sample material and this has to be taken into account in analysis. However, hydrothermal oxides can, in most cases, be excluded on the basis of the shapes of Fe_{ox}/Fe_{tot} and Mn_{ox}/Mn_{tot} profiles, because low-temperature oxidation in reduced rocks is limited close to the fracture surface (see Figure 70).

In the research period 2002–2005 a large body of data on U mobilisation was collected. Different fractures were studied. Fractures exhibited U-series disequilibrium signatures of: (i) recent U accumulation; (ii) recent U release, and (iii) long-term immobilisation of U. The observations were important in giving credibility to our methodology and, independently, illustrated the complexity of the flow system (Suksi et al. 2002, 2003; Suksi 2003).

After the general U mobilisation at Palmottu had been documented (Suksi et al. 2002, 2003; Suksi & Rasilainen 2002), U release from rock was studied in more detail using U-series modelling and mass balance calculations (Rasilainen et al. 2003). For the first time U release was simulated in a glaciation scenario. The purpose was to see if modelling could reproduce the measured $^{230}Th/^{234}U$ activity ratio that unambiguously indicated U release. Mass balance calculations were used to obtain reasonable constraints for the U release scenarios (Figures 68 and 69).

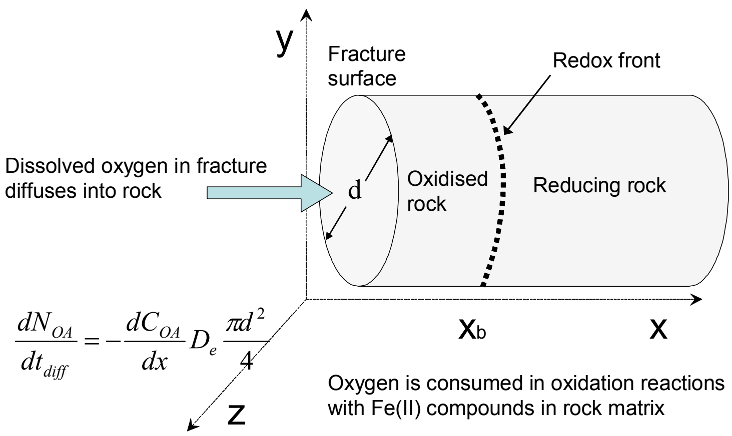

Figure 68. Conceptual model of the advance of a redox front into rock matrix as a result of oxygen diffusion and subsequent release of U. The mass flow of dissolved oxygen is multiplied by the stoichiometric factor of the oxidation reactions and set to be equal to the rate of Fe(II) consumption. The consumption rate of stationary Fe(II) was derived from the propagation rate of the redox front (Rasilainen et al. 2003).

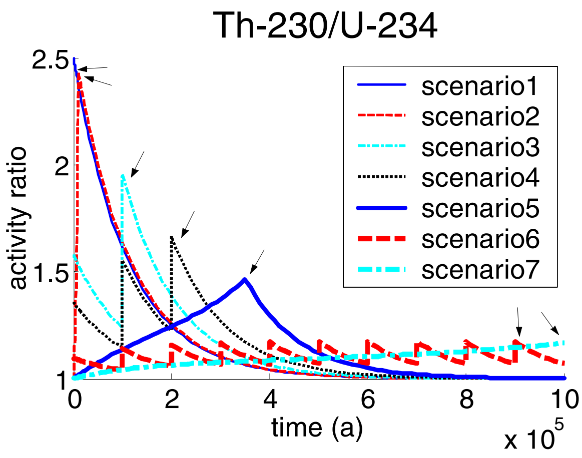

Figure 69. Simulation of Th-230/U-234 activity ratio in different U release scenarios. The response of any release episode is equilibrated within approximately 350 ka. We originally assumed radioactive equilibrium as an initial condition (at t = 0), but in multi-stage release scenarios the first release occurs at t = 0. The small arrows on each curve indicate the postulated sampling point of the respective release scenario (details in Rasilainen et al. 2003).

The result was not surprising; current oxidising groundwater circulation cannot trigger U release from rock centimetres from a water-conducting fracture. Therefore, the only process that could convey sufficient amounts of oxygen into fractures, cause U dissolution centimetres away from the fracture, and last long enough for U out-diffusion was most probably glacial melt water intrusion with a large oxidising capacity. Our first attempt to model the process described above was successful and offered a sound basis for more detailed modelling in the future with more detailed experimental parameters. Studies were enlarged to embrace other redox-sensitive elements. The reasoning for using Fe and Mn is as follows: oxygen diffusion into reduced rock oxidises Fe and Mn. The longer the oxygen transient persists in fractures, the deeper the redox front can advance into the rock. Thus, by studying the oxidation of Fe and Mn as a function of distance from fracture (Fe_{ox}/Fe_{tot} and Mn_{ox}/Mn_{tot}), one might observe the redox front. An example of using Fe and Mn is seen in Figure 70. A similar type of fracture-controlled oxidation was observed in some other drill core samples as well. The preliminary results show that we have the basic tools to study fracture-controlled oxidation (Suksi and Marcos 2005).

Evidence of fracture-controlled natural oxidation helped in the planning of further studies. The possibility of quantifying oxidised Fe and Mn offered a tool to quantify oxygen consumption as well. Because Fe concentration in rocks is normally much higher than Mn concentration, a good estimate of oxygen consumption could be obtained by studying Fe alone. One of the remaining tasks is to model the duration of oxygen transient in fractures, using an oxygen inventory deduced from Fe and Mn oxidation.

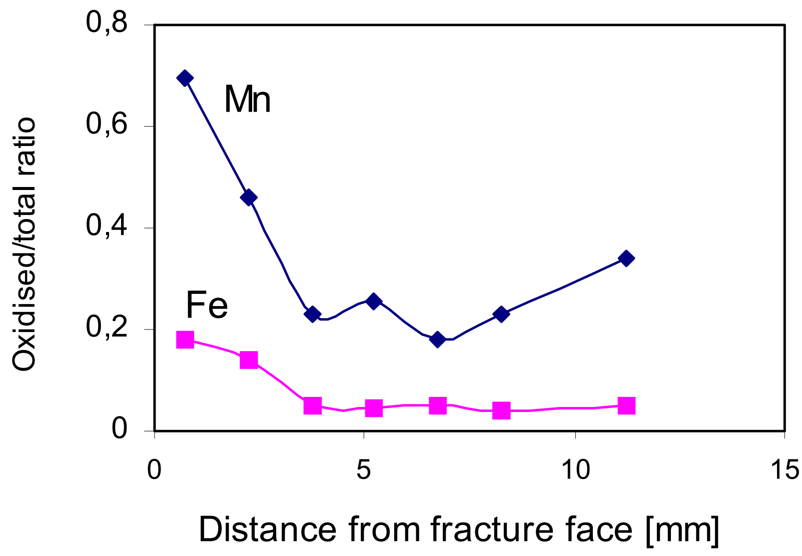

Figure 70. Natural oxidation of Fe and Mn adjacent to a small aperture flow channel interpreted from the proportions of oxidised Fe and Mn (oxidised/total ratio). Oxidised Fe and Mn have been quantified using selective chemical extraction (see details in Suksi and Marcos 2005).

DATING OF U RELEASE

Current oxidising groundwaters and melt waters should in principle yield similar effects on U. In order to separate their influence dating is necessary. Dating of U release is not as straightforward as is dating of U accumulation, which has been traditionally used in Quaternary geology. In principle, U release can be dated provided that the U can be quantified. A suitable case for the dating study was found in a sample where around a flow channel an abrupt decrease in U concentration from 285 ppm to 16 ppm close to the aperture was observed (Figure 71). The released amount of U could be thus obtained from the U concentration profile.

A special feature observed in U release was the exceptionally large $^{230}Th/^{234}U$ activity ratio (12.5), which as such indicates strong and recent U release, giving a very extreme reference for modelling result. The results indicated that U had released rapidly during the postglacial period (Figure 72). The simulations further indicated that U accumulation (and subsequent ^{230}Th generation) previous to its release occurred most probably some 200 ky ago. Modelling was done using the URSE code presented in Rasilainen and Suksi (1997).

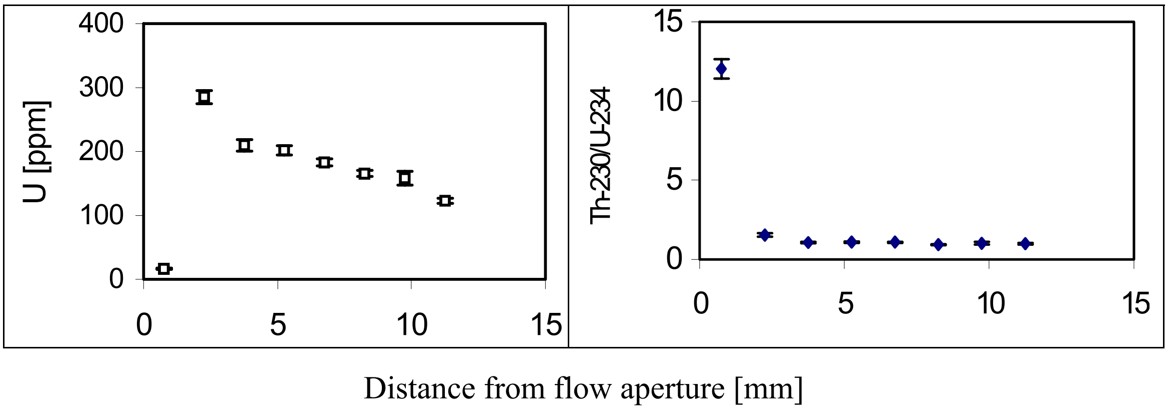

Figure 71. U release from rock around a flow channel as a function of distance from the flow channel aperture (Palmottu DH384/34.5m). U out leach was interpreted from the drop in U concentration and the $^{230}Th/^{234}U$ activity ratio, which showed an exceptionally high value (12.5).

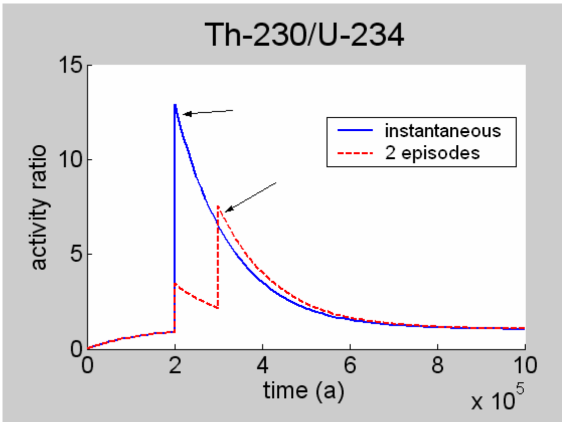

Figure 72. Simulations of U out leach. Simulations were performed using instantaneous and two release episodes with an equal amount of U released in both scenarios. The arrows indicate the postulated sampling points in respective release scenarios. The simulation $^{230}Th/^{234}U$ activity ratio of 12.5 indicates a single rapid U release most probably after a 200 ky accumulation time.

$^{234}U/^{238}U$ ACTIVITY RATIO (AR) IN FLOW SYSTEM CHARACTERISATION

The observation of ^{234}U depletion seen in rock samples close to water conducting fractures (Blomqvist et al. 2000; Marcos et al. 1999) yielded good experimental evidence of U isotope fractionation caused by changes in groundwater conditions. One such example was the measured $^{234}U/^{238}U$ activity ratio value of around 0.25, which is much lower than the theoretical value of 0.5 obtained for direct α-recoil on geometrical basis.

The above finding made us scrutinise the literature of U isotope fractionation. The scrutiny was necessary also because ^{234}U and its parent ^{238}U, more precisely their activity ratio (AR), have long been used as hydrogeological tracers; the first comprehensive review published in 1976 (Osmond & Cowart 1976) and the second one in 1992 (Osmond & Cowart 1992). It is well known that the reason for variations in AR is isotope fractionation at the rock-water interface. Fractionation is known to be due to the more mobile behaviour of ^{234}U, which is generated via chain decay and emerges in the more mobile valence state 6+, promoting preferential ^{234}U release with respect to ^{238}U (Adloff & Rössler, 1991; Ordonez-Regil et al. 1989; Suksi & Rasilainen, 2002). However, the reason why AR varies in groundwaters and to the observed extent is not clear. A study of the problem was started in the KIVES-project by examining first a large body of U data from Finnish and Swedish investigation sites (Figure 73).

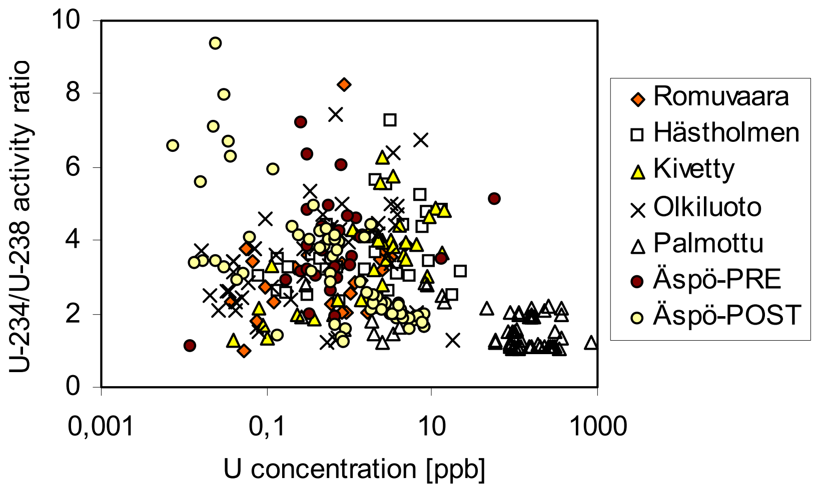

Figure 73. Measured $^{234}U/^{238}U$ activity ratios and U concentrations in some Finnish and Swedish groundwaters. Data from Kaija et al. (1999), Pitkänen et al. (1996, 1998, 1999, 2001, 2004) and Tullborg et al. (2004). Äspö-PRE and Äspö-POST refer to the data previous to and after the construction of Äspö Hard Rock Laboratory, respectively (Rasilainen et al. 2005).

Figure 73 shows how U data are generally presented. As such, no information can be derived on the hydrogeological evolution of any of the sites. However, a link may exist between the hydrogeological evolution at the investigation sites and the observed AR data in groundwater.

For a more detailed interpretation, the U data in Figure 73 were presented as ^{234}U versus ^{238}U using activity unit Bq/l, e.g. Figure 74 shows data from Palmottu and Olkiluoto. In both cases the plots yielded exceptionally good correlations, the slopes giving the $^{234}U/^{238}U$ release ratio (i.e. the activity ratio in which the isotopes were released from rock). An interesting finding when comparing the release ratio values obtained is their large variations between and within the investigation sites. If a link existed between the $^{234}U/^{238}U$ release ratio and the groundwater chemical conditions then the variations in the $^{234}U/^{238}U$ release ratio between the sites might be explained by the hydrogeological past of the sites. A link could be established being the case that $^{234}U/^{238}U$ release ratio is 1 in oxidizing conditions at Palmottu (that is, isotopes have not fractionated during release) whereas the release ratio is 6.8 at Olkiluoto, where conditions change from oxidising to anoxic. A more detailed study on the subject will be published in Physics and Chemistry of the Earth (Suksi et al. 2006).

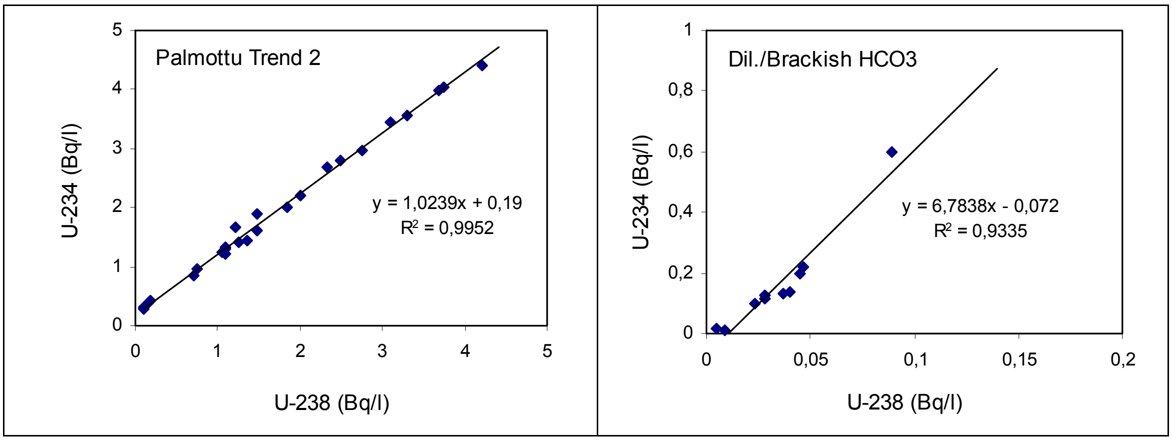

Figure 74. Examples of regression analysis of Palmottu (left) and Olkiluoto data (right). The slope gives directly the $^{234}U/^{238}U$ release ratio. Palmottu Trend 2 data are from oxidising groundwater. Dilute brackish HCO_3 data represent groundwater that rapidly changed from oxic to anoxic (Pitkänen et al. 2004).

It is known that the rock types and thus also U sources vary between sampling locations, which means that the correlations are mainly due to groundwater conditions. The consistent behaviour of ^{234}U versus ^{238}U under the same conditions gives us an interesting possibility to use the correlation to identify data points that may represent different flow paths because the "differing" data points are expected to deviate from the correlation. This idea can be tested because outlier data points can be substantiated by independent hydrogeochemical data. Testing is underway.

A crucial step forward in interpretation was made after finding that physical ^{234}U flux is in most cases insignificant compared to the respective chemical flux. This was observed in a direct α-recoil simulation exercise (Rasilainen et al. 2005). Thus the possibility to exclude direct α-recoil flux from the total ^{234}U flux significantly simplified interpretation. A simulation example of physical ^{234}U flux into water is given in Figure 75.

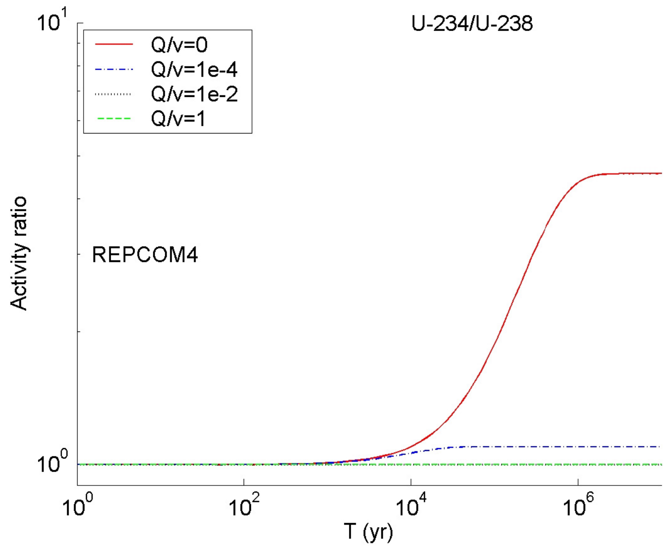

Figure 75. A simulation example of direct α-recoil for $^{234}U/^{238}U$ in groundwater flowing in a fracture with aperture of $1 \cdot 10^{-5}$ m. U concentration in groundwater is initially 10 ppb and at the fracture surface 10 ppm. Water turnover (Q/v) has been varied using values 0 (stagnant), $1 \cdot 10^{-4}$, $1 \cdot 10^{-2}$, and 1 (1/a) (from Rasilainen et al. 2005).

The result from data analyses and from recoil simulations supported the overall hypothesis that groundwater chemical conditions control the $^{234}U/^{238}U$ release ratio and thus AR in groundwater. The hypothesis can be tested in the laboratory and such testing is underway.

DISEQUILIBRIUM SIGNATURES PRODUCED BY UNDERGROUND EXCAVATIONS

Natural decay series nuclides can be used to establish relevant time correlation for man-made changes. We studied fracture surface samples from the Pyhäsalmi mine to see if possible disequilibrium signatures from excavations can be seen (Marcos et al. 2004; Marcos & Suksi 2005). We studied the ^{232}Th decay chain sequence $^{232}Th \rightarrow {}^{228}Ra \rightarrow {}^{228}Ac \rightarrow {}^{228}Th$ which reaches equilibrium within 40 years. If excavations have affected the flow system, ^{228}Ra may have been mobilised and transported down flow generating locally extra ^{228}Th which imprints fracture surfaces as the $^{228}Th/^{232}Th$ activity ratio >1. Th has been assumed immobile, as is the normal practice in natural waters.

The study of the Pyhäsalmi samples proposed a methodology based on natural decay series studies to monitor natural and excavation-induced disturbances. The observed $^{228}Th/^{232}Th \gg 1$ unequivocally indicated changes within the last 30 years. More detailed distinguishing between excavation-induced and natural disturbances would require systematic and detailed studies at the selected site(s).

Reactivation of the flow system due to excavations can mobilise U and thus affect U isotope composition in groundwater. The effect can be studied provided that data before and after excavations are available. Such a case study was provided by the data from the Äspö Hard Rock laboratory. The data are presented in Figure 76. Comparison of the data shows that the data from before the excavations (PRE-HRL data) scatter "broadly" around one line whereas the data from after the excavations (POST-HRL data) appear tightly grouped into two alignments with different $^{234}U/^{238}U$ release ratios (Trend 1 and Trend 2). The observation is noteworthy because it may indicate U mobilisation in different conditions and separate flow systems. More detailed interpretation of the observation will proceed in the future by consultation with the data providers.

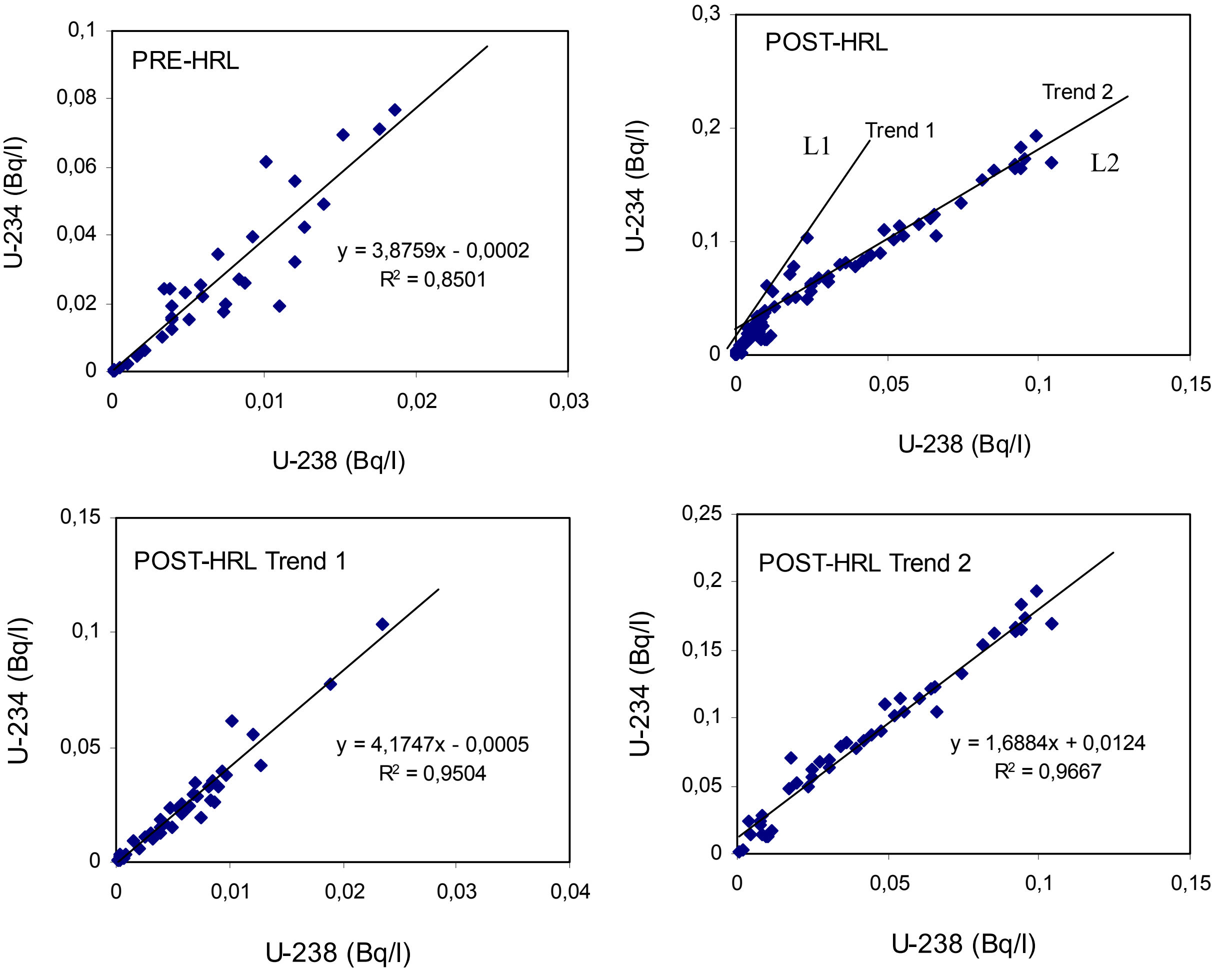

Figure 76. Examination of the Äspö data (Smellie et al. 1995; Tullborg et al. 2004) before and after the construction of Äspö Hard Rock Laboratory. Note the aligning of data points after the construction process, which yielded different slopes (Rasilainen et al. 2005).

Both, the study of the Pyhäsalmi samples and the analysis of the Äspö data show that we have caught the signatures of man-made disturbances around the excavations and that we have the basic tools to study the range of such disturbances.

CONCLUSIONS

Methodology for the interpretation of geochemical signals in bedrock has been developed in order to link the signals to U mobilisation events. Two signals were studied in particular: U-series disequilibria (USD) in rock and water and the occurrence of Fe and Mn oxy-hydroxides in rock.

A link has been established between USD data and redox front in rock matrix surrounding a water-conducting fracture. The work was based on parallel use of experimental USD-observations, USD-modelling, diffusion modelling and mass balance considerations. The conceptual model was that oxic water penetrates the surrounding rock matrix and oxidises it to the extent controlled by the duration of the oxic pulse, diffusion kinetics, and the abundance of oxygen consuming compounds in the rock matrix. The existence of a redox front can further be supported by the occurrence of Fe and Mn oxy-hydroxides as these will be formed in the oxidised part of the rock matrix.

The dominant role of geochemical conditions in controlling radioactive disequilibria in groundwater was established when the role of direct alpha recoil was shown to be insignificant in strongly flowing systems. The role of alpha recoil was studied quantitatively in numerous sensitivity studies varying for instance U source strength, fracture geometry, and groundwater turnover.

Based on the observation above a new methodology was outlined to interpret $^{234}U/^{238}U$ activity ratios in groundwater. The methodology, based on presenting ^{234}U vs. ^{238}U graphically, is assumed to yield the ratio in which the isotopes were released from rock. This in turn would provide a tool to estimate the geochemical conditions in investigation sites. The methodology was tested with an extensive experimental material from Finland and Sweden and the measured activities of the U isotopes grouped surprisingly well along trend lines considered characterising the sites. The methodology was also tested for its ability to show signs of underground excavations, e.g. mines or underground research facilities, and the results were encouraging.

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2.3.5 Gas studies in the Olkiluoto sea area[18]

INTRODUCTION

The gas study was made to examine gas or groundwater anomalies found recently (Kotilainen & Hutri 2004) in Holocene submarine sediments in the Olkiluoto area, in the Gulf of Bothnia, which is the selected site for the Finnish nuclear waste repository. The reflections of the anomalies in echo-sounding profiles recorded from the area, a “mushroom” or “cloudy”-looking appearance or rounded depressions (pockmarks), imply that discharge is still constant in some places.

Some gas anomalies lie only in the Holocene soft sediment but some anomalies seem to derive from deeper in the sea bottom. The possible cause of this phenomenon might be gas derived from the organic-bearing Litorina and recent gyttja clays, or gas which is derived from the bedrock, the so-called deep gas. Several observations around the world (e.g. in the Stockholm archipelago, Söderberg & Flodén 1992) connect deep gas seepages (especially methane) to fault creep and (micro) seismicity, which has relevance to the safety assessment of a spent nuclear fuel repository. Although the seismic activity in the area has been very sparse, indications of bedrock creep have been measured with an accurate GPS net on Olkiluoto island (Ollikainen & Kakkuri, 2000).

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This study aimed to reveal the origin of the observed gas/fluid release through sampling and chemical analyses and to investigate whether there is a connection between the gas observations and bedrock fault creep.

SUMMARY OF THE CONDUCTED STUDIES

The previously recorded study area (together ~350 km survey lines, situated approximately 500 m from each other), about 100 km^2 (Figure 77), was revisited for 45 new acoustic-seismic survey kilometres. The Geological Survey of Finland (GTK) uses a MD DSS sonar system© incorporating both software and hardware. For the present study, a 28-kHz echo-sounder was used. This frequency of echo-sounding provides resolutions as high as ~10 cm (in favourable weather conditions, which were met during surveying). In addition to echo-sounding, we used single-channel seismic reflection (Electro-Magnetic implosion-type sound source, ELMA, 400–700 Hz, depth resolution of ± 2 m) and side scan sonar (Klein SA 350, 100 kHz) surveys. The geospatial position was based on the DGPS (Differential Global Positioning System) system with ± 2 m accuracy. During the surveys both the acoustic echoes and the 3D positioning of each sounding observation were stored digitally. The interpretation of the profiles was carried out on a PC screen, using TOPOS mapping software.

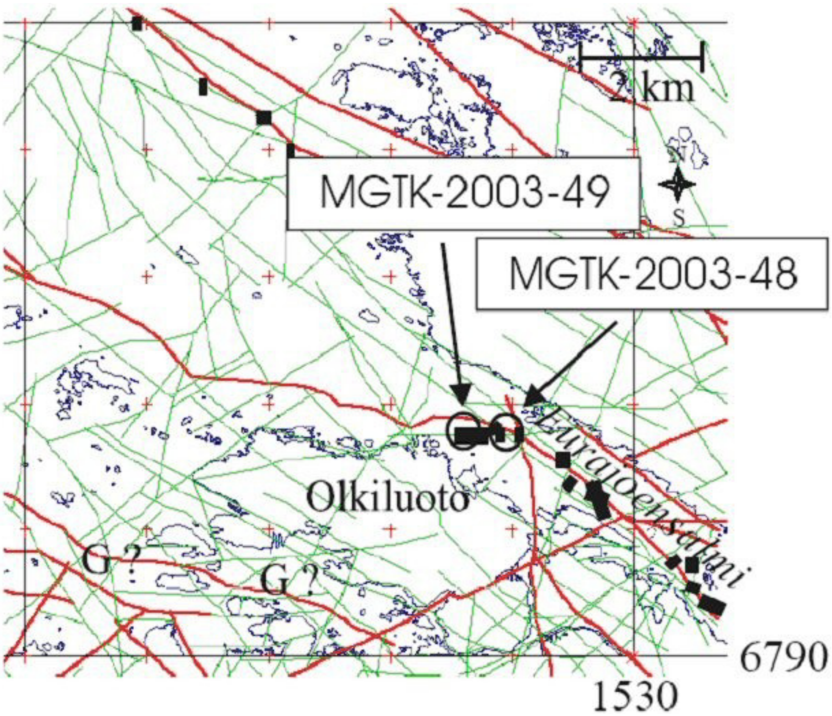

Figure 77. Study area and methane sampling sites (Hämäläinen & Kotilainen 2004).

In the echo-sounding and side-scan profiles several sediment structures, like pockmarks (Figure 78), and mud mounds associated with gas/groundwater seepages were observed.

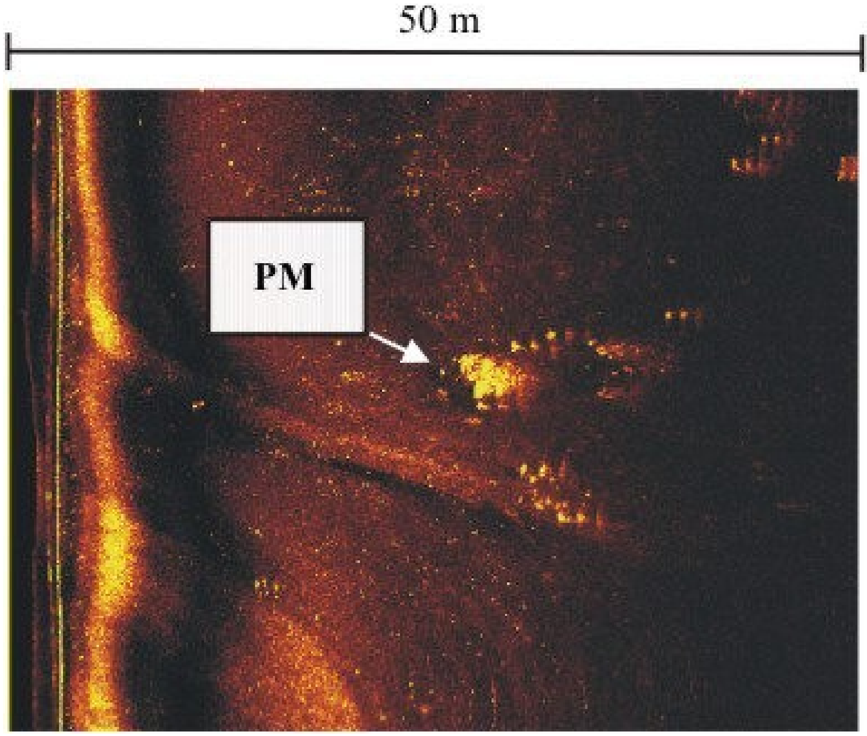

Figure 78. A side-scan profile of a pockmark (Kotilainen 2002).

Because of acoustic-seismic anomalies, the sampling was directed to six sites where gas-rich sediments occur. The main part of the sampling was carried out in the summer of 2002 with a "Söderberg"-type sampler (Figure 79), which was developed at Stockholm University. Two types of samples were taken: gas samples direct from the sampler chamber equipped with ventilators, and sediment samples from the same sampler, from which additional gas samples were taken (Figure 80). The sampling method and the laboratory results are reported in Kotilainen (2002). The aim of that study was to confirm the occurrence of methane in the submarine sediments. The methane analyses were performed by gas chromatography at the Lahti research laboratory and they confirmed the existence of methane in the sediments. Methane concentrations in the chamber samples were relatively low, at 0.007–1.2 mg/l. In the sediment samples the methane concentrations were higher, at 0.11–18.7 mg/l (Kotilainen 2002).

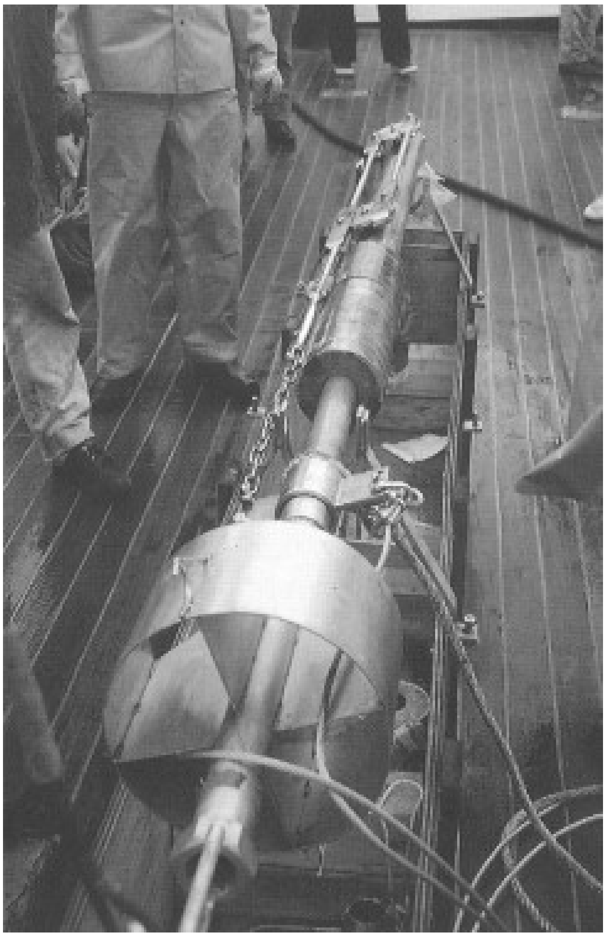

Figure 79. "Söderberg"-type sampler (Photo Per Söderberg).

Figure 80. Additional gas samples taken from a sediment core (Photo Jyrki Hämäläinen).

Since the methane concentrations themselves did not reveal information on the origin of the gas, a new expedition to the study area was planned. The isotope abundances of carbon and hydrogen may indicate the origin of the gas. It has to be remembered, though, that the isotope ratios can also be modified by subsequent processes, e.g. CH_4-oxidising bacteria can change isotope composition.

Additional sampling was carried out during the 2003 field season, when two carefully selected sites (Figure 77) were recovered using a vibrohammer corer. Carbon isotopic ratios of $^{13}C/^{12}C$ from the sediment samples MGTK-2003-48 and MGTK-2003-49 from both sites were measured on an isotope ratio mass spectrometer (IRMS; Thermofinnigan, Bremen, Germany) at the Dating Laboratory of the Finnish Museum of Natural History, University of Helsinki. The methane gas was oxidised to carbon dioxide (at 1000ºC) in a PreCon device (Thermofinnigan, Bremen, Germany) and introduced on-line to the IRMS.). The results, as follows, are given as $\delta^{13}C$ values against VPDB. $\delta^{13}C_{CH4}$ measurements of the sediment core MGTK-2003-48, -80‰, indicate the biogenic origin of methane. $\delta^{13}C_{CH4}$ values from the sediment core MGTK-2003-49, -46‰ and -50‰, are close to the atmospheric $\delta^{13}C$ values (-47‰), and thus it is presumable that the samples were contaminated and the results are relatively unreliable (Hämäläinen & Kotilainen 2004).

A new gas sampling technique was tested in the winter of 2004 (Figure 81) through the ice cover from the same area, with a self-developed sampler (Figure 82). The method used was, however, not successful for collecting gas samples from submarine sediments. This methodological development work was done outside KYT funding.

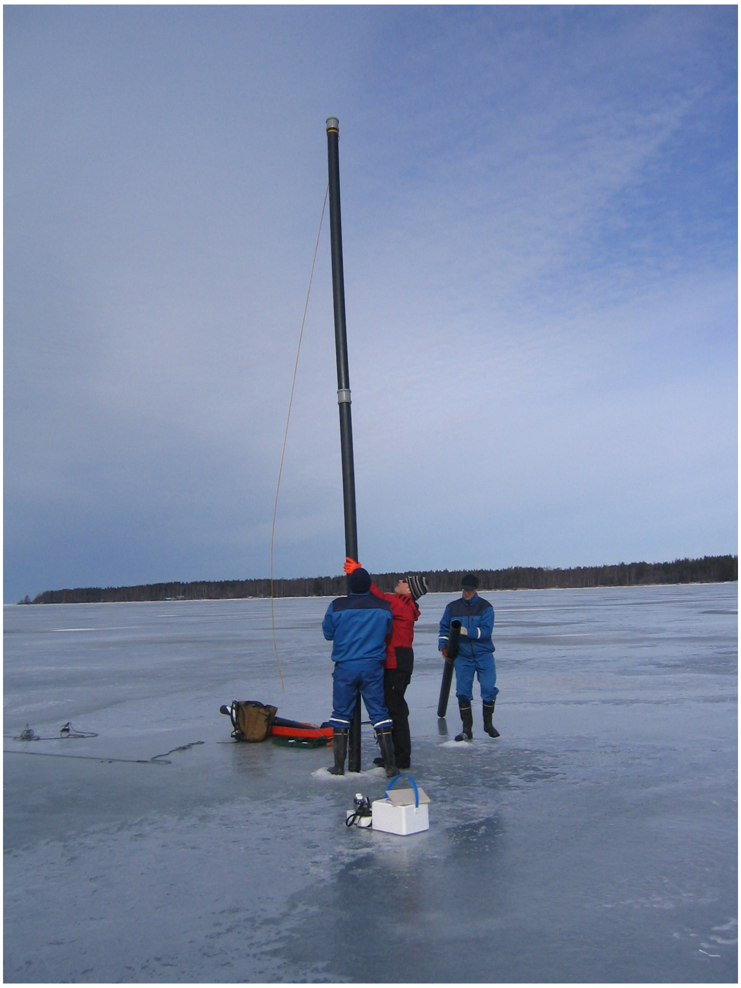

Figure 81. Winter sampling on the Olkiluoto ice (Photo Kaisa-Leena Hutri).

Figure 82. Sampling tube (Photo Kaisa-Leena Hutri).

Since the results actually did not add anything remarkable to the previous studies of the authors, the results have been reported so far only in GSF's work reports (Kotilainen 2002; Hämäläinen & Kotilainen 2004).

CONCLUSIONS

On the basis of the laboratory analyses performed so far, the origin of the gases and their connection to seismic creep cannot be proven. However, the reflections of the seepages in the echo-sounding profiles imply that part of the gas is derived from the bedrock, thus being so-called deep gas, and the formation of the pockmarks seems to be related to fracture zones. Additionally, the groundwater samples from the deep drill holes in the island of Olkiluoto include a high volume of gas (methane) (Karttunen et al. 1999; Haveman et al. 2000), which supports the bedrock origin. One possibility is that some of the observations are relicts (buried pockmarks) of strong hydraulic gradients connected with the Weichselian deglaciation (Hutri et al. 2005).

Tectonic movements and high hydraulic gradients are recognised in safety analysis as major threats to repository safety related to glacial scenarios (Vieno & Nordman 1999). To get better and more reliable results, further research should be conducted with successful sampling techniques and broader analyses. This is, however, expensive and time-consuming. The authors did not submit a new application for KYT funding, since the principal researcher was nominated to the Steering Group of KYT. However, the research has been going on outside KYT, focusing on similar observations around the Baltic proper and the Bothnian Sea. These results were presented at the 4th Geological Research Meeting in Turku (Hutri et al. 2005) and as a scientific paper (Hutri & Kotilainen 2005).

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2.3.6 Permafrost studies at the Lupin Mine[19]

INTRODUCTION

In assessing the safety of the geologic disposal of spent fuel, it is essential to assess what physical and chemical conditions could evolve within the repository and surrounding environment. The objective of performance and safety assessments is to demonstrate the long-term safety of a repository for long-lived radioactive waste and most management agencies consider scenarios of up to 1 million years (My) into the future.

During at least the past 2 My, repeated cycles of glaciation have occurred in the Northern Hemisphere. At present, a major part of the area to the north of latitude 60°N is under permafrost conditions, although temperate conditions prevail within

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Fennoscandia at the same latitudes. Climate models based on astronomical forcing predict a cold period after approximately 60,000 years. Cold, dry periods favour the formation of periglacial conditions and the development of deep permafrost is expected to reoccur in northern and central Europe and North America.

In order to evaluate the long-term performance of a geological repository, the influence of periglacial and glacial conditions must be considered. A key goal of this research project is to enhance the scientific basis for safety and performance assessment and to derive constrained permafrost scenarios relevant to the evolution of crystalline groundwater flow systems. On the basis of an earlier literature study and theoretical considerations (Ahonen 2001), a number of issues were identified as possible targets of study: 1) freezing rate and depth extent of permafrost in crystalline bedrock; 2) cold saline water segregations (cryopegs) and their role in transport processes; 3) the role of non-frozen areas (taliks) as pathways for groundwater flow; 4) the aggregation of solid methane hydrates (clathrates), and 5) the effects of freezing on bedrock stability.

All these aspects are related to the long-term stability of hydrogeological and hydrogeochemical conditions. Some of these phenomena are reviewed at length in the literature and much is known about their role in shallow systems or in sedimentary environments. However, much less has been published regarding cryogenic processes and hydrogeology in crystalline rock under deep permafrost conditions. To be able to address the relevant questions with respect to the geologic disposal concept, the Permafrost Project was initiated in 2001. The fundamental idea was to investigate permafrost at a representative field site in order to evaluate the various hypotheses presented and reviewed in the literature. The project is jointly funded by the Geological Survey of Finland, Posiva, Svensk kärnbränslehatering AB (SKB), Nirex Ltd (UK), Ontario Power Generation (Canada), and the University of Waterloo (Canada). During the years 2002 and 2003 the project was administratively related to the Finnish Research Programme on Nuclear Waste Management (KYT). Since 2003 the project has continued its work as an independent research project. In this document the essence of the work carried out before the year 2004 (Phases I and II) is reported.

THE RESEARCH SITE: LUPIN MINE

The Lupin Mine is located in the Nunavut Territory (eastern part of the former Northwest Territory), about 1300 km north of Edmonton, Canada, and some 80 km south of the Arctic Circle. The distance to the White Sea in the north is about 300 km. The gold mine was opened in 1982 and closed in January 2005. All the mining activities at Lupin took place underground. Access to the mine was facilitated by a main shaft which extends down to the 1210-m level. The final depth of the mine reached almost 1500 m.

The site is within the continuous permafrost zone (Figure 83). The southern limit of the continuous permafrost corresponds roughly with a mean annual air temperature of -6° to -8°C (it is -11°C at Lupin), with its southern discontinuous limit at about -1°C. The region is located in the continental sub-arctic climate zone above the timberline. The vegetation is typical for tundra: grass and low-growing shrub species and dwarf birch. The annual mean precipitation is low, about 270 mm. The major part of this is obtained between July and September; from mid-September until mid-May as snow. The snow cover is thin, but with strong wind, thick drifts can form in suitable spots.

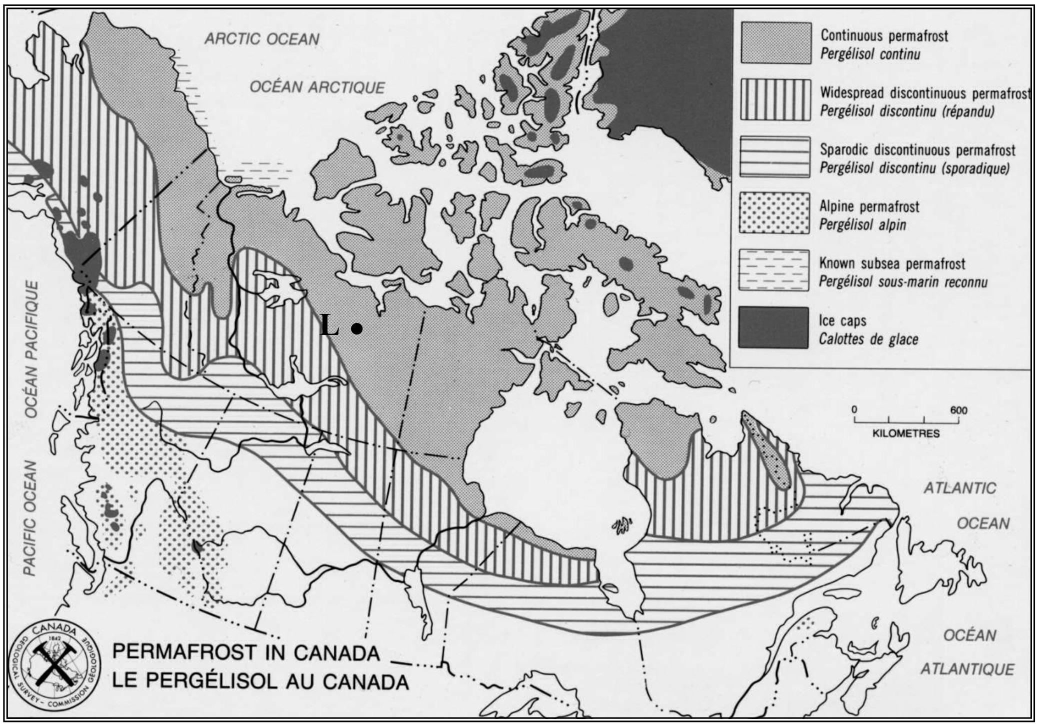

Figure 83. Distribution of permafrost within the territory of Canada. Lupin (L) is located in the zone of continuous permafrost.

Today the depth of the permafrost in the area extends to about 400–500 m. Most of the deep permafrost in Canada was formed during the Holocene, *i.e.* during less than the past 10,000 years. The temperature measurements conducted in the mine indicate that permafrost persisted down to the 541-m level, where 0°C was recorded (Figure 84). The lowest value was recorded at a depth of 87 m (Sandhu & Tansey 1996). The uppermost measurement was from the 27-m level, with a value of -3°C. However, this temperature is rather high compared to the local mean annual air temperature (-11°C), and it is probably disturbed by the mining activities. The depth of the active layer (unfrozen) developed in the overburden during the short summer depends on the soil type, but is less than 2 m.

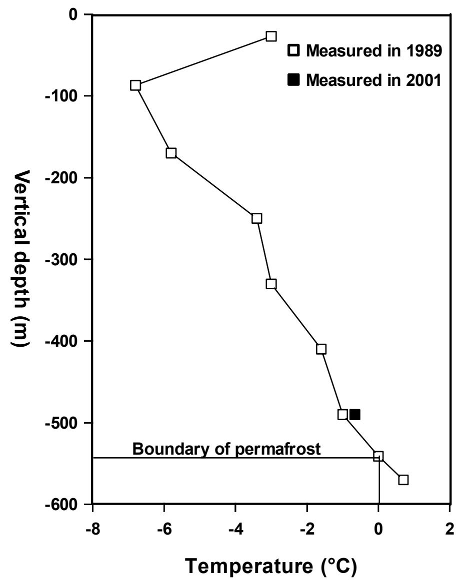

Figure 84. Temperature versus depth, Lupin Mine. The measurements were taken in the main shaft in 1989 (open squares) and at the 490-m level in 2001 (black squares).

RESEARCH ACTIVITIES

During the studies in Phase I of the Permafrost project (Ruskeeniemi et al. 2002), a moderate understanding was achieved of the general geological, structural, Quaternary geological, and permafrost features of the Lupin Mine area. The target of the fieldwork was to collect geological and structural information from the site in order to locate potential water-conducting fracture zones for future hydrogeochemical sampling. A seismic survey at the surface and review of the various sources of information were performed. An important task was the acquisition of the necessary databases from the mining company to construct a 3D model of the mine. Video surveys were performed in more than 10 water-producing boreholes to gain information on the water-conducting fracture zones. Their location and orientation was of major interest. Preliminary characterisation of the deep groundwaters and the dissolved gas phase was also carried out.

However, very little was learned about the natural groundwaters within or just below the permafrost or at median depths. All the samples collected from the upper part of the mine seem to be more or less contaminated by the mining activities. It was considered important to identify and characterise the permafrost water, an end-member water type, at Lupin (and potentially in Fennoscandia). Therefore, the temporal priority in Phase II was given to activities aiming to sample water or activities providing support for a better understanding of the distribution of groundwater and permafrost (Ruskeeniemi et al. 2004). Electromagnetic SAMPO soundings were carried out to observe the depth of permafrost in the surroundings of the mine and to locate possible intensely fractured hydraulically active zones (Paananen & Ruskeeniemi 2003). Groundwater and gas compositions in the research boreholes were monitored and a comprehensive chemistry and isotope database was continuously updated to facilitate geochemical characterisation, geochemical modelling, and dating. In parallel to the field work at Lupin, freezing experiments were conducted at the University of Waterloo on assignment from Ontario Power Generation.

RESULTS

The existence of structural heterogeneity at the Lupin site is evidenced by: (i) salinity (2.2 to 36.3 g/L) and gas differences in the 1130-m boreholes, which follow a general trend in the same direction through the same group of faults and geological units across the foliation. As an example, the proximity of highly saline zones on the 1130-m level to a zone containing very fresh, non-tritiated water suggests variable, poorly-connected structural pathways at the site; (ii) the heterogeneity of the hydraulic heads at the site; (iii) the apparent lack of hydraulic connection between boreholes during hydraulic testing, and (iv) the observed heterogeneity in gas flow from fractures versus fluid flow during down-hole video surveys.

One of the major implications of such a poorly connected system would be the *limitation on recharge* to depth or across the site. This is apparent from the large unsaturated zone (≈ 100 m thick) beneath the permafrost. The results of the electromagnetic soundings suggest that the groundwater table is consistently deeper than the assumed base of the permafrost for a distance of 2–3 kilometres away from the mine. In addition, the inflow of natural water into the mine is low, reflecting the low bulk permeability of the fractured rocks at this site (only about 53,000 m^3 of water annually is pumped out of the mine). The possibility that crustal rebound also affects the unsaturated zone needs further investigation. In addition, the heterogeneity in salinity and the observed head measurements (with the possible exception of the non-tritiated fresh water sampled at the 1130-m level), would suggest that the connections to the assumed large talik structure (unfrozen bedrock) connected to a nearby major lake are poor.

Talik is an unfrozen section of ground within permafrost. Three different situations can be distinguished: open talik (open to the ground surface, but otherwise surrounded by permafrost), closed talik (all within permafrost) and through talik (open to the ground surface and to an area of unfrozen ground beneath it. Permafrost encases it along the sides). In the Lupin area all shallow lakes will freeze down to the bottom during winter. The bottoms of larger lakes deeper than 2–3 m will remain unfrozen all year. These lakes have the potential to support an open talik. Still larger lakes are likely to conceal taliks extending through the deep permafrost. Their impact on the hydrogeological regime depends on the geological setting beneath. The taliks within an intact bedrock would be hydrogeologically isolated “worm-holes”. However, some chains of lakes related to regional lineaments may have partly interconnected unfrozen channels beneath. The most significant talik structure in the Lupin area would be related to Lake Contwoyto, which is only 1.5 km N of the mine. The lake is 1–3 km wide and more than 100 km long. Although there is no direct evidence of the existence of a talik, there is no reason to doubt the existence of one. In principle, taliks provide routes for vertical flow, if a gradient exists.

Highly saline waters under high pressures have not been observed in or directly under the permafrost. The lack of such a large reservoir of high-pressure, gas-rich saline fluid, coupled with the structural heterogeneity discussed above, constrains our conceptual model of the site. There are mine-induced contaminants moving slowly downwards in one large fault system. However, there appears to be a mixing or dilution of this saline, nitrate-rich contamination by a matrix fluid (i.e. stagnant pore water), as seen in the results for a variety of stable isotopes (^{2}H, ^{18}O, ^{37}Cl) used to fingerprint the water and solutes. In addition, anomalies in some parameters, such as sulphate concentrations in fluids from the permafrost zone, may prove to be precipitates from the freeze-out process (e.g., $Na_2SO_4 \cdot 10H_2O$; $MgSO_4 \cdot 6H_2O$). If this proves to be the case, then these very soluble precipitates have remained undisturbed (possibly in a stagnant fracture system) until the man-made brine was introduced into the fault zone. This would have major implications in terms of demonstrating the lack of flow or active hydraulic systems during the natural evolution of the flow system.

Gases found in the deep groundwater systems below the permafrost are apparently thermogenic in origin. The site does have considerable metasedimentary rocks, which were originally deep ocean sediments. The composition and age of the gas would therefore most likely be tied to the age of the metamorphic event (i.e. hundreds of millions to billions of years). In most cases, it is assumed that by association, the gases, matrix fluids, and much of the saline groundwater have similar evolutionary histories.

CONCLUSIONS

One of the key topics of the Permafrost project is to explore whether the out-freezing process can result in the formation of a saline groundwater layer at the base of the permafrost in crystalline bedrock. Highly saline waters might have undesired effects on the function of the bentonite sealing and might increase the corrosion of the other engineered barriers. The available data do not provide any evidence of highly saline waters below the permafrost. However, there is not yet enough information to unequivocally rule out the hypothesis of the presence of a saline freeze-out front. Zang and Frape (2003) conducted a series of laboratory experiments in which the effects of the freezing-out process were studied using a number of groundwater samples from Canada and Fennoscandia. Among other things, they concluded that it is very difficult to envision a situation where any significant amount of concentrated saline fluid could be generated in crystalline bedrock by this mechanism. The amount of water in fractured crystalline rock is limited and the bedrock temperatures necessary to support the successive freezing corresponding to increasing salinity levels are unrealistically low.

It seems that deep permafrost prevents any significant recharge of surface waters. Even in the disturbed mining environment, where a strong artificial hydraulic gradient exists (mine drainage) and a through talik is located next to the mine, the infiltration of recent waters is insignificant. This is, of course, partly due to the poor hydraulic connections, which prevent or slow down the potential flow.

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2.3.7 Post-glacial bedrock movements[20]

INTRODUCTION

One of the starting points of the Finnish site selection procedure was the long-term stability of the bedrock, which is due to its mosaic-like block structure. According to present knowledge, stress in the bedrock is released by the reactivation of old fractures and fracture zones, while the bedrock in between will stay intact. The bedrock model can be verified by analysing the spatial distribution of the younger faulting and by following present-day bedrock movements. This is why evidence of postglacial faulting and present-day bedrock movements has been the subject of intensive study in co-operation with a number of research institutes in Finland and abroad.

Postglacial and currently occurring bedrock deformation can be divided into episodic and continuous. Episodic deformation happens in the form of measurable seismic events, which may be related to certain faults and are mostly brittle. Continuous deformation needs a longer observation period and is detected by accurate geodetic methods such as traditional levelling and modern GPS measurements. It includes elastic, plastic, and brittle deformation, which occur in such small events that they cannot be separated with currently used instrumentation (Ojala et al. 2004).

The purpose of the work was to summarise the current knowledge of the postglacial deformation of bedrock in Finland. These are divided into recent earthquake activity, postglacial faulting, glacial isostatic rebound, and current horizontal crustal deformation. There are exhaustive and detailed case studies on postglacial faults or other deformational features and structures which have resulted from bedrock deformation after glaciations and cut glacial or later deposits in Finland and Fennoscandia (e.g. Kuivamäki et al. 1998; Kuivamäki & Vuorela 2002, Olesen et al. 2004). In addition, tectonics and deformation related to glaciations are discussed in several papers in a special issue of the Quaternary Science Reviews Volume 19, Issues 14–15 (2000).

SEISMIC ACTIVITY

Earthquakes are the most obvious manifestation of the currently occurring bedrock deformation. In Fennoscandia, seismic activity is low and is concentrated offshore near the coastline of Southern and Middle Norway and a few onshore regions (Figure 85). Onshore there is lower activity and magnitudes, especially in Finland, where the magnitude of all earthquakes in human records is less than five. However, the large postglacial fault would

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have had magnitudes up to eight (Kuivamäki et al. 1998). The most active earthquake areas are Southern Sweden, the Swedish coast of the Gulf of Bothnia, the Northern Fennoscandian postglacial fault province, and Kuusamo. Note that Kuusamo and Southern Sweden are the only onshore areas where postglacial faults have not yet been recognised. In Finland, within the modern observation period, it has not been possible so far unequivocally to point out individual faults which have moved during earthquakes.

Several studies demonstrated that the brittle crust is near the point of failure, and that very small changes (0.1 Mpa) in the state of stress can trigger earthquakes. Consequently, the stress build-up resulting from the deformation of the brittle crust is released mainly through microseismicity below the detection limit of the sparse seismic network and large earthquakes are rare. In addition, most of the earthquakes happen at a depth of 5 to 20 kilometres, without any surface trace. Faults may eventually break through to the bedrock surface and disturb glacial deposits or features.

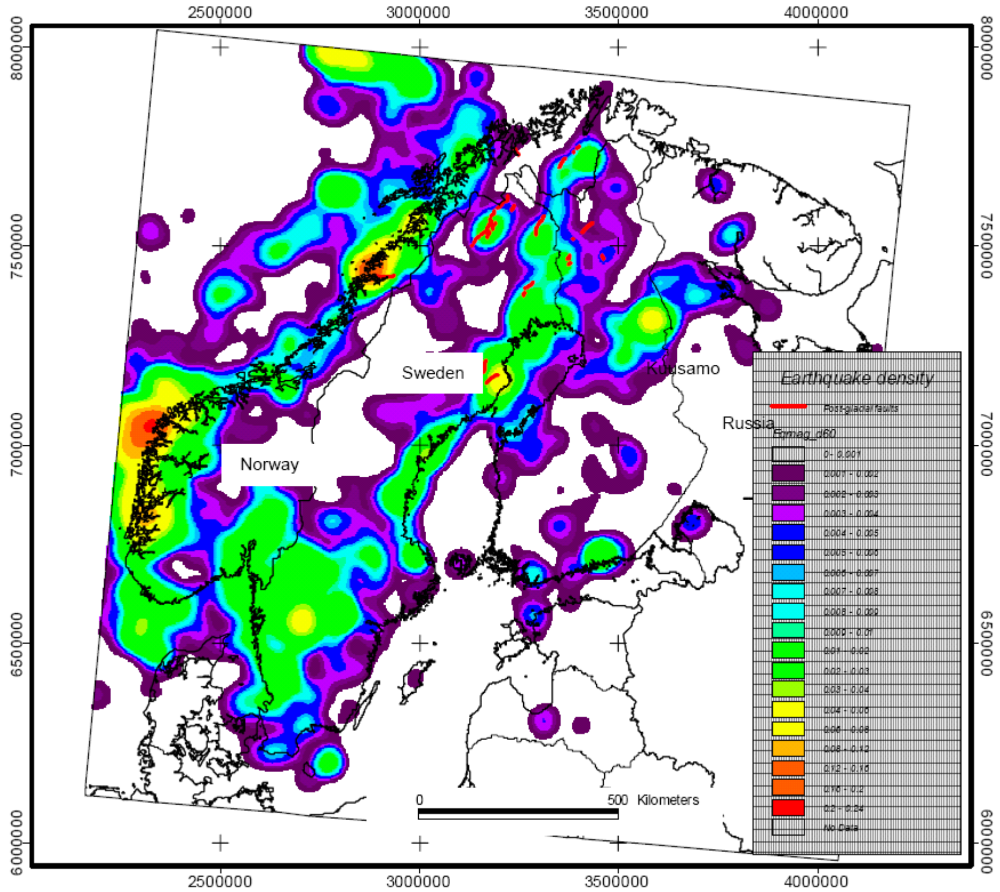

Figure 85. Earthquake density and main post-glacial faults in Fennoscandia. All earthquakes from the year 1375 to 2003 from the Bulletins of Earthquakes of the Department of Seismology, University of Helsinki. Earthquake density has been calculated using a 60-km search radius. Post-glacial faults from Kuivamäki et al. (1998).

POSTGLACIAL FAULTS

The bedrock has been deformed several times during its geological history and it has resulted in a mosaic-like block structure where fracture zones surround more or less intact blocks of different sizes. The old block structure existing now is very stable, i.e. the sizes of blocks and the fracture zones surrounding these are at optimum orientation for any tectonic stress direction to reactivate them with very small loads and to release tectonic stresses.

Large-scale postglacial (PG) faults were discovered in the 1960s in northern Finland (Kujansuu 1964, 1972) and later elsewhere in northern Fennoscandia (Lagerbäck 1979, Olesen et al. 2004). The faults are classified as postglacial as the faults cut the Quaternary deposits overlaying the fault zone or the bedrock surface polished by glacial ice and have displacements from a few centimetres to 30 metres. They formed very soon after deglaciation and were probably accompanied by the most remarkable seismic events in Fennoscandia, with magnitudes up to eight (Kuivamäki et al. 1998; Olesen et al. 2004). Significant postglacial faults have been recognised only in the northern part of Fennoscandia (Figure 85). Only small PG faults located in ice-polished bedrock outcrops and with displacements up to 20 cm have been found in southern Finland. This does not, however, rule out the possibility of larger displacements. The main part of southern Finland belongs to an area which has been below the highest shoreline and the coastal erosion and deposition may mask the faults. However, there might not be any large faults (Ojala et al. 2004).

All large postglacial faults oriented NE-SW have a reverse sense of slip and they dip SE (Kuivamäki et al. 1998, Olesen et al. 2004). Johnston (1987, 1989) noted an apparent tendency for large continental ice sheets to suppress tectonic activity. Conversely, unloading would increase seismic activity, as illustrated in Figure 86 (Muir-Wood 1989). The ice load was removed last from the western parts of Fennoscandia; rebounding eastern parts with NW-SE compression favoured reactivation weaknesses trending NE-SW and dipping SE. Studies of the large Fennoscandian postglacial faults indicate that they are reactivated old faults (Kuivamäki et al. 1998; Olesen et al. 2004) whose history started in the Precambrian.

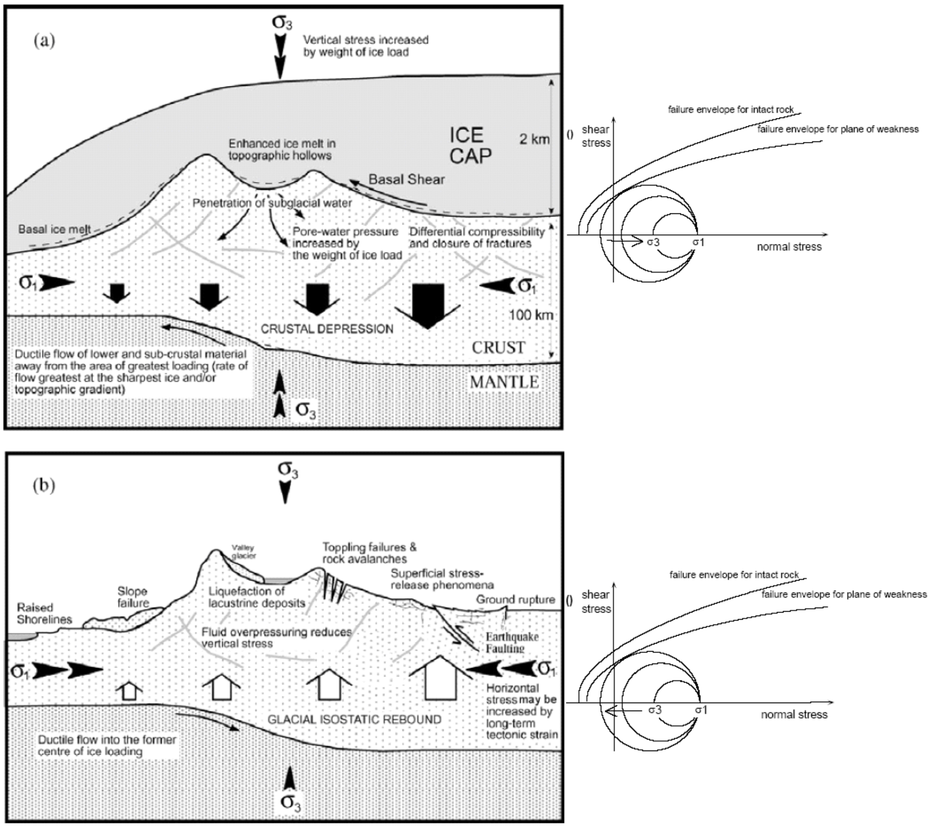

Figure 86. Diagrammatic representation of the impact of glacial loading (a) and unloading (b) on the crust and the resulting stress changes in a region with a compressive stress regime. Note the considerably exaggerated vertical scale difference between the crustal thickness (~100 km) and the ice-sheet thickness (~3 km) (Modified from Stewart et al. 2000). Schematic Mohr diagrams illustrating the stabilisation of structures under the ice load as the vertical stress (lithostatic load plus glacial load) (σ_3) increases and differential stress decreases. During unloading (σ_3) decreases, differential stress increases, which can lead to failure of a plane of weakness.

RECENT BEDROCK MOVEMENTS

The crust of the earth deforms in three dimensions but for practical reasons the traditional geodetic practice has been to observe these separately. For just over a decade, GPS positioning has made possible three-dimensional observations of crustal movements (e.g. Milne et al. 2001).

The land uplift rate in Fennoscandia was first measured from tide gauge recording and precise levelling. This also allowed the total amount of uplift to be estimated. In addition, these data were used to model the tectonic and glacial rebound components in the present uplift. The principle was that the misfit between observations and the isostatic uplift modelling was interpreted to reflect a tectonic component of the uplift (Figure 87). Anomalies in the uplift have been studied by several workers (e.g. Chen

1991; Fjeldskaar et al. 2000; Milne et al. 2001) and all of them have come to the same conclusion, that the tectonic component of the uplift is of the order of 10% (or 1 mm/yr).

Figure 87. Areas with significant (< 1.0 mm/yr) negative deviations (left) and positive deviations (> 1 mm/yr) (right) between the observations and the calculated glacial isostatic uplift (modified from Fjeldskaar et al. 2000).

Veriö et al. (1993) found few areas where the uplift deviates substantially from predictions based on uniform elastic uplift. These results, together with the results of the third precise levelling of Finland carried out by the Geodetic Institute of Finland, suggest that the present-day land uplift can be considered plastic on a regional scale (deformation is distributed into a large number of structures), but that on a local scale there can be small block movements. These block movements are preferentially concentrated within zones of pre-existing weakness. However, it has not been possible to pinpoint individual structures which would have controlled the deformation and the deformation distributed in a number of structures.

First estimates of the horizontal crustal deformations in Finland were based on measurements of the first-order triangulation network. Chen (1991) analysed the horizontal crustal deformations in Finland using the measurements of the first-order triangulation network with observations spanning from 1920 to 1985. On a large scale, compression in the NW-SE direction dominates and the strain pattern is heterogeneous (or has a “block-like structure”). Chen (1991) calculated the change rates of some lines across proposed geological boundaries and found extension rates up to 21.8±10.4 mm/yr on a line 140 km long in Pohjanmaa and compression rates up to 10.2±6.6 mm/yr along an 83-km-long line in Kajaani. Although the results are consistent, these measurements display large errors compared to the strain rates. However, the stress field correlates well with the seismological data.

The first analysis of the BIFROST GPS network data confirms that glacial isostatic adjustment dominates the ongoing three-dimensional crustal deformation in Fennoscandia (Milne et al. 2001). The vertical uplift maximum of 11.2 ± 0.2 mm/year is located near the site at Umeå. Horizontal movements are directed outward from this location on all sides. In further agreement with the numerical predictions, these rates increase with their distance from the uplift centre, and they reach about 2 mm/year at sites marking the perimeter of the BIFROST network. Consequently, the uplift in Fennoscandia results in an extensional component to the stress field. The horizontal rates are higher on the western side of the Gulf of Bothnia than on the eastern side (Milne et al. 2001).

CONCLUSIONS

Besides earthquakes, glacial isostatic adjustment and related land uplift horizontal motions are the most significant indications of the ongoing bedrock deformation in Fennoscandia, and these are closely connected. Based on GPS measurements, maximum vertical uplift rates are about 11 mm/yr and horizontal motions are up to 2 mm/yr. The tectonic component is about 10% of the land uplift (or 1 mm/yr). Horizontal motions are directed outward from the area of the fastest uplift. Horizontal tectonic motions are also less than 1 mm/yr.

Seismic activity in Finland is low and heterogeneously distributed and the earthquake density maximums and the areas of postglacial faults have a spatial correlation. Detailed geodetic surveys indicate that crustal deformation occurs unevenly. However, the bedrock in Finland is so fractured that the deformation is distributed over a number of structures and deformations and displacements along individual structures are very small and difficult to resolve. Fault intersections can form a locked area where stresses large enough to trigger intraplate earthquakes can build up. In the absence of intersections, the pre-existing faults can creep at a lower stress threshold.

In Fennoscandia, plate-boundary tectonic stresses drive the regional compressive stress field, but the glacial isostatic adjustment has a very important role in accounting for the current level of seismicity. The brittle crust is near the point of failure, and, consequently, small changes, like a glacial rebound (0.1 Mpa) can, in this state of stress, lead to earthquakes sufficient to reactivate optimally oriented pre-excising weaknesses.

Stress orientations inferred from the strain measurements of the first-order triangulation network and seismological stress data show: a) the dominant ridge-push/mantle drag-related compression, and b) evidence of significant local variations in the surface stress field influenced by the orientation of major fracture zones.

Postglacial faults are re-activated old faults and the areas of postglacial faulting are still the most seismically active areas in Fennoscandia. The association of seismicity with glacial rebound suggests that in areas experiencing diminishing rebound, the level of seismicity decreases over time. Therefore, palaeoseismicity and geological modelling have to be combined to predict the likely incidence, magnitude, and frequency of future earthquake activity.

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2.3.8 Brief summary of natural analogue studies at Palmottu[21]

GENERAL FRAMEWORK

From their beginning in 1987, natural analogue studies at Palmottu were conducted mainly under the auspices of the public sector's research programmes. During the first years the study was financed by the Ministry of Trade and Industry and by the STUK. Research was conducted as a cooperative project between research groups from GTK, Helsinki University of Technology (Rock Engineering), the University of Helsinki (Radiochemistry) and, later, the Technical Research Centre of Finland (VTT).

In 1996 the study grew into an international effort jointly funded by the European Commission, GTK, STUK, Svensk Kärnbränslehantering (SKB), Empresa Nacional de Residuos Radioactivos S.A (ENRESA), Centro de Investicationes Energéticas, Medioambientales y Tecnológicas (Ciemat), and Bureau de Recherches Géologiques et Minières (BRGM).

[21] By Lasse Ahonen, GTK.

After the EU project, Palmottu was a part of a Coordinated Research Project (CRP) launched by the IAEA "The use of selected safety indicators (concentrations; fluxes) in the assessment of radioactive waste disposal" for the period 1999–2003 (cf. Chapter 2.1.2). During recent years, material and data from Palmottu have been utilised in various studies addressing detailed questions related to transport, retardation, and paleohydrogeology. A summary of the Palmottu natural analogue studies, including a comprehensive bibliography, was made in the framework of the present KYT programme (Ahonen et al. 2004).

GEOLOGICAL FRAMEWORK

Palmottu is a small uranium deposit in Nummi-Pusula, south-western Finland. The site was discovered in 1979 by GTK in the course of a systematic airborne radiometric survey, and the uranium reserves were discovered at the beginning of the ’80s.

The crystalline bedrock of the Palmottu site represents a fairly good analogue to the planned nuclear waste disposal site at Olkiluoto. Both are situated within the Proterozoic Svecofennian orogenic belt, which was formed about 1.9 billion years ago. In general, the bedrock of both sites consists of migmatitic mica gneisses (Figure 88).

HYDROGEOLOGICAL STUDIES

In the first phase of the international EU Palmottu project, the hydrogeological framework of the Palmottu site was extensively studied (Jan 1996 – May 1997). The work was, however, preceded by GTK's own earlier borehole testing and development work, as well as the pre-feasibility studies conducted in cooperation with SKB in 1994–1995.

Regional-scale hydrogeological studies at Palmottu comprised an area of about 15–20 km^2, and the work aimed at an integration of the bedrock structures (lineament interpretation), Quaternary geology (glaciofluvial deposits), and topography (catchment of Lake Palmottu). Site-scale studies mainly utilised the existing borehole field of the ore exploration studies, but new and deeper research boreholes were also drilled. Bedrock fracturing and hydrogeological properties were thoroughly investigated, using a wide variety of research methods from single-hole measurements to large pumping tests and tracer experiments.

The results of the fieldwork were interpreted using different approaches, from semi-qualitative estimates to numerical flow modelling (Lampinen 1997, Ludvigson 1997, Koskinen and Kattilakoski 1997). Because of the project structure, Phase 1 of the EU Palmottu Project had a strict time frame, imposing severe demands on the integration of the field data for the development of appropriate hydrogeological models.

HYDROGEOCHEMICAL STUDIES

A remarkable amount of bedrock groundwaters were sampled and analysed during the Palmottu studies. Altogether, 33 boreholes from different depths (down to 417 m) were sampled, many of them several times. A total of 184 borehole water analyses are included in the Palmottu hydrogeochemical database (Kaija et al. 1998); see Figure 88. Additionally, about 30 surface and soil water samples were analysed. The major part (70%) of the bedrock (fracture) water samples were taken from packed-off sections after pre-pumping, while the tube sampling method (30% of samples) revealed the vertical distribution of groundwater types among the boreholes.

The main part of the samples analysed represents fresh calcium and/or sodium-dominated waters, with bicarbonate as the predominant anion. Brackish waters with salinities up to about 2 g/L (sulphate as the predominant anion) were commonly observed deeper in the bedrock, while saline chloride waters were not met in the depth range studied. A major conclusion was the bedrock structure controls of the existence of different groundwater types (Blomqvist et al. 2000).

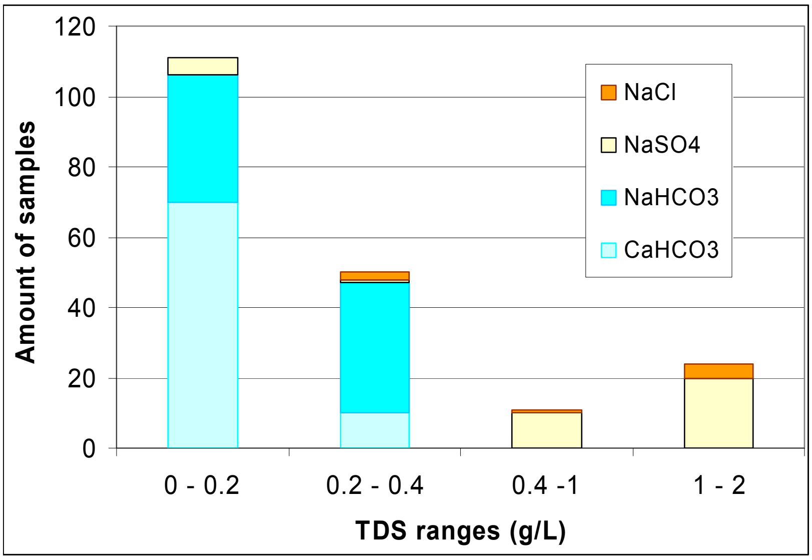

Figure 88. Distribution of water types and salinity (TDS) ranges at Palmottu. Average TDS of Finnish potable water is about 0.2 g/L. Surface and soil waters are included.

Considerable effort was put into the characterisation of the redox conditions of the Palmottu bedrock-groundwater system. During the field study, redox measurement methodologies were tested and developed. Finally, during the EU-financed study phase, the mobile laboratory of SKB was used to measure redox potential in the bedrock in situ. The results indicated the presence of a redox evolution sequence from near-surface oxidic conditions to deep reducing conditions. The redox control of uranium mobility was clearly demonstrated.

MIGRATION PROCESSES AND MODELLING

The ultimate goal of the Palmottu study was a quantitative description of radionuclide transport processes. Different migration phenomena were studied on the process level and incorporated into models which use the groundwater flow model as a basis.

The sorption of radionuclides on rock is a broad set of processes retarding transport. Sorption processes were studied using different techniques elucidating the nature and distribution of the sorbing phases and physicochemical processes involved in sorption. Matrix diffusion is a well-defined process in which sorption also plays a role. The outcome of these studies included estimates of the distribution coefficient (K_D) for U and matrix diffusion depth. Uranium series disequilibrium (USD) study is a versatile method for demonstrating uranium mobilisation/retardation processes and their time scale. The application of USD studies is described in Chapter 2.3.4. Colloidal material in groundwater is considered a potential carrier of radionuclides. Consequently, both inorganic (mainly iron and silica) and organic (humics fulvics) colloids were studied. The microbes in the Palmottu groundwaters were studied using conventional cultivation techniques.

Different teams and modelling approaches were included in the transport modelling exercise performed: the FTRANS code was used by VTT, the RETRASO code by CSIC (Barcelona), and ALLAN-NEPTUNIX software by BRGM. The models were based on at least partially dissimilar conceptualisations of the transport phenomena, implying that different sets of (or different emphases on) processes were chosen by different teams. Another modelling exercise was directed to the geochemical modelling of the speciation and solubility of selected elements. Three separate groups (Ciemat, GTK-VTT, QuantiSci) conducted the same "blind predictive modelling" (BPM) exercise using the same input but their own codes and data.

CONCLUDING REMARKS

The Palmottu natural analogue study has been a major effort within the Finnish public sector's nuclear waste disposal research. An additional credit to the study was its acceptance into a Europe-wide EU-funded project. As natural analogue studies often do, it has its roots in geological thinking; processes in the geological past can be considered as keys in predicting the future. One of the strengths of the study was its interdisciplinary nature. In addition to combining different branches of geosciences, the Palmottu study also encompassed radiochemistry from the beginning. Colloid and microbe studies widened the scope into the fields of applied chemistry and environmental microbiology, while matrix diffusion studies included the first initiatives to apply a modelling approach to the Palmottu project.

A very detailed hydrogeological site characterisation programme was carried out at the Palmottu study site. The work, including comprehensive research, testing, and development of field study methods, resulted in the construction of a hydrogeological model of the site, which was the primary input of the transport models developed. One of the major findings of the site study was that the extensive groundwater chemical data could be used as an additional support to the hydrogeological model. Additionally, the bedrock redox conditions of the Palmottu site were studied in depth, using the best available measuring methodologies. All these facts together make Palmottu an excellent reference site for the actual nuclear waste disposal sites planned in crystalline bedrock.

In a relatively early phase of the Palmottu study, it became clear that a dialogue between the analogue study and the performance assessment (PA) of the nuclear waste disposal should be an essential part of the project. The needs and outputs of the PA were finally coupled to the study when the EU-funded project started. Now, the results of the Palmottu analogue study provide valuable information for the safety aspects of nuclear waste disposal.

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2.4 Radionuclide transport in bedrock

Radionuclide transport studies in bedrock consist of four basic components. Laboratory-scale tracer experiments and related modelling have been pursued as domestic collaboration. The characterisation of the pore space geometry of crystalline bedrock by applying at the same time C-14-PMMA method, confocal microscopy method, and tomographic methods has been performed in domestic and international collaboration. The sorption of radionuclides in tunnel filling materials has been studied by applying experimental and modelling approaches. The KYT research programme has also participated in the OECD NEA Sorption Forum as sorption is a process of utmost importance in radionuclide transport considerations.

2.4.1 Laboratory-scale tracer experiments[22]

INTRODUCTION

The retention of radionuclides in the geosphere can be an important component of the safety case for the deep geological disposal of radioactive wastes. It represents an essential part of the performance of the geosphere as a barrier for the migration of radionuclides. The most important retention process of solute transport in fractured rock is matrix diffusion. However, because of the very low flow rates and long time scales required, it is not possible to study matrix diffusion directly under the flow conditions that will prevail around the closed underground repository. Estimation of radionuclide transport and retention needs to be based on the identification and understanding of the processes.

According to the literature, the majority of experiments carried out in natural fractures have been performed as in situ tracer experiments. Under in situ conditions the control of the flow conditions is difficult because of the natural background flows. The possibilities of controlling the local flow field are much better in laboratory-scale experiments.

Rock-block migration experiments were performed to examine fracture flow and radionuclide transport in a horizontal natural fracture. Rock-core column experiments were carried out to estimate the diffusion and sorption properties of the Kuru Grey

[22] By Pirkko Hölttä, HYRL; Antti Poteri, VTT.

granite used in block-scale experiments. The results of the column experiments are used to estimate radionuclide transport times and retardation parameters in the fracture before block-scale experiments.

The objective of this study is to examine the processes that cause retention in solute transport through rock fractures. In particular, the focus of the work is on matrix diffusion. The results can be used to estimate the importance of the retention processes during transport on different scales and under different flow conditions.

ROCK MATRIX CHARACTERIZATION

Kuru Grey granite was obtained from Kuru Quarry, Tampereen Kovakivi Oy. The total porosity and the surface areas of the mineral grains available for the sorption and migration of species were determined by the ^{14}C-PMMA method. Pore apertures and geometry in mineral phases were analysed by scanning electron microscopy (SEM) and the minerals and sorbed tracer were quantified by energy-dispersive X-ray micro analysis (EDX). The specific surface area of the solid rock was determined by the B.E.T. Hg impregnation method.

Kuru Grey granite is fine-grained, non-foliated, and equigranular and is composed of 36% potassium feldspar, 35% quartz, 21% plagioclase, and 8% amphibole and micas. The rock density given by Tampereen Kovakivi Oy is 2,660 $kg \cdot m^{-3}$. The total porosity determined by water gravimetry and the ^{14}C-PMMA method was 0.4%. The average grain size determined by SEM was 0.5–1.5 mm. The specific surface area was 0.03 $m^2 \cdot g^{-1}$ and the average pore diameter was 300–400 nm. The porosity value determined by Hg porosimetry was slightly higher as a result of artefacts caused by the small sample preparation. Grain boundary porosity dominated, though intragranular porosity was observed in biotite and feldspar grains. A disturbed zone was found about 1 mm from the surface, because of the drilling core and sawing of the sample.

THE ROCK CORE

The experimental design for rock-core column experiments is shown in Figure 89. Cores drilled into the fracture of the Kuru Grey granite block were glued one after the other to form 74.5-cm, 68.5-cm, and 28-cm long columns. Cores 14 mm in diameter are placed inside a tube to form a flow channel in the 0.5-mm gap between the core and the tube. The volume of the flow channel in the 74.5-cm column is about 17 ml and the tubing takes about 1.3 ml.

The rock core columns were used in experiments to determine the transport properties of the rock material. Tracer experiments were performed using a peristaltic pump to control the water flow rate. Water was fed into the column using different flow rates of 2–50 μl·min^{-1}. A short tracer pulse (5 μl) was injected into the water flow using an injection loop (Rheodyne) and the outflowing tracer was collected. In all experiments synthetic granitic groundwater equilibrated with crushed Kuru Grey granite was used. The main cation concentrations were determined using ICP-MS spectrometry.

Several tracer tests were performed using Uranine, HTO, and ^{36}Cl as non-sorbing, ^{22}Na as a slightly sorbing, and ^{85}Sr as a sorbing tracer with injection flow rates of 2–50 μl·min^{-1}. The transport of tracers through the core columns was modelled by applying the same advection-dispersion model as used in the block-scale fracture flow experiments. The model is based on matrix diffusion to the rock matrix and generalised Taylor dispersion in the description of the flow field. The Taylor dispersion model is based on the assumption of a linear velocity profile across the flow channel.

The most interesting results were obtained with the 68.5-cm long column. Both the experimental and modelled breakthrough curves of Uranine and ^{22}Na for total flow rates of 20, 6, and 3 μl·min^{-1} are presented in Figure 90. According to the modelling results, clear signs of matrix diffusion can be seen in the tailings of the sorbing tracer (^{22}Na). The highest flow rate (20 μl/min) is advection-dominated for both of the tracers and also the smallest flow rates, 6 and 3 μl/min, for the non-sorbing tracer. This can be verified from Figure 90, where the breakthrough curves are presented both with and without matrix diffusion. The tailings of the breakthrough curves for the smallest flow rates of the ^{22}Na are well reproduced by the matrix diffusion in the modelled results. Note that the only difference between the Uranine and ^{22}Na breakthrough curves in the model is the effect of the stronger matrix diffusion for ^{22}Na resulting from the sorption in the rock pore space. The distribution coefficient of $K_d = 0.0006$ m^3/kg applied for the ^{22}Na in the model was measured for the mica gneiss in the rock column experiment.

A stronger sorbing tracer (Strontium) was applied in the experiments with the shorter 28-cm rock column. However, the system was not as stable as was the case with the longer column. This means that the non-sorbing breakthrough curves required for checking the influence of the flow field are not as reliable as those for the 68.5-cm rock column presented in Figure 90.

The parameters used to model the breakthrough curves in Figure 90 are summarised in Table 11.

Table 11. Parameters applied in the modelling of the breakthrough curves in Figure 90.

Parameter	**Channel 2**
Transport and retention processes	Advection (linear velocity profile), Taylor dispersion, Matrix diffusion
Channel length	68.5 cm
Channel width	4.4 cm (circumference of the column)
Channel aperture	0.7 mm
Flow rate	20 μl/min, 6 μl/min and 3 μl/min
Width of the velocity profile	2.2 cm
Matrix porosity	0.4%
Matrix density	2 660 kg/m^3
Diffusivity in free water	Uranine: $2\cdot10^{-9}$ m^2/s ^{22}Na: $2\cdot10^{-9}$ m^2/s
Pore diffusivity (based on the Archie's law for the formation factor $F = 0.71\,\varepsilon^{1.58}$)	Uranine: $6.6\cdot10^{-11}$ m^2/s ^{22}Na: $6.6\cdot10^{-11}$ m^2/s
K_d	Uranine: 0 m^3/kg ^{22}Na: 0.0006 m^3/kg
Ka	Uranine: 0 m ^{22}Na: $2\cdot10^{-5}$ m

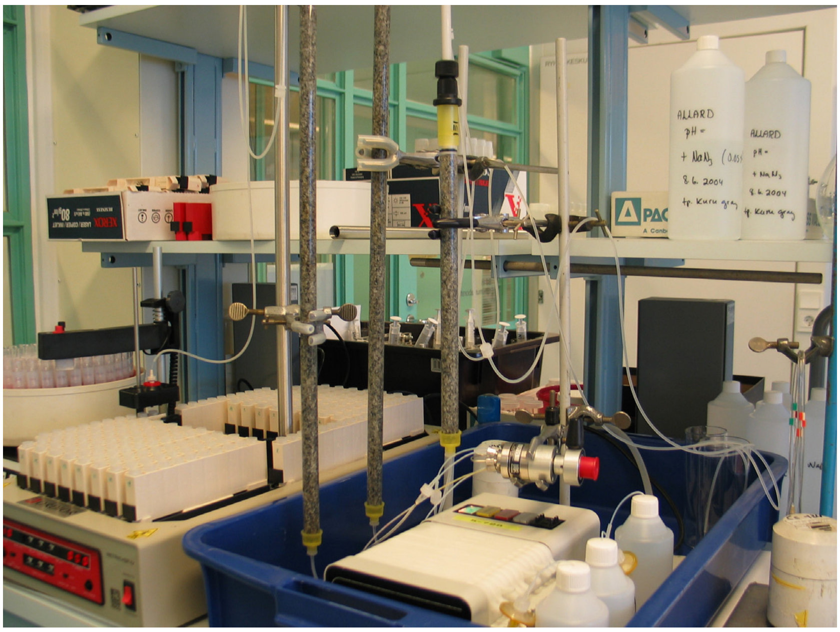
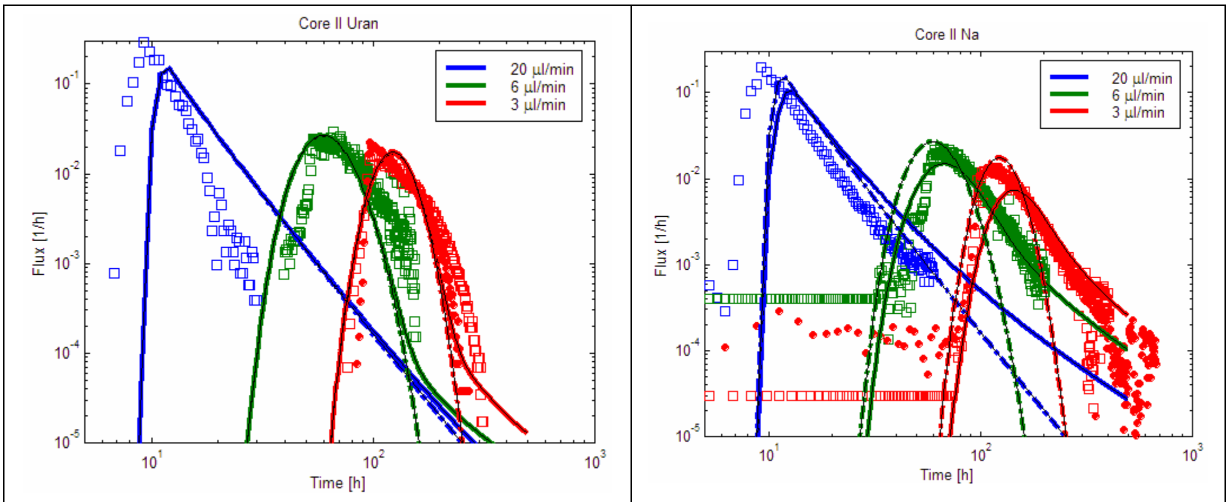


Figure 89. Experimental design used in rock-core column experiments. Cores drilled to the fracture of the Kuru Grey granite are placed inside a tube to form 74.5-cm-, 68.5-cm-, and 28-cm-long columns. Core diameter is 14 mm and flow channel is in the 0.5 mm gap between the core and the tube.



Column 2, Uranine　　Column 2, ^{22}Na

Figure 90. Modelled and measured breakthrough curves (BTC) for the 68.5-cm-long rock column. Markers indicate measured BTC and lines indicate the modelling results. Modelled BTCs are shown for advection-dispersion only (dashed line) and including matrix diffusion (solid line). The only difference in the model between Uranine and ^{22}Na BTC is the sorption of the ^{22}Na, which enhances the matrix diffusion of the ^{22}Na. on the basis of the modelling; the tailings of the measured 6- and 3-µl/min BTC for the ^{22}Na show clear signs of matrix diffusion.

THE ROCK BLOCK

The rock block is medium-grained grey granite and it contains a natural hydraulically conducting fracture. The size of the block is approximately 0.9 m x 0.9 m x 0.7 m and the horizontal fracture is located about 17 cm below the top of the block (see Figure 91). The fracture is intersected by nine vertical boreholes. The borehole in the middle of the block has a diameter of about 3 cm and all the other boreholes are 2 cm in diameter. The locations of the boreholes are presented in Figure 92. The cores of the boreholes were used in the concurrent rock column experiments that were carried out to verify the transport properties of the rock.

The rock block is equipped with water pools that are installed at the vertical sides and top of the block. The purpose of these water pools is to ensure the saturation of the block and also to stabilise the hydraulic head around the vertical faces.

The rock block is also instrumented, besides the boreholes, at the outer vertical boundary of the block where the horizontal fracture intersects the faces of the block. One face is equipped with tracer collection cells. Preliminary tracer tests with Uranine showed that migration may take place through distinct channels. The tracer collection cells are used to directly measure the breakthrough curves of the different transport channels.

Figure 91. The experiments were carried out in the horizontal natural fracture located in the upper part of the rock block. Water pools installed on the vertical faces of the rock block are used to apply appropriate boundary conditions to the outer boundary of the fracture.

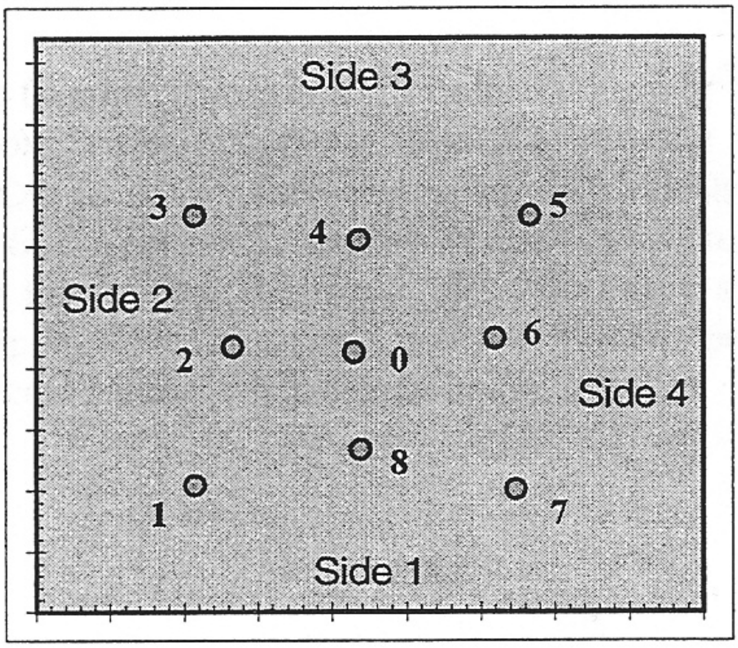


Figure 92. Locations of the boreholes at the top of the rock block (picture from Hölttä and Hakanen (2002)). The size of the block is about 90 cm x 90 cm. Side 3 is the one facing the viewer in Figure 91.

HYDRAULIC CHARACTERISATION

The hydraulic characterisation of the fracture is an essential part of the interpretation and planning of the transport and retention experiments. The hydraulic properties of the fracture were determined by measuring water inflow for different hydraulic heads and different boreholes, as shown in Figure 93. The fracture aperture is very small at one of the corners. The fracture is practically closed at the corner between Side 1 and Side 4 (see Figure 92). The aperture increases towards Side 3. This can also be deduced from Figure 94. Boreholes 7 and 8 did not conduct water at all and they are not included in Figure 93.

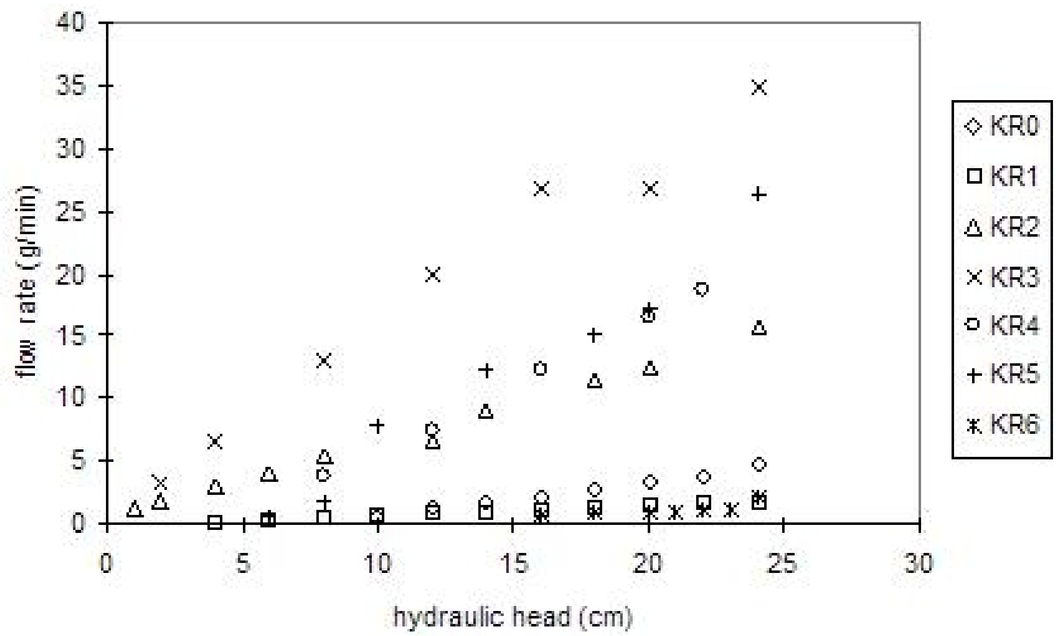


Figure 93. Water inflow to the fracture at different boreholes as a function of the applied hydraulic head (based on data in Hölttä & Hakanen (2002)).

Interpretation of the pumping experiments in Figure 93 by applying a simple radial flow field gives estimates of the local transmissivities around the different boreholes. Figure 94 shows the estimated transmissivities. This figure shows clearly the increase in the transmissivity (and very probably also in the aperture) as Side 3 is approached. The locations of boreholes 7 and 8 are indicated by dots, as they had zero transmissivity in the hydraulic tests.

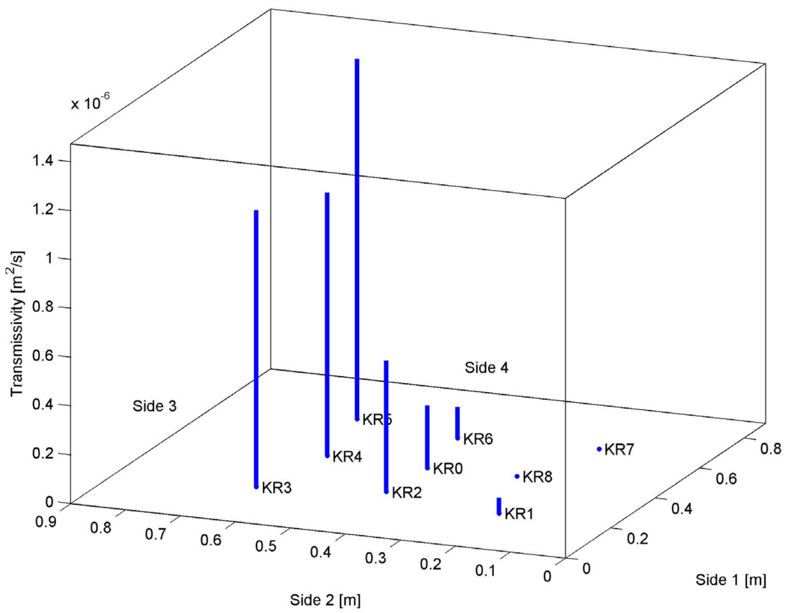


Figure 94. Locations and estimated local transmissivities for different boreholes. Locations of boreholes 7 and 8 are also indicated, although they showed zero transmissivity in the test (figure is based on Hölttä et al. (2004)).

TRACER EXPERIMENTS

A suitable configuration for the tracer experiments was studied extensively in the first phase of the experiment (Poteri & Hölttä 2005). Tracer experiments were mainly carried out from the borehole KR1 to Side 3 of the rock block. This gives the longest transport distance along the fracture, about 70 cm. The majority of the tracer tests were carried out using injection flow rates from 10 µl/min to 50 µl/min. All tests were performed in a radially diverging flow field by pumping the tracer injection borehole and keeping the outer boundary of the fracture in the constant hydraulic head. Tracer was collected at Side 3 of the outer boundary of the fracture. The majority of the experiments give tracer recovery in adjacent locations at the outer boundary of the fracture. This proves that several parallel flow paths exist. On the basis of the tracer experiments it was noticed that the stability of the flow field in the fracture is a problem. Successive tests applying the same experimental configuration led to rather large variations between the transport channels, i.e. the main transport channel may vary from test to test. This also makes it difficult to determine the division of the total pumped flow rate into the individual transport paths.

However, a test was carried out using a 10-µl/min pumping rate and using a tracer cocktail of Uranine and ^{22}Na (Figure 95). This test shows recovery from two transport channels for both Uranine and ^{22}Na in a very consistent way. Modelling this test shows that the difference between the Uranine and ^{22}Na breakthrough curves can be consistently explained by the matrix diffusion for both channels and using parameters that are also consistent with the rock column experiments. The main difference from the rock column experiment is a slight increase in the porosity of the rock, from 0.4% in the column experiments to 0.5% in this fracture experiment. The transport and retention processes were the same in both column and fracture experiments.

Figure 95 shows the measured and calculated breakthrough curves for the two channels (Channel 2 and 4) and two tracers (Uranine and ^{22}Na). The modelling results of the Uranine for both channels are advection-dominated and there is no difference, regardless of whether matrix diffusion is taken into account or not. However, the difference in the breakthrough curves between the modelled Uranine and ^{22}Na comes from the matrix diffusion. It is caused by the enhancement of the matrix diffusion as a result of the sorption of the ^{22}Na in the rock matrix ($K_d = 0.0006$ m^3/kg). Note that the only difference between the Uranine and ^{22}Na in the model is in the K_d.

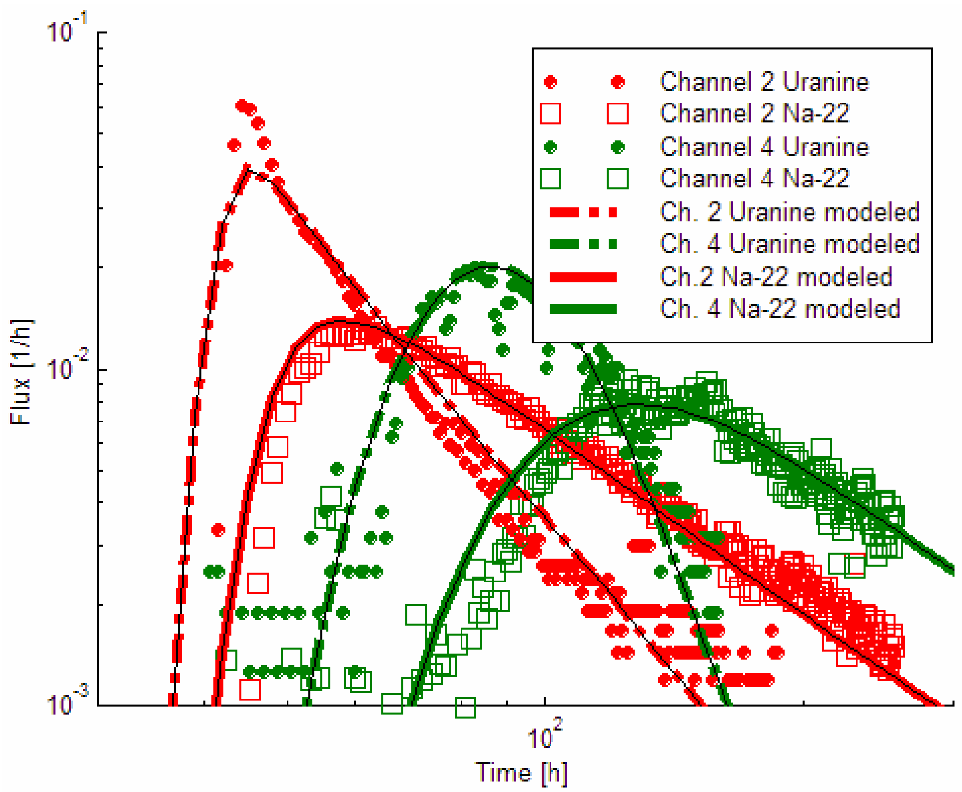


Figure 95. Measured and modelled breakthrough curves for tracer transport from borehole KR1 to tracer collection cells 2 (red) and 4 (green). Measurements are presented by means of markers and modelling results by lines. Two different tracers were used in this experiment. The only difference in the model between the tracers is in the tracer sorption properties (see Table 12). Both measured and modelled breakthrough curves have been normalised to give a unit mass recovery at the termination time of the measurements.

The parameters used to model the breakthrough curves in Figure 95 are summarised in Table 12.

Table 12. Parameters applied in the modelling of the results in Figure 95.

Parameter	**Channel 2**	**Channel 4**
Transport and retention processes	Advection (linear velocity profile), Taylor dispersion, Matrix diffusion	Advection (linear velocity profile), Taylor dispersion, Matrix diffusion
Channel length	75 cm	80 cm
Channel width	6 cm	3.5 cm
Channel aperture	0.65 mm	0.5 mm
Flow rate	7 μl/min	3 μl/min
Width of the velocity profile	3 cm	2.4 cm
Matrix porosity	0.5%	0.5%
Matrix density	2 660 kg/m^3	2 660 kg/m^3
Diffusivity in free water	Uranine: $2 \cdot 10^{-9}$ m^2/s ^{22}Na: $2 \cdot 10^{-9}$ m^2/s	Uranine: $2 \cdot 10^{-9}$ m^2/s ^{22}Na: $2 \cdot 10^{-9}$ m^2/s
Pore diffusivity (based on the Archie's law for the formation factor $F = 0.71\,\varepsilon^{1.58}$)	Uranine: $6.6 \cdot 10^{-11}$ m^2/s ^{22}Na: $6.6 \cdot 10^{-11}$ m^2/s	Uranine: $6.6 \cdot 10^{-11}$ m^2/s ^{22}Na: $6.6 \cdot 10^{-11}$ m^2/s
K_d	Uranine: 0 m^3/kg ^{22}Na: 0.0006 m^3/kg	Uranine: 0 m^3/kg ^{22}Na: 0.0006 m^3/kg
Ka	Uranine: 0 m ^{22}Na: $2 \cdot 10^{-5}$ m	Uranine: 0 m ^{22}Na: $2 \cdot 10^{-5}$ m

CONCLUSIONS

The results of this experiment provide evidence that it is possible to investigate matrix diffusion in a natural fracture on the laboratory scale. Two different experimental configurations, a rock core column experiment and fracture experiment, can be modelled using consistent parameterisation. The effects of the matrix diffusion on the tracer breakthrough curves can be demonstrated by modelling the different tracer which, because of the sorption, has a different residence time history in the rock matrix.

This experiment builds confidence in the model predictions of radionuclide retention through the network fractures in bedrock. The experimental results presented here cannot be directly transferred to the spatial and temporal scales that prevail in the underground repository. However, this exemplifies that the advection-dispersion and matrix diffusion processes can be conceptualised with sufficient accuracy to reproduce the experimental results. This knowledge and understanding of the transport and retention processes is transferable to different scales.

The variability observed in the successive tests using the same experimental configuration underlines the importance of the flow field in the assessment of the transport and retention of the solutes. This is an important result for the assessment of radionuclide transport in the expected repository conditions. The experiments performed provide variability in the flow conditions, but repeated tests under well-controlled flow conditions and further analysis of all the tests can provide a firm basis for the flow coupling of the retention.

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2.4.2 Studies of pore-space geometry with the C-14-PMMA method[23]

INTRODUCTION

Because of uncertainties in the transport pathway and real pore space characterisation, there has been a tendency to play down the potential role of the geosphere as a safety barrier in repository performance assessment. When evaluating repository safety, today it is necessary to go from laboratory and surface-based field work to the repository level

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underground. For upscaling matrix diffusion and sorption processes to the real metre-scale conditions *in situ*, realistic data on the properties of the physical rock matrix are needed. Little is known about changes in rock transport properties during sampling and decompression. Information is needed about the reliability of laboratory-based rock characterisation methods, and this can be achieved by quantitative investigation of the effects of rock stress release on total and mineral-specific porosities. This is the basis for an evaluation of changes in the transport properties of different rock types under different stress conditions.

The ^{14}C-labelled polymethylmethacrylate (^{14}C-PMMA) method has been developed for the characterisation of the pore-space geometry of low-permeability granitic rocks. So far rock matrices have been studied in the laboratory. The porosity of the rock, the morphology of the pore space, i.e. tortuosity, constrictivity, pore apertures and connectivity, as well as the accessible pore space, have been studied from the submicrometre to the decimetre scales. Impregnation with ^{14}C-labelled methylmethacrylate (^{14}C-MMA), together with autoradiography, makes possible the determination of the spatial distribution of porosity. Quantitative measurement of the total or mineral-specific local porosity has also been developed using image analysis tools.

The development of the ^{14}C-PMMA method for *in situ* applications comprises the impregnation of decimetre-long rock cores (Kelokaski et al. 2005), the impregnation of rock blocks (30 cm × 30 cm × 30 cm) in the laboratory, and, furthermore, *in situ* impregnations conducted at the Grimsel Test Site (GTS) within the Phase VI project cluster (Pore Space Geometry project). The results of the block-scale investigations, as well as those of the *in situ* experiment, are reported here. *In situ* conditions were simulated in the block-scale experiments by intruding ^{14}C-MMA into water-saturated Kuru grey granite (permeability 10^{-18}m^2, porosity 0.4%). The tests were focused on the drying of the matrix, impregnation in a vacuum, and optimising the heating-polymerisation conditions. The visualisation of the conductive pore space was performed by autoradiography, which produced 2D images of the intra- and intergranular pores of the granite sample. In these experiments intragranular porosity was revealed in unsaturated zones, implying that water effectively inhibits the intrusion of ^{14}C-PMMA. In addition, the intrusion *in situ* of ^{14}C-MMA into Grimsel granodiorite was verified.

The aim of this work was to perform an *in situ* experiment using ^{14}C-MMA resin impregnation in the underground rock laboratory of Nagra (Swiss National Cooperative for the Disposal of Radioactive Waste). This Grimsel Test Site (GTS) is located in the central Swiss Alps. The first results of the *in situ* development work were presented earlier by Kelokaski et al. (2005). The results of the block-scale investigations, as well as those of the *in situ* experiment, are reported here.

EXPERIMENTAL

Samples

The rock matrix used in the core-scale experiments, as well as the rock material in the *in situ* experiment, was Grimsel granodiorite. The core-scale samples were taken from the site which was earlier studied by Möri et al. (2003). Figure 96 illustrates the site at GTS where the boreholes for the PMMA *in situ* experiment were cored. The length of the injection borehole was 127 cm and its diameter 4 cm. The ventilation/temperature observation boreholes were 15 cm away from the injection borehole. They were 160 cm long and had a diameter of 1.6 cm. The injection area (length 20 cm) was confined by packers (length 7 cm) at each end. Overcoring was performed with an 80-cm-long and-300-mm-wide single core barrel which provided an intact 1.17-m-long drill core (PSG 04.001-OC). Sub-samples for different porosimetry analyses were sawn from five separate overcore slices (thickness 4–5 cm).

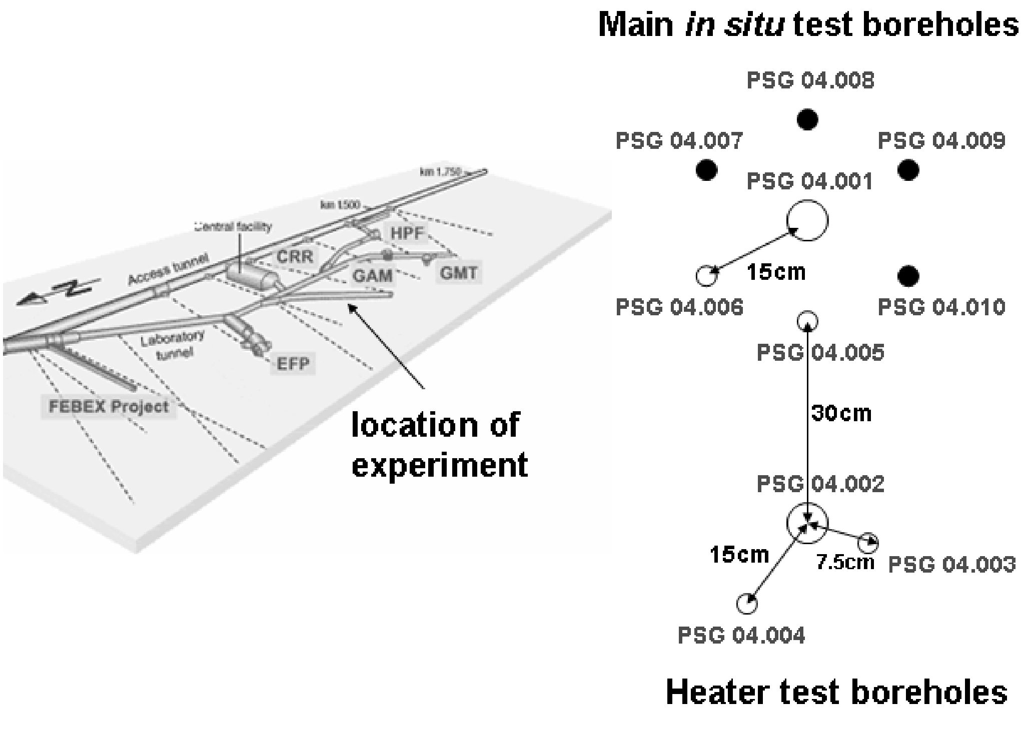

Figure 96. Location and setup of the in situ *experiment at GTS.*

Kuru grey granite was chosen for the block-scale experiments because the porosity and permeability of this rock were found to be close to those of Grimsel granite. The sizes of the rock blocks were 30 cm × 30 cm × 30 cm. To simulate *in situ* conditions the block was placed in a water bath. An airtight casing prevented water loss during drying, impregnation, and polymerisation. Experimental procedures for the core and block scale, as well as the *in situ* test, are given in Table 13.

Table 13. Experimental procedures for the core-scale, block-scale, and in situ *tests. The results obtained for porosity are also shown.*

	Core scale	**Block scale**	***In situ***
Rock matrix	Grimsel granodiorite	Kuru grey granite	Grimsel granodiorite
Dimensions	length 3–11 cm Ø 3–7 cm	30 cm × 30 cm × 30 cm	at the depth of 1 meter from the tunnel wall
Drying /time	vacuum, aluminium chamber /1–2 weeks	air flushing + vacuum /1–3 weeks	air flushing /3 weeks
Impregnation /time	vacuum, aluminium chamber /3–14 days	initial vacuum /7–12 days	5 bar pressure /36 days
Polymerization	irradiation with ^{60}Co source	heating, chemical initiator	heating, chemical initiator
Tracer activity	888 kBq mL^{-1}	888 kBq mL^{-1}	22 kBq mL^{-1}
Analysis	* autoradiography (ag) * water gravimetry * MMA gravimetry * PMMA porosimetry * Hg porosimetry * SEM + CLSM	* autoradiography * water gravimetry * PMMA porosimetry * Hg porosimetry	* PMMA ag * water gravimetry * thermo gravimetry * Hg porosimetry * CLSM * tracer activity too low for ag
Depth of penetration	samples fully impregnated	max. 6 cm	max. 5 cm
Porosity determined	0.7 ± 0.2%	0.4 ± 0.1%	from 0.6 to 0.1%

The ^{14}C-PMMA method

The ^{14}C-PMMA method makes it possible to study the spatial distribution of pore space in rock and also the heterogeneity of the rock matrix on the submicrometre to centimetre scale (Hellmuth et al. 1993, 1995; Siitari-Kauppi et al. 1998, 2003; Autio & Siitari-Kauppi 1998). The method involves the impregnation of centimetre-scale rock cores with ^{14}C-MMA in a vacuum. This is followed by irradiation polymerisation, sample partitioning, autoradiography, optical densitometry, and porosity determination using digital image-processing techniques. The labelled low-molecular-weight and low-viscosity monomer ^{14}C-MMA, which wets silicate surfaces well, provides information about the accessible pore space in crystalline rock. Total porosity was determined by employing 2D autoradiographs of sawn rock surfaces. The preconditions for applying this method are: (i) a known local bulk rock density; (ii) the presence of only two phases – mineral and ^{14}C-PMMA, and (iii) the homogeneous distribution of pores and minerals below the limit of the lateral resolution of autoradiography. The method effectively fills the gap between macroscopic and microscopic investigation methods. In addition, it provides quantitative information about nanometre-range porosity which is beyond the scope of most standard methods of microscopic investigation.

The conventional ^{14}C-PMMA method had to be changed for *in situ* conditions. Drying and impregnation had to be performed in atmospheric pressure, and irradiation polymerisation was not possible. The rock matrix was dried by pumping air for several weeks into the ends of the outer boreholes. In one block experiment, vacuum drying was used. Resin was polymerised by heating according to the methodology developed by Möri et al. (2003). Bentsoyl peroxide was added as a chemical initiator (1.25%) to initiate the polymerisation of the acrylic resin. Two downhole heaters were placed into injection (PSG 04.001) and heater test (PSG 04.002) boreholes. The rock temperature in the observation boreholes rose to 32ºC.

Determination of porosity and characterisation of microscopic porosity

In addition to the ^{14}C-PMMA method, three different conventional techniques were applied for a quantitative porosimetry investigation on the core-scale, block-scale, and *in situ* impregnated samples along with non-impregnated samples. The procedure for water saturation porosimetry basically follows the standard method of the International Society for Rock Mechanics (1979). It was modified so that the water saturation time was followed up to 32 days. Standard mercury injection porosimetry was applied (AutoPore III Micromeritics), for the determination of porosity and pore size distribution. Finally, a method based on thermo-gravimetric analyses was applied. Sliced sub-samples from the impregnated granodiorite *in situ* slabs were crushed into powder. Polymethylmetacrylate was determined by heating the crushed rock samples up to 500ºC (Mettler Toledo TA8000), which is the temperature at which MMA decomposes into carbon dioxide. The amount of MMA and the porosity can then be determined from the weight loss in the crushed rock sample (Kelokaski et al. 2006).

Scanning electron microscope examinations were executed on the core-scale samples to determine in detail the mineral-specific porosity and to characterise the geometry of the pores. ^{14}C-MMA resin was doped with fluorescent dye for confocal laser-scanning-microscope (CLSM) studies. This method produces 3D figures with 1-µm resolution of the impregnated pore space; mainly micro fissures are revealed by this method (Montoto et al. 1995).

RESULTS

Core scale

It was possible completely to impregnate the pore spaces of the core samples of varying size under laboratory conditions once the samples were properly dried. The total porosities determined by the ^{14}C-PMMA method (0.5–0.8%) were in good agreement with those measured by conventional methods (0.6–0.9%). All mineral phases were found to be porous and could be visualised with ^{14}C-PMMA autoradiographs. Intergranular and intragranural fissures and pore apertures in biotite and feldspar grains were also visualised by CLSM when ^{14}C -MMA was doped with a fluorescent dye. More detailed results of the core-scale experiments on Grimsel granodiorite have been presented by Kelokaski et al. (2005).

A photographic image of Grimsel granodiorite, its corresponding autoradiograph, and a magnified image which shows the pore structure in detail (porosity 0.55%) are shown in Figure 97. Different shades of grey in the autoradiographs correspond to different porosities; the darker the shade, the higher the porosity.

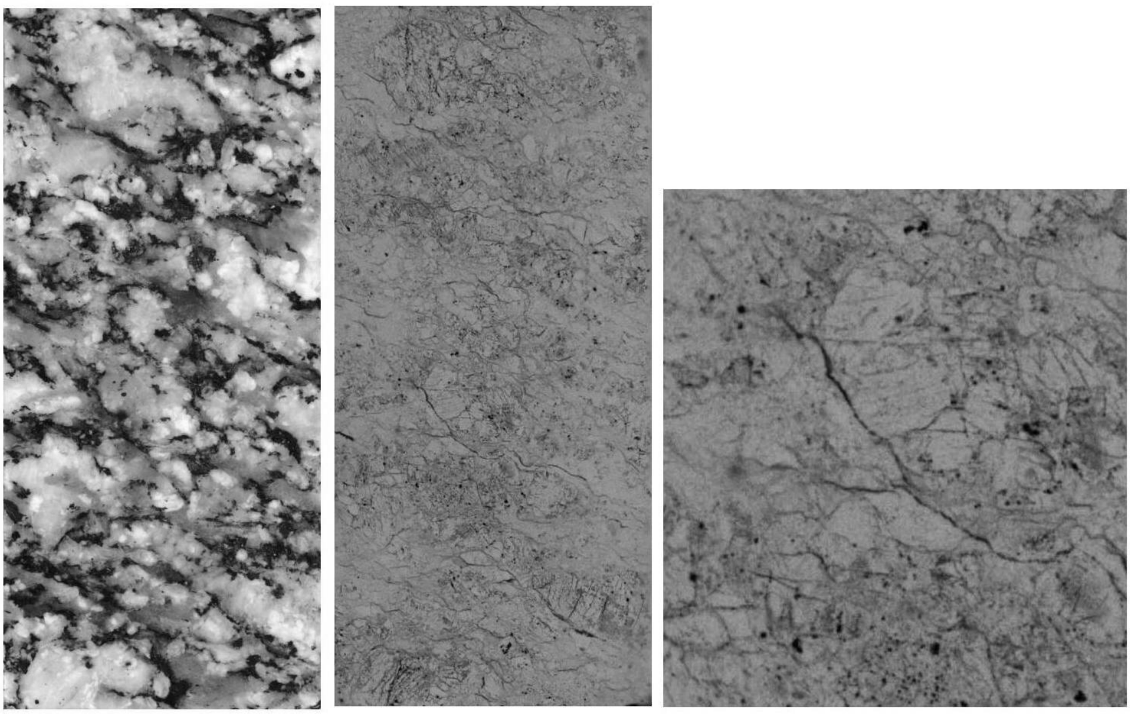

Figure 97. A photographic image of Grimsel granodiorite, its corresponding autoradiograph (sample height 5 cm), and a magnified image which shows the pore structure in detail.

Block scale

Figure 98 illustrates the experimental setup for the block-scale tests. Six sub-samples (CO1, CO2…) were cored and sawn into two pieces for autoradiography. A photographic image of a sample of Kuru grey granite and its corresponding autoradiograph are shown on the right. The resin intruded to a depth of 6 cm at the maximum. Vacuum drying and impregnation clearly improved the penetration of the tracer. The porosity values found (0.3–0.4%) were in line with the results achieved in earlier laboratory experiments (0.4%). Porosity was slightly lower at the interface of the ^{14}C-MMA and water (0.2–0.3%). Grain boundary porosity dominated in that zone; otherwise, intragranular pores were also impregnated. The main reason for the insufficient intrusion of the resin was found to be the residual water in small pores.

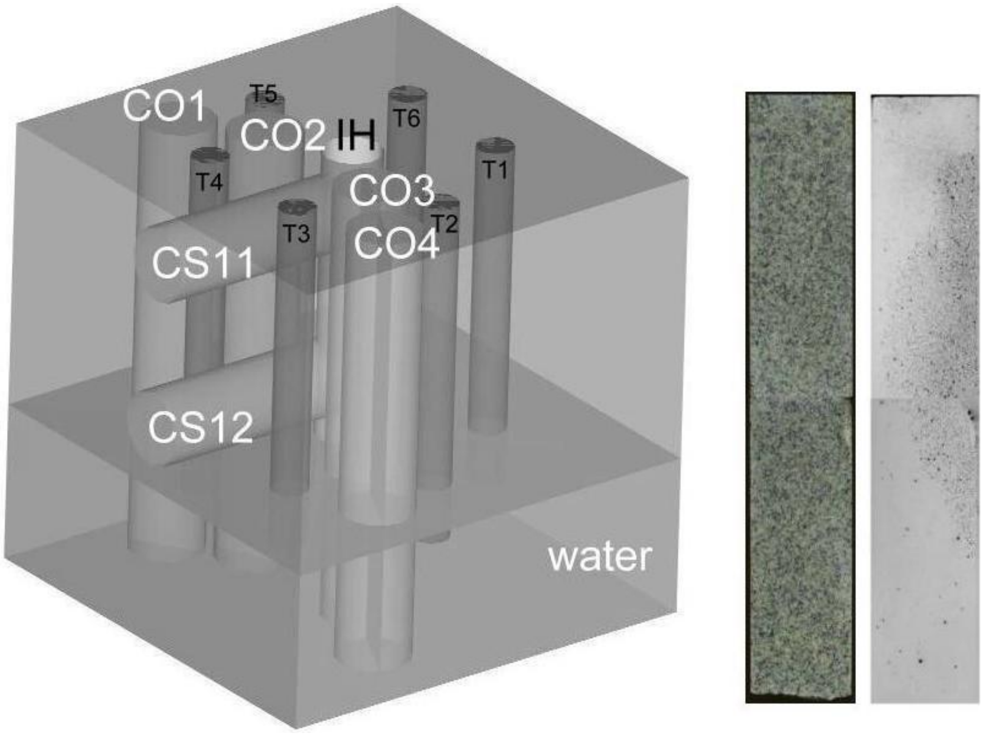

Figure 98. Setup for the block-scale tests, in which the air ventilation/temperature holes (T1-T6) surrounding the injection borehole (IH) are also shown. Six samples (CO1-CO4, CS11, CS12) were cored for autoradiography. A photographic image of Kuru grey granite sample CO2 and its corresponding autoradiograph (sample height 28 cm, width 5 cm) are shown on the right.

In situ

^{14}C-MMA intruded in the rock matrix only to depths of 3–5 cm. It was clearly observed that air- ventilation drying around the injection borehole was not effective enough. The resin autopolymerised during the impregnation step in the vessels, but heating polymerisation was executed to assure polymerisation in the rock matrix. The overcored rock sample was sawn into five slices and their surfaces were autoradiographed. Unfortunately, the tracer activity (22 kBq mL^{-1}) was too low for it to be possible to achieve PMMA autoradiographs (film or digital). Part of one slice was re-impregnated in the laboratory to achieve the porosity distribution shown in Figure 99. Several other sub-samples of the overcore slices were analysed with different porosimetry methods. Because of the shrinking of MMA, water-saturation gravimetry and mercury porosimetry showed 0.10–0.15% residual porosity for core-scale impregnated samples. The porosity of the sub-samples close to the injection borehole surface was 0.32% by Hg porosimetry and 0.35% by water porosimetry. Sub-samples taken from more than 7 cm away from the injection borehole had a 0.7% porosity by Hg porosimetry and a 0.8% porosity by water porosimetry. The thermo-gravimetry (TG) porosities from the corresponding places were 0.5–0.7%. Table 14 presents the porosity results obtained.

Table 14. Results of the porosimetry analyses of the sub-samples taken from the in situ *overcore PSG 04.001-OC, and the core-scale results for comparison.*

Water-saturation gravimetry	
0.6–0.9%	virgin Grimsel granodiorite, core scale
0.10–0.15%	residual porosity after impregnation due to shrinking
0.35%	*in situ* impregnated sample, close to the injection-borehole surface
0.7–0.9%	*in situ* impregnated sample, far from the injection-borehole surface
Mercury porosimetry	
0.7%	virgin Grimsel granodiorite, core scale
0.12%	residual porosity after impregnation due to shrinking
0.32%	*in situ* impregnated sample, close to the injection-borehole surface
0.52–0.81%	*in situ* impregnated sample, far from the injection-borehole surface

It was possible to analyse the real *in situ* porosities by TG and the results are shown in Figure 99. The porosity profile provides evidence of the insufficient intrusion of the resin, which might be due to insufficient drying of the open pore space. In addition, observations by CLSM showed the intrusion of ^{14}C-MMA into all other pore types and minerals except the biotite phases.

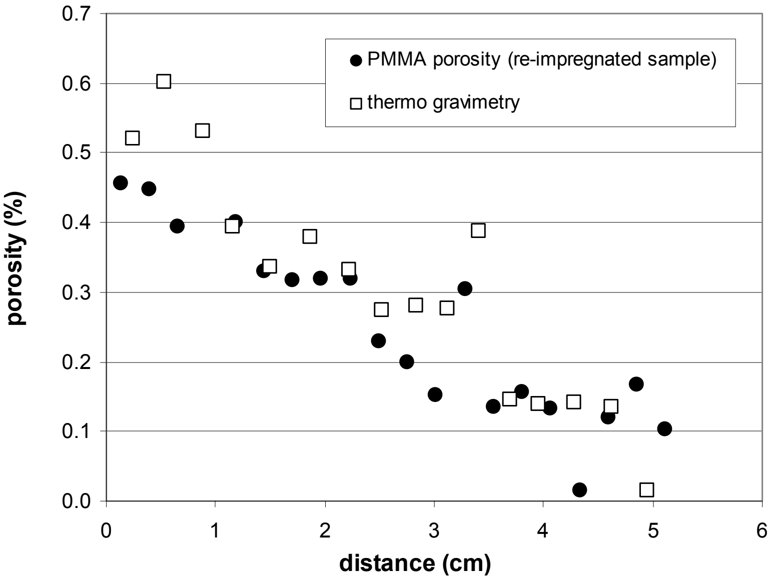

Figure 99. Porosity as a function of distance from the injection borehole surface. It decreases rapidly with increasing distance. Porosity was measured by the TG method and by PMMA autoradiography after re-impregnation.

The penetration of ^{14}C-MMA into Grimsel granodiorite *in situ* was successfully achieved. Porosity values were 20–30% lower under the *in situ* conditions than in the laboratory experiments, which is in agreement with earlier *in situ* experiments (Möri et al. 2003). This suggests that measurements in the laboratory might overestimate the porosity of the rock matrix as a result of sub-sampling artefacts. However, water effectively inhibits the intrusion into micro-pore apertures, and therefore the reason for the lower porosity values might be insufficient drying during impregnation.

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2.4.3 Studies of pore-space geometry with confocal microscopy[24]

INTRODUCTION

Within the framework of geological disposal of high-level radioactive waste, water movement and the related pathways, which involve many relevant length scales in the

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rock mass of a possible disposal site, are among the key questions to be understood and predicted. On the rock matrix scale, one has to determine the connected pores and fissures through which the flow takes place.

The imaging, mapping, and quantification of open effective porosity in low-permeable granitic rocks were traditionally performed by fluorescence microscopy and scanning electron microscopy (secondary electron detector and/or back-scattered electron detector). Since the beginning of the 1990s, new techniques and procedures have, however, been developed: confocal laser scanning microscopy (CLSM) (Montoto et al. 1995), ^{14}C-polymethylmethacrylate (PMMA) (Hellmuth et al. 1993, 1995; Siitari-Kauppi et al. 1998), the impregnation technique, and x-ray microcomputed tomography µCT (Lindquist & Venkatarangan 1999). More specifically, all these techniques aim at the 3D imaging of the possible water pathways.

The main objective of this work was to demonstrate the advantages the three new techniques (µCT, CLSM, and ^{14}C-PMMA) can offer for the imaging and mapping of these pathways, *i.e.* water-bearing fissures, at length scales that extend from µm to cm. Three granitic rocks: Grimsel granodiorite; Kuru grey granite, and muscovite granite, were used to compare the results provided by the three techniques.

METHODS

The CLSM and PMMA techniques require the impregnation of the rock sample with epoxy or acrylic resin mixed with a fluorescent dye. Thereafter, flat surfaces must be made in the sample for further analyses without disturbing its original characteristics. CLSM offers the potential to collect a series of 2D images from the focal plane alone of the laser beam used for illumination. A stack of consecutive 2D images can then be used to construct a 3D image of the scanned volume. Figure 100 shows the principles of CLSM measurements.

The ^{14}C-PMMA method involves the impregnation of centimetre-scale rock cores with ^{14}C-labelled methylmethacrylate (^{14}C-MMA), which is then polymerised, as described in Chapter 2.4.2. Subsequent autoradiography of the planar surfaces of the sample makes possible the visualisation of the conducting pore space with a 10-µm resolution.

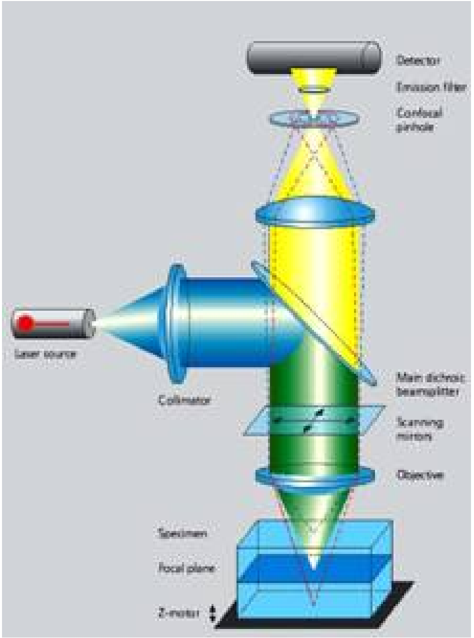

Figure 100. A pinhole conjugated to the focal plane (i.e. confocal) shields the detector from light reflected/emitted from elsewhere than the focal plane. The laser beam scans the sample sequentially, point by point and line by line, and the pixel information is assembled into a 2D image with high contrast and resolution. By moving the focus plane 2D images are gathered into a stack from which a 3D image can be constructed afterwards. ("The Confocal Laser Scanning Microscope" (2005). Carl Zeiss. www.zeiss.de/lsm).

Figure 101 shows a partition diagram for PMMA autoradiography analysis. The sawn surfaces shaded in the partition diagram were exposed to an autoradiography film.

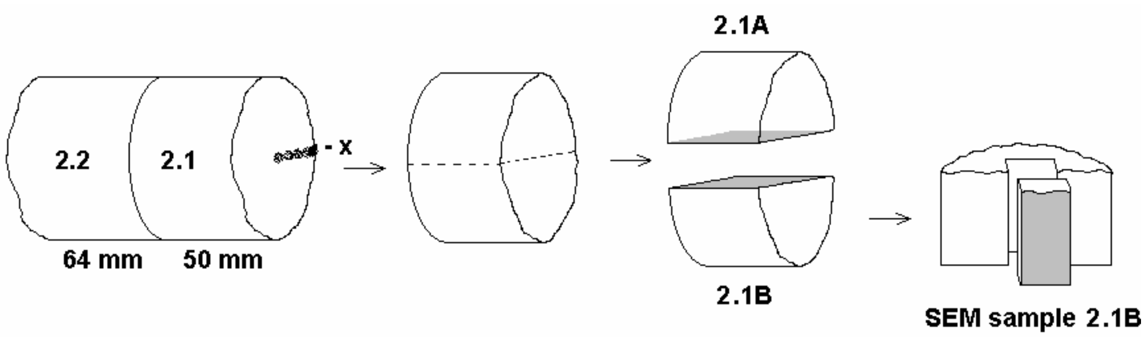

Figure 101. A partition diagram for a Grimsel granite sample with a diameter of 10 cm.

RESULTS

The main advantage of using the three techniques in parallel is the wide range of length scales they cover: the results of the ^{14}C-PMMA method are on the cm-dm scale, while the CLSM and μCT results are on the μm-mm scale and can be extended to the cm scale by lowering the resolution somewhat. The μCT method produces the best 3D information and the CLSM method can be used to depths of 200–300 μm, while the ^{14}C-PMMA method only provides 2D information on the exposed surface.

Table 15 shows the samples chosen for this study and Table 16 shows the conditions for the impregnation with the dye for each sample. Three different types of granite samples were imaged with CLSM: Grimsel granodiorite, Kuru grey granite, and muscovite granite. The CLSM studies were performed both in the Biotechnical Institute of the University of Helsinki and in the Geology Department of the University of Oviedo in Spain (a Leica TCS SP2 microscope). Better results were obtained in Oviedo, especially as it was possible to achieve imaging to depths of up to 300 μm. The equipment and image software were optimised for studying hard solid materials.

The impregnation of rocks with ^{14}C-MMA that included a dissolved fluorescent dye succeeded well. Polymerisation with radiation did not reduce the fluorescent emission of the dye, nor did the dissolved dye inhibit the MMA polymerisation inside the rock.

Table 15. Samples used to study the structure of rocks on different scales.

Sample		**Epoxy resin or PMMA /HYRL**	**Confocal CLSM /Oviedo**	**Confocal CLSM /Helsinki**	**Micro tomography /JYFL**
Kuru grey granite	**sample 1**	**x**		**x**	
Grimsel granodiorite	**sample 2**	**x**	**x**	**x**	**x**
Grimsel granodiorite	**sample 3**	**x**	**x**		
Muscovite granite	**sample 4**	**x**	**x**	**x**	
Rapakivi granite	**sample 5**	**x**			
Grimsel granite	**sample 6 (3 pieces)**				**x**

Table 16. The impregnation methods used for the samples.

	tracer	impregnation	polymerization	dye
Sample 1	^{14}C-MMA	Normal air pressure	Heating with chemical initiator BPO	Epo Dye Struers
Sample 2	^{14}C-MMA	vacuum	Radiation with Co-60	Epo Dye Struers
Sample 3	sampl-kvick acryl resin Buehler	vacuum	Two component resin, polymerizes when mixed	Epo Dye Struers
Sample 4	epoxy/ Oviedo 1993	vacuum and pressure	Two component resin, polymerizes when mixed	Rhodamine B
Sample 5	epoxy/ Oviedo 1993	vacuum and pressure	Two component resin, polymerizes when mixed	Rhodamine B

The rock samples impregnated with ^{14}C-MMA mixed with a fluorescent dye had volumes of about 100 cm^3. In this study, the PMMA porosities and the centimetre-scale connected pore structures of the granite samples were determined by means of film autoradiography. The PMMA porosities for Grimsel granodiorites varied between 0.5% and 0.8%. The PMMA porosity for Kuru grey granite was 0.3%, for muscovite granite 0.5–1.2% (altered rock caused increased porosity values), and for rapakivi granite 1%.

In Figures 102–104 photographic images of sawn and polished sample surfaces and the corresponding autoradiograms are shown. Different shades of grey in the autoradiograph represent different porosities; the darker the shade, the higher the porosity. The resolution achieved with the PMMA autoradiography method is about 10–20 µm, while in confocal microscopy it is about 1 µm. These images are from a rock sample with a size of several cubic centimetres.

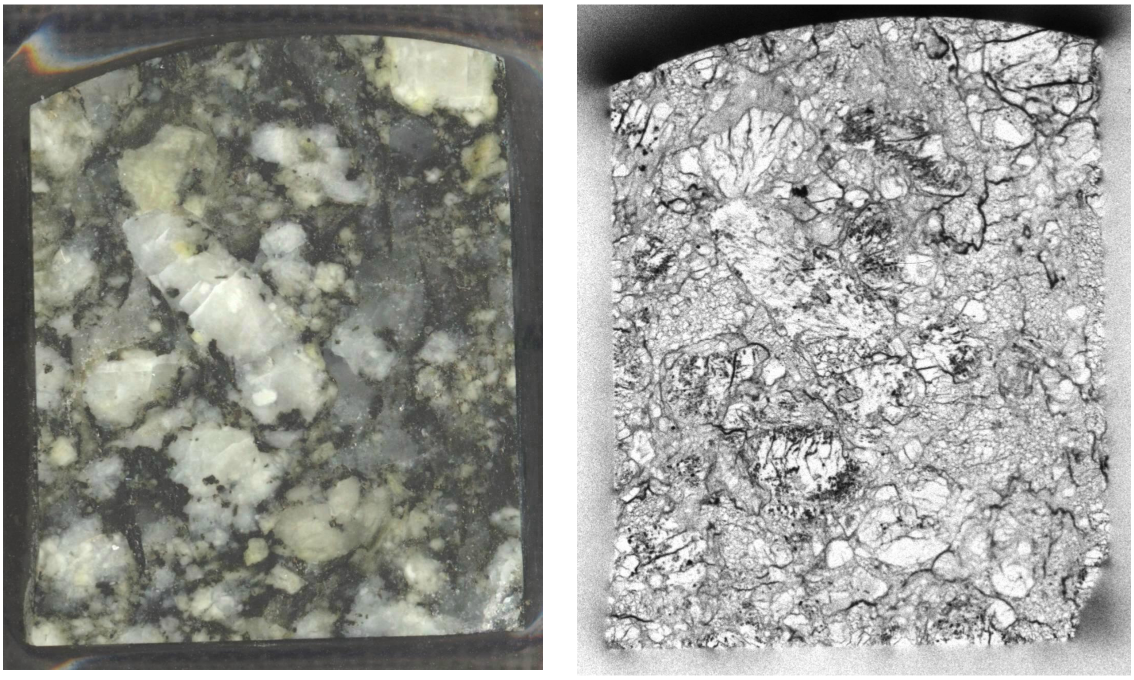

Figure 102. Photographic image of impregnated Grimsel granodiorite (left) and the corresponding autoradiogram showing the porous areas in the sample (right). PMMA porosity was 0.6%. Sample width is 3 cm.

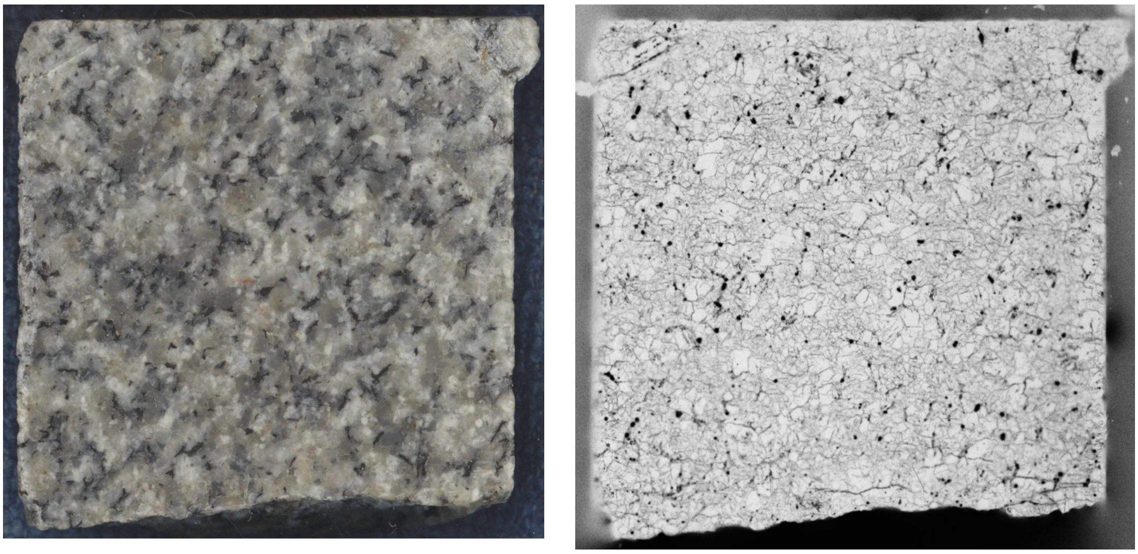

Figure 103. Photographic image of impregnated Kuru grey granite (left) and the corresponding autoradiogram showing the porous areas in the sample (right). PMMA porosity was 0.3%. Sample width is 4 cm.

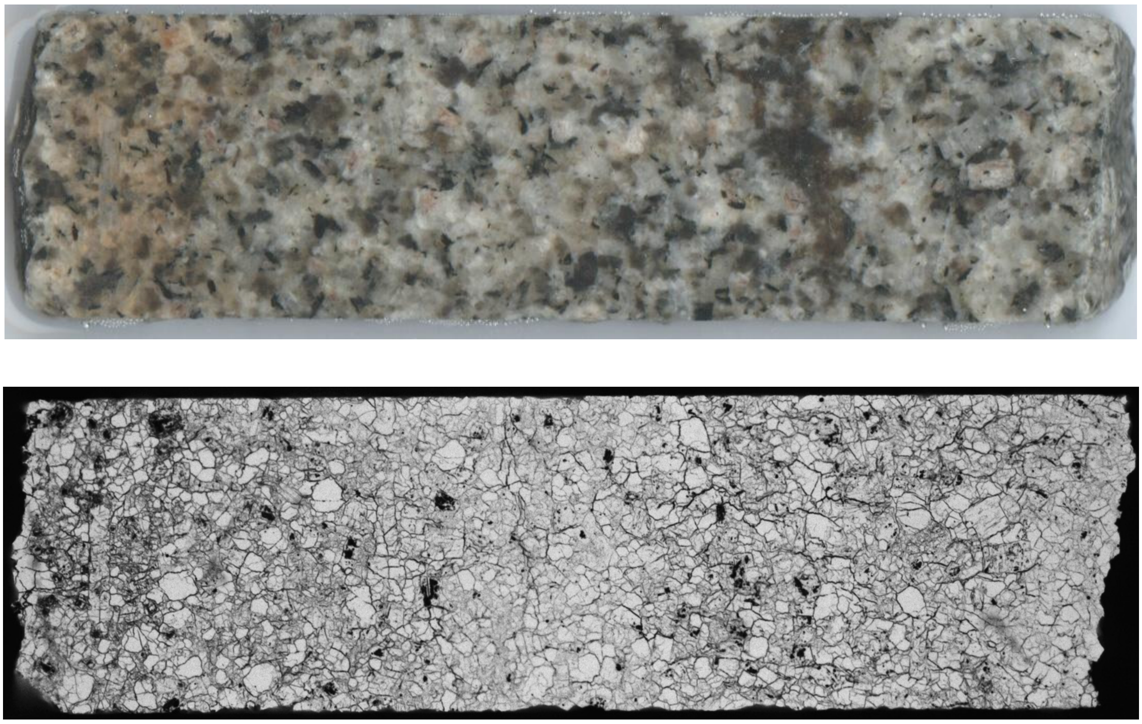

Figure 104. Photographic image of impregnated muscovite granite (upper) and the corresponding autoradiogram showing the porous areas in the sample (lower). PMMA porosity was 0.5–0.8%. Sample width is 7 cm.

The confocal microscopy evaluations of the granite samples succeeded well, except for that of Kuru grey granite. The grain boundary pores, solution pores, and microfissures had apertures below the resolution limit of this method. For Grimsel granodiorite and muscovite granite, it was possible top determine the 3D connected porosity with an accuracy of a few micrometres. It is not possible to get reliable data with CLSM if the pore apertures are clearly under 1 µm.

Figure 105 illustrates the results of the CMSL analyses. The branched microfissure can be assigned to a biotite mineral vein with apertures of 1–10 µm. In an autoradiogram this microfissure accumulation would only leave a single trace. The areas studied were located in the autoradiogram that had been cross-referenced with the mineralogy of the rock. The magnifications used in the analyses were 10x, 20x, and 40x, with the image sizes 1.5 mm x 1.5 mm, 0.75 mm x 0.75 mm, and 0.37 mm x 0.37 mm, respectively. Figure 106 shows an image of a feldspar grain.

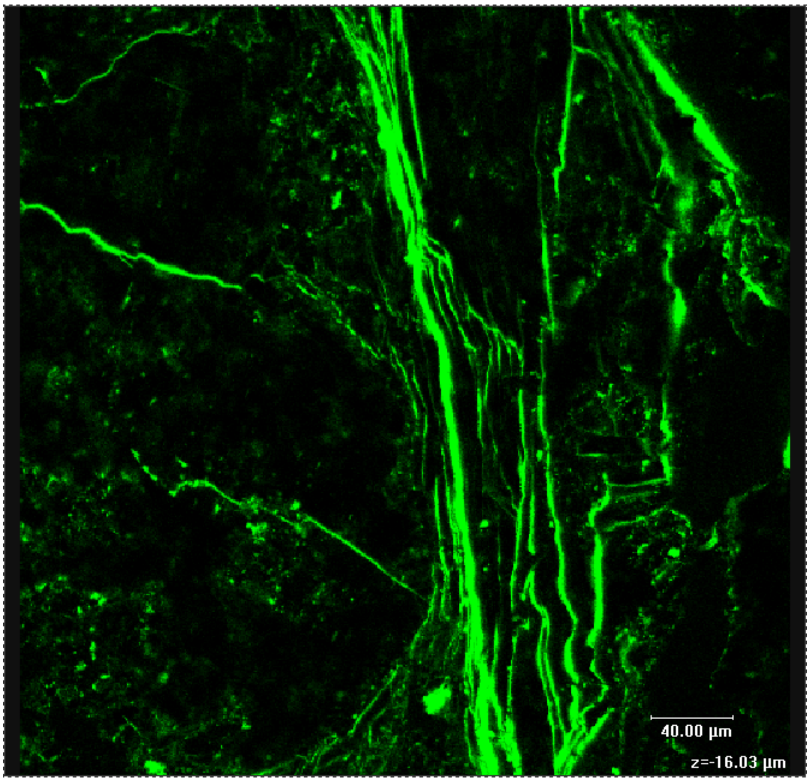

Figure 105. A 0.37 mm x 0.37-mm image from a depth of 16 µm. In an autoradiogram this microfissure accumulation would only leave a single trace.

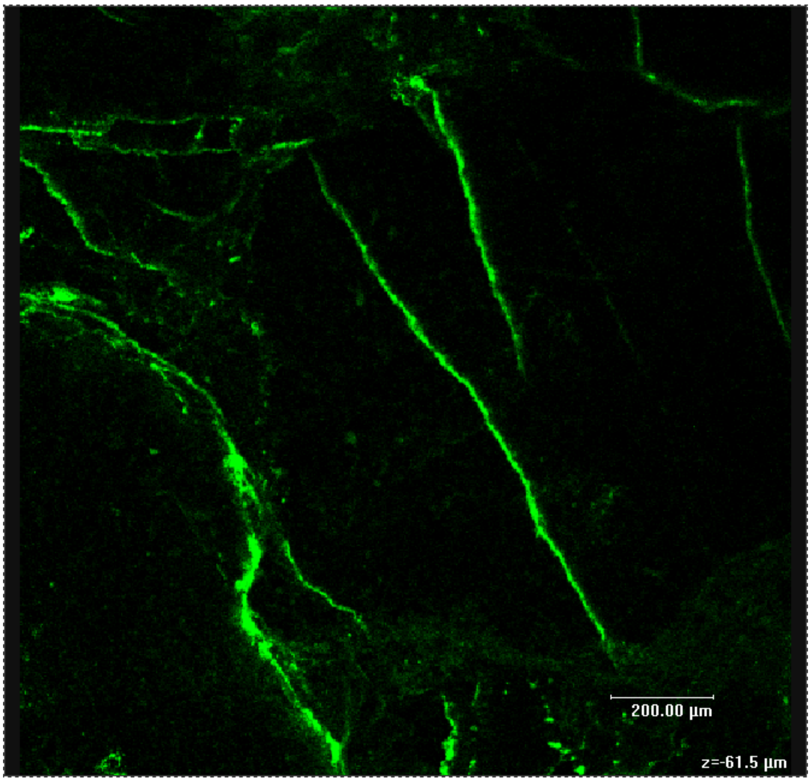

Figure 106. A 1.5 mm x 1.5 mm image from a depth of 61 µm showing intra-granular microfissures in feldspar minerals.

Figure 107 presents the pore network composed of 2D images of quartz crystals. The 3D presentation illustrates the change in the pore network depending on the depth. The sizes of the pore apertures in the crystal are 1–10 µm. In the PMMA autoradiogram the area in question is seen as a uniform dark area, because the penetration depth of the beta radiation of the radioactive carbon in rock (with 2.7 g/cm^3 density) is about 100µm.

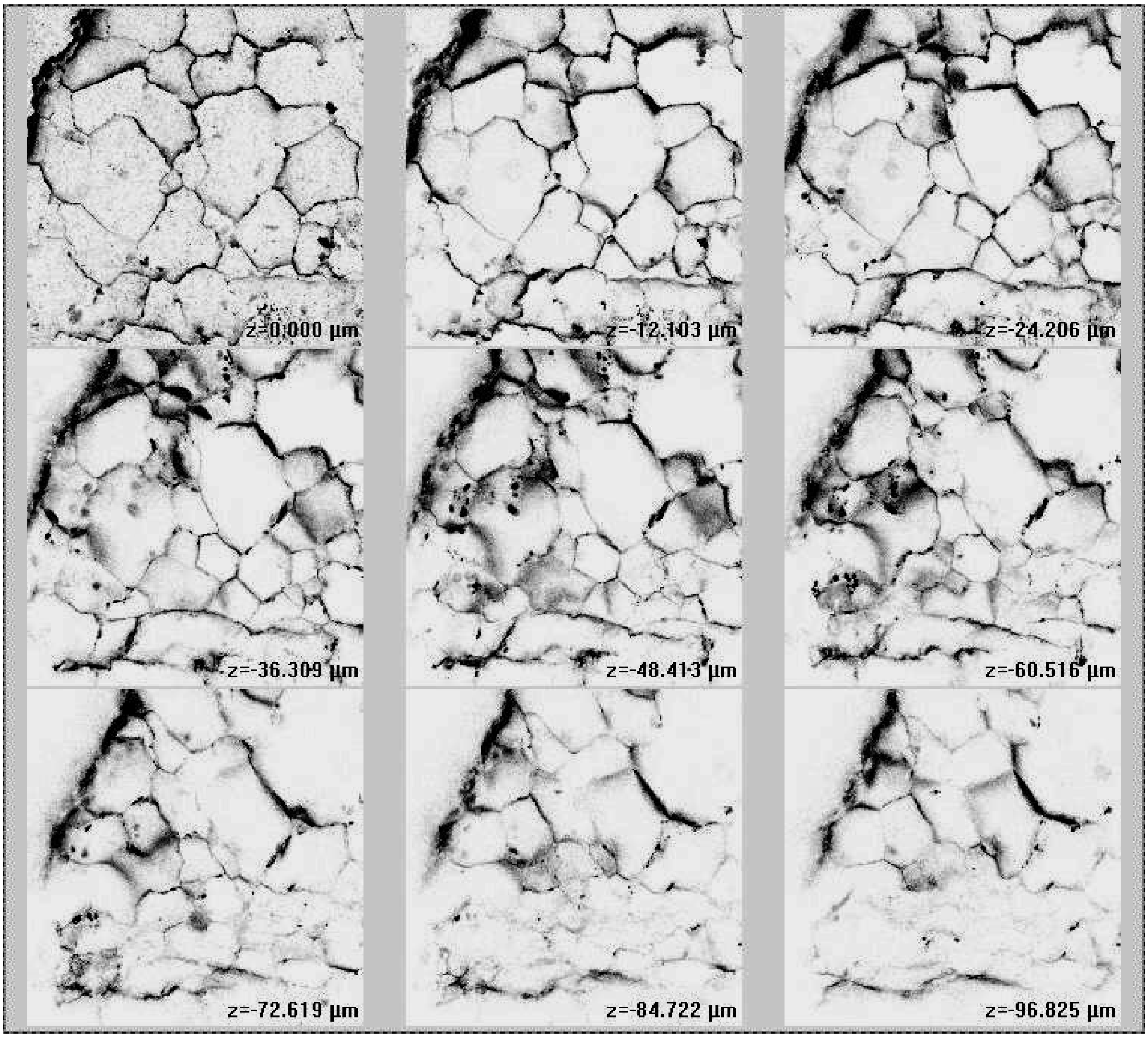

Figure 107. The structure of a quartz granule. The size of the image is 750 µm x 750 µm x 100 µm. The quartz cluster is composed of numerous mineral grains (100–200 µm in size). The grain boundary pores form a connective pore network in quartz.

The PMMA method provides a good way to characterise the centimetre-scale pore network in poorly permeable granitic rocks. It fills the gap between macroscopic and microscopic methods of investigation, and also provides some information on nanometre-scale porosity for centimetre-scale areas. However, if better understanding of the interactions between the pore structure and the pore water is important, studies with microscopic techniques such as CLSM and µCT must be included in the analyses.

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2.4.4 Studies of pore-space geometry and mineral contents with x-ray tomography[25]

INTRODUCTION

Computed tomography is an imaging method in which cross-sectional images of an object are determined from transmission or reflection data obtained by illuminating the object from different directions. Illumination can be performed e.g. by electromagnetic radiation in different frequency regions or ultrasound. The method of reconstructing a three-dimensional (3D) structure from its 2D projections was introduced as early as 1917 by Radon (see Radon 1917). Since the 1970s, x-ray CT scanners and reconstruction algorithms have been actively developed for many different applications, including geosciences (Ketcham and Carlson 2001).

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PRINCIPLES OF X-RAY TOMOGRAPHY

Computed x-ray tomography (CT) can be used to obtain a 3D construction of the internal mass distribution of the sample on the basis of the measured attenuation of x-rays. The internal structure of the sample can thus be disclosed by varying mass density. A couple of hundred 2D images of the sample at different angles are typically required for a reliable 3D structure. The principle of data collection via 2D images for the cone-beam reconstruction routine is shown in Figure 108. The sample needs to be rotated only through 180 degrees when the Feldkamp algorithm (Natterer and Wübbeling 2001) is used to reconstruct the 3D data set.

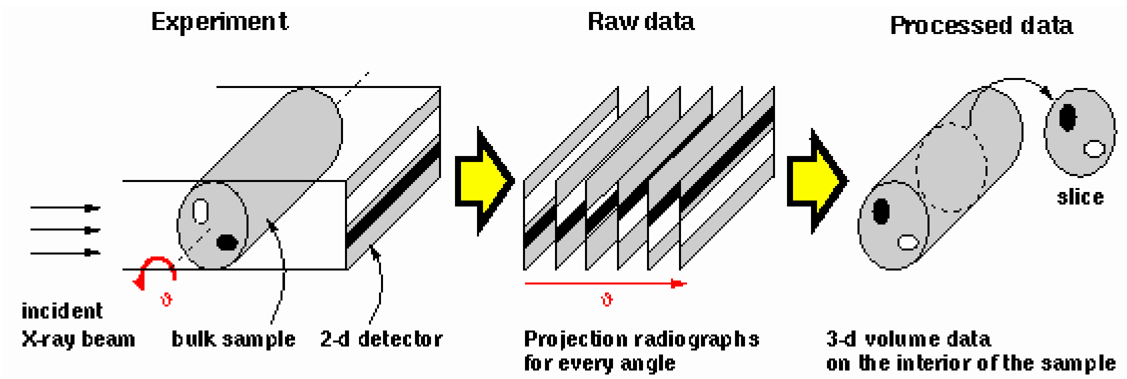

Figure 108. Principle of x-ray tomography.

In conventional CT, non-coherent x-rays are passed through the sample, and the transmitted intensity recorded is a projection of the absorption density of the sample onto the plane perpendicular to the direction of the x-ray beam. When illumination is by coherent radiation, the 2D images recorded contain information about the scattering and absorption of x-rays in the sample. Using this kind of phase-contrast imaging, one can obtain a good detail resolution, but because of the limited energy in a monochromatic and coherent x-ray beam, this method is applicable to light materials only (for instance organic materials). For conventional absorptive-contrast imaging, the reconstruction algorithms assume the pure absorption of x-rays, and the non-monochromatic x-rays are produced with an x-ray tube. Absorptive-contrast imaging is also applicable to relatively heavy materials such as minerals, with a best resolution typically of about 1 µm (microtomography).

SKYSCAN 1172 HIGH-RESOLUTION DESK-TOP MICRO-CT SYSTEM

The SkyScan 1172 micro-CT is a high-resolution desktop device based on the absorptive-contrast imaging of x-rays; see Figure 109. It includes a sealed micro-focus x-ray tube (20–100kV, 0–250µA) with a spot size smaller than 5 µm, a precision object manipulator, a 10-megapixel (4000 x 2300) 12-bit digital CCD camera, and an external computer to control both the x-ray tube and the CCD camera. An automatic filter changer is used for beam-hardening compensation and multi-energy scanning. For the reconstruction of the sample, 2D transmission x-ray images are acquired for up to 400 different angles over 180 degrees of rotation. The maximal object diameter that can be scanned is 68 mm. The pixel size at maximum magnification is 0.8 μm.

Figure 109. SkyScan 1172 micro-CT system.

A special software package has been developed for system control and sample reconstruction. The reconstruction algorithm is based on the filtered back-projection procedure for the fan-beam geometry or the Feldkamp cone-beam reconstruction for circular and spiral acquisition with specific noise reduction corrections. The software and hardware are optimised for multiprocessing.

For the preparation of samples, the microtomography laboratory must also be equipped with a high-quality diamond saw and optical microscope. A multi-processor PC cluster has been acquired for fast image reconstruction as otherwise reconstructions for the highest possible resolutions would take much too long a time. Since the image files are very big, a terabyte disc server and a fast tape robot have been installed in the laboratory as well.

FILTERING OF NOISE

Beam hardening

If a broad band beam of x-rays passes through a high-density sample, the low-energy end of the spectrum is absorbed more than the high-energy end. This effect is called beam hardening, and it means that the edges of dense materials appear brighter in the images than their inner parts. This effect can also be seen in Figure 110 below. Certain techniques used in making the shadowgrams will partly remove this effect, and they were also used in this work. The simplest way (Turpeinen et al. 2005) to remove the problems related to beam hardening which remain after the x-ray beam has been filtered is to cut the outer edges of the sample away from the image that is being analysed.

Ring artefacts

The scattering of x-rays, which is neglected in the reconstruction algorithm used, creates ring-shaped artefacts in the images produced by rotating the sample.

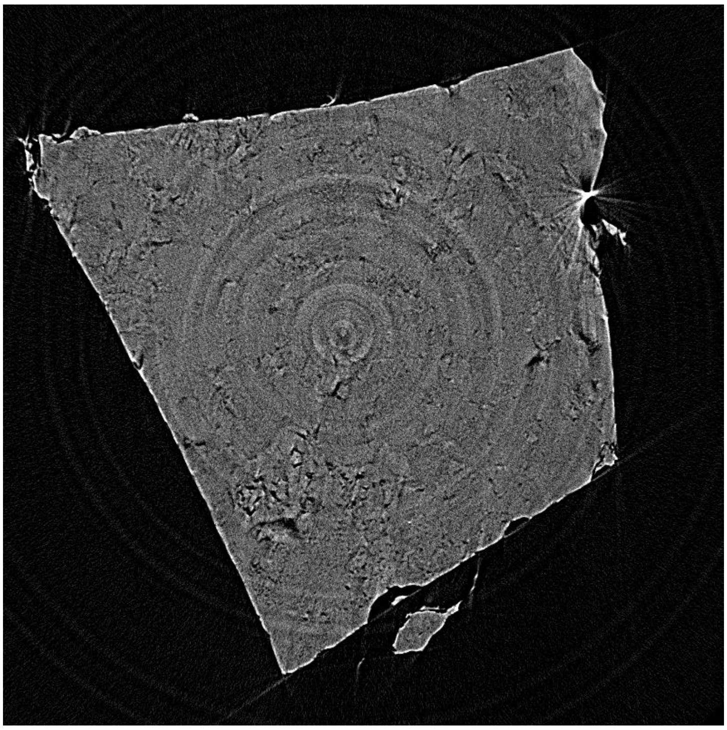

Figure 110. Ring artefacts in a mineral sample.

We applied the method of Sijbers and Postnov (Turpeinen et al. 2005) for removing homogeneous ring artefacts, which were pronounced in some of the images. We found that this method worked well for images that contain large uniform objects. In images with many small uniform objects, this method cannot satisfactorily detect artefacts as problems appear when the variance value is calculated. When the image has a lot of details, object boundaries will be mixed with ring artefacts.

As a result of this test we thought that ring artefacts could be removed more easily by transforming the image to polar coordinates. Rings can then be seen as vertical or horizontal lines and they can be parametrised more easily. Horizontal and vertical lines can be removed from the image by using high-pass and low-pass filters. We tested this method on polar-coordinate images that were corrupted by ring artefacts and the method worked quite efficiently (Figure 111). The negative side of the method is that it loses some details from the image. Such object boundaries as happened to be in the same direction as the ring artefacts were also affected when the artefacts were removed by this method.

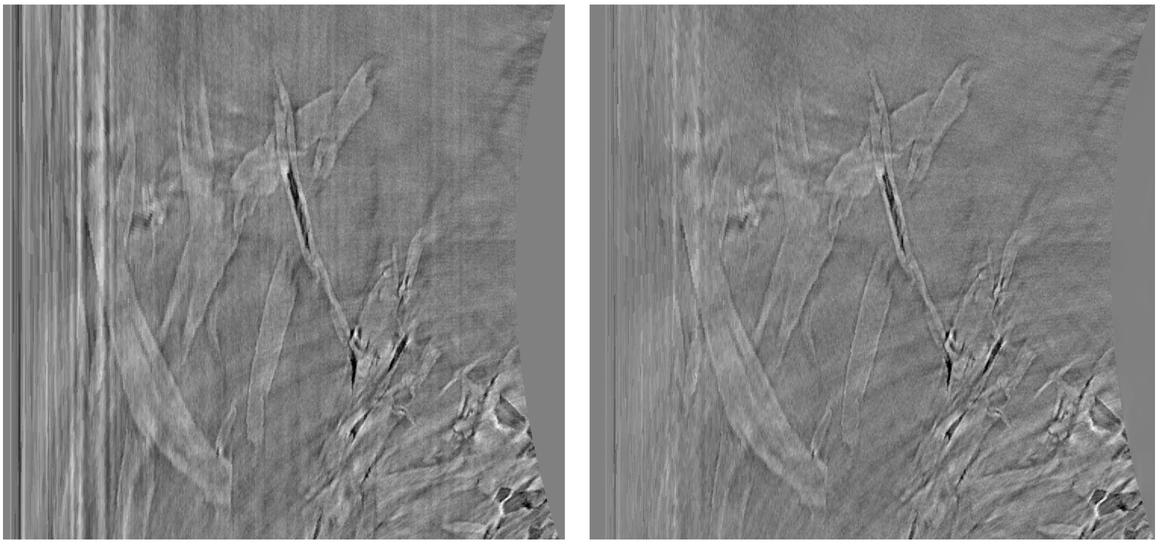

Figure 111. An unfiltered image (left) and lowpass/highpass filtered image (right).

Stochastic noise

Stochastic variations in the x-ray beam, random scattering events, and thermal noise in the electronics and CCD cells all create stochastic noise in x-ray images. This noise makes the segmentation of especially small features in the image rather difficult and its amount must be reduced before image analysis. To this end we mainly used different kinds of median filters, even though many other methods were also tested (Turpeinen et al. 2005). New robust filtering techniques will be applied in 2006.

IMAGE ANALYSIS

The main purpose of this study was to infer detailed information about the pore-space structure of rock samples by analysing in different ways the tomographic images taken of these samples. If the image quality is satisfactory, there are many properties that can be determined by image analysis. To this end we have developed a few analysis methods.

Rendering of tomographic reconstructions

Before three-dimensional tomographic reconstructions of samples can be quantitatively analysed, they must be rendered. There are two different rendering methods that are commonly used for tomographic images: isosurface rendering and direct volume rendering (Turpeinen et al. 2005). There are also two different coordinate systems used for this purpose, orthographic projection and perspective projection (Turpeinen et al. 2005). The properties of all these methods were analysed (Turpeinen et al. 2005), and both isosurface and volume rendering were used in analysing the rock samples considered.

A stack of slices can be considered as a 3D matrix containing intensity values that provide a gray-scale colour code for each voxel (volumetric pixel) in the matrix. In direct volume rendering this matrix of gray-scale values is rendered directly to the 3D space. The advantage of this method is that the image looks quite natural as every pixel can be seen as in the original data. The noise in the images is also less of a problem than it is in isosurface rendering. Noise usually appears in the images as a semitransparent veil around the object. The image can be very easily filtered to show only the areas of higher or lower density.

One can also consider a stack of slices as a 3D matrix containing the object of interest and the space around it. In isosurface rendering the boundary and the interior of the object are rendered using only one gray-scale value. In this method the gray-scale value of an individual pixel is determined by the position of the light source and the shape of the object, so that the surface points that are in shadow will be darker. The boundary (and thereby also the interior) of the object is determined by one gray-scale value (threshold limit) and gradient information of the image. This method makes voxels more opaque the closer their intensity is to the threshold and the higher their surface gradient is.

Segmentation

Segmentation is used to divide an object into its components. A rock sample can be divided into areas of different density. There are different methods for segmentation. The method to be applied depends on the quality of the data. If the boundaries of the object are clear and different objects can be distinguished in the histogram presentation, segmentation is easy.

If an image contains material with only a few different densities that are not close to each other, segmentation can be carried out using gray-scale values (Turpeinen et al. 2005) (Figure 112). One should be able to determine a gray-scale value that separates two different density values. A histogram can be helpful in this procedure. A histogram shows a peak at the gray-scale values that describe one of the components, and the threshold value can be chosen quite easily.

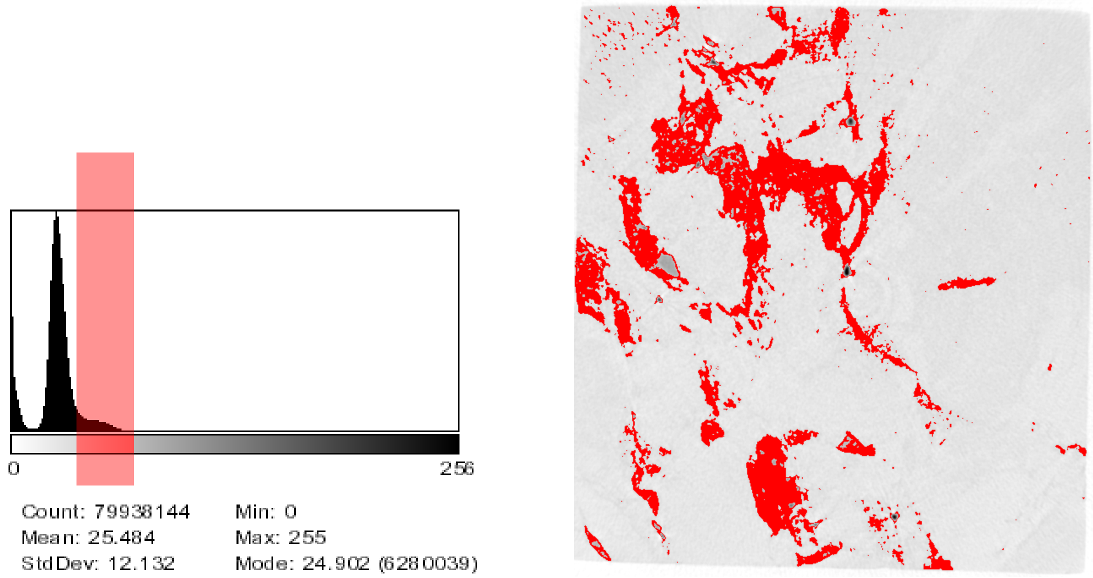

Figure 112. Histogram of a tomographic image of a rock sample and the selected density range highlighted in the original image.

When there is a need to detect an area with an edge, a line-tracing algorithm is one approach that can be used to solve the problem (Turpeinen et al. 2005). The algorithm starts at a chosen image point and moves to an adjacent point by choosing the greatest gray-scale or gradient value in its neighbourhood. As there is noise in the image, it is clear that one cannot choose the adjacent point by only choosing the point of greatest gradient in the neighbourhood. To solve this problem we decided to use an average of more than one point in each direction. Another improvement is to have forbidden angles for movement, so that backwards movement is forbidden. This method works quite well for clear boundaries. In the event that there are crossing boundaries, problems may appear. As a result of testing we concluded that a line-tracing algorithm can be used to define the boundary between air and solid material, but inside the material the algorithm is not very reliable.

Recently we have constructed a new region-growth algorithm that appears to work quite well in the inner regions of many different images. Region-growth segmentation connects voxels or subregions into larger regions according to predefined criteria. The basic approach is to start with a set of points and enlarge these into regions by appending to each point those neighbouring voxels that have the same properties (such as the grey level). We can e.g. iteratively choose the seed voxel that minimises the standard deviation of the appended region.

Beam hardening causes the edges of the object to appear brighter and the gray-scale values to become darker towards the centre. Therefore gray-scale-based segmentation is not possible. The local variance of the image can be used (Turpeinen et al. 2005) to mark the areas of low gradient. The boundaries of the object are emphasised as local variance is large near the boundaries. Local variance is calculated over a small neighbourhood around every pixel in the image. The size of the neighbourhood should be chosen according to the resolution of the image. For our images we used a 4 x 4-pixel neighbourhood. The resulting variance image can now be thresholded to outline the area of interest. As a result of testing we concluded that variance can be used to segment images that contain quite clear boundaries with small variances inside the objects. In the future we will also develop a general edge-detection algorithm.

Porosity

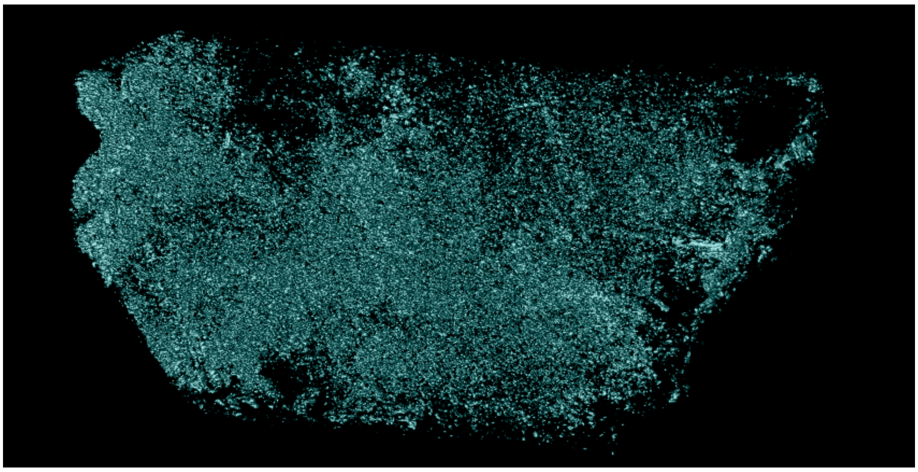

Figure 113. Pores of a granodiorite sample: porosity is 2%.

Porosity can be determined fairly easily from a 3D reconstruction of a porous sample by many software tools (Turpeinen et al. 2005). The basic steps in the porosity determination are the following. 1) threshold the image so that the pores are marked; 2) divide the intensity values of the image so that background pixels are denoted by zero and pore pixels by 1; 3) create a z-projection of the image as a sum of values. The gray-scale value of a certain pixel tells us how many pixels with the same xy-coordinates belong to a pore; 4) create a histogram of the data (number of pixels as a function of gray-scale value), and 5) determine the number of pore pixels.

The pore space structure of a porous material is typically characterised by the pore size distribution or the average pore size. Quite often, however, the concept of a pore in such a material is somewhat vague as a result of a complex pore space structure, and the void space has to be modelled with simple geometrical shapes. The pore space is often considered as a set of capillaries with varying radii, and the pore size is simply the radius of the capillary. This capillary assumption is used, e.g., in extracting the pore size distribution from the results of mercury-intrusion porosimetry.

We have implemented (Turpeinen et al. 2005) a computer algorithm to calculate the pore size distribution, the porosity, and the specific surface of a 3D binary image of a porous sample; see Figure 113. We define a pore as a local region in the pore space, and the local pore radius at a selected point as the radius of the largest sphere included in the pore space and including the selected point. We calculate the pore size distribution with a Monte-Carlo method where we select a large number of initial points randomly from the pore space, and for each point determine the local pore radius with the algorithm described above. To increase the performance we can save the information of the determined pore size in the data structure of the binary image for later retrieval. We calibrated the pore size-distribution algorithm for a set of binary images obtained by x-ray tomography, and the results were in good agreement with those measured by mercury-intrusion porosimetry.

Pore size distribution alone is not enough to characterise the permeability or diffusivity of a porous structure. A common belief is that the narrow throats between pores are most important in such transport properties. In order to analyse the throat distribution we have already constructed a skeleton algorithm on which such an analysis can be based. A 3D skeleton of the pore space is a group of connected object voxels so that for every such voxel there is a sphere centred at that voxel in such a way that one cannot find a larger sphere that would contain this sphere and include only pore voxels. There are two requirements that a skeleton has to meet. It must retain the topology and the geometry of the original pore space. The skeleton thus defines the connected centre lines of the open pores.

RESULTS FOR MINERAL DISTRIBUTION AND MICRO-FRACTURES

Parts of the samples were imaged with x-ray tomography, as explained above. The tomographic reconstructions of these samples were analysed with the methods developed. We do not report all the analysis results here but concentrate on identifying and quantifying the uranium minerals in a sample from Palmottu, and on segmenting a micro-fracture in a sample of muscovite granite. A sample from Palmottu which contains a number of uranium compounds is shown in Figure 114.

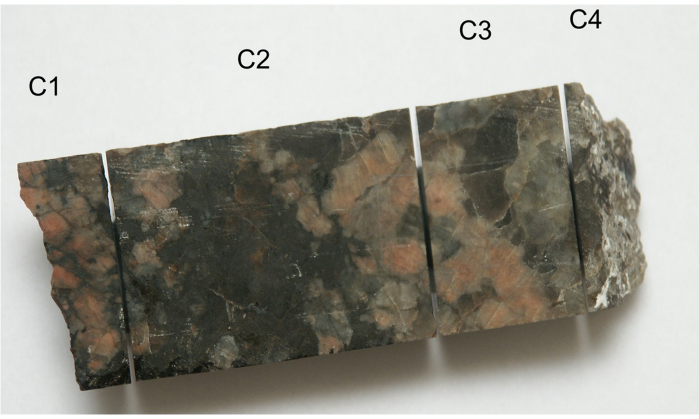

Figure 114. Sample C with subdivision from the Palmottu samples.

For the Palmottu samples the goal was first to identify the uranium compounds, which are much heavier than the other mineral components, and then determine the relative amount of these compounds. The uranium compounds can clearly be identified as dark areas in the shadowgrams of the samples. In Figure 115 we show a shadowgram of sample C4.

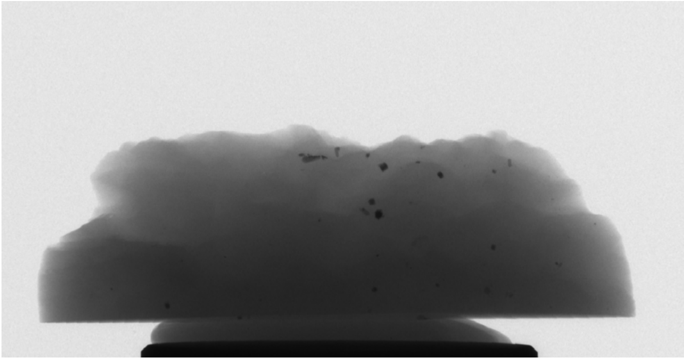

Figure 115. A shadowgram of sample C4. Uranium compounds appear as dark areas.

Shadowgrams do not, however, contain 3D information about the size of the grains of uranium minerals and for this purpose tomographic reconstructions of the samples must be made. In high-resolution imaging the sample size is restricted to about 2 mm, and the amount of uranium compounds is estimated from a relatively small amount of sub-samples of this size. In one direction these sub-samples can be longer, as it is possible to use one-dimensional scanning in the imaging of the sample. In Figure 116 we show a photograph of a sub-sample that has been scanned in one direction. Its cross-section is about 4 mm^2.

Figure 116. A sub-sample of sample C2, which has been used in tomographic imaging.

The uranium compounds of this sub-sample could be quite easily segmented by thresholding the gray-scale value, as these compounds are clearly heavier than the other mineral components. In Figure 117 we show the 3D distribution of these compounds in the sub-sample of Figure 116.

Figure 117. The 3D distribution of uranium compounds in the sub-sample of Figure 116.

Five sub-samples of samples C2 and C3 were imaged and the amount of uranium compounds in them was determined. In sample C2 their amount was found to be 1.7% and 2.3% in sample C3. It is evident from Figure 117 that the distribution of uranium compounds is very heterogeneous and they mostly appear in bands of much higher density than the average density.

Imaging of micro-fractures

For imaging micro-fractures we used a sample of muscovite granite in which the openings were found by the PMMA method and confocal microscopy to be large enough for the current resolution of x-ray tomography. Micro-fractures whose openings were larger than a couple of micrometres could be seen in the tomographic reconstructions. Because of noise, their segmentation was, however, difficult. To begin with we thus concentrated on the largest micro-fractures for which segmentation could quite easily be performed. In Figure 118 we show a 3D image of a micro-fracture that has been segmented from the mineral components by using the most advanced segmentation methods described above.

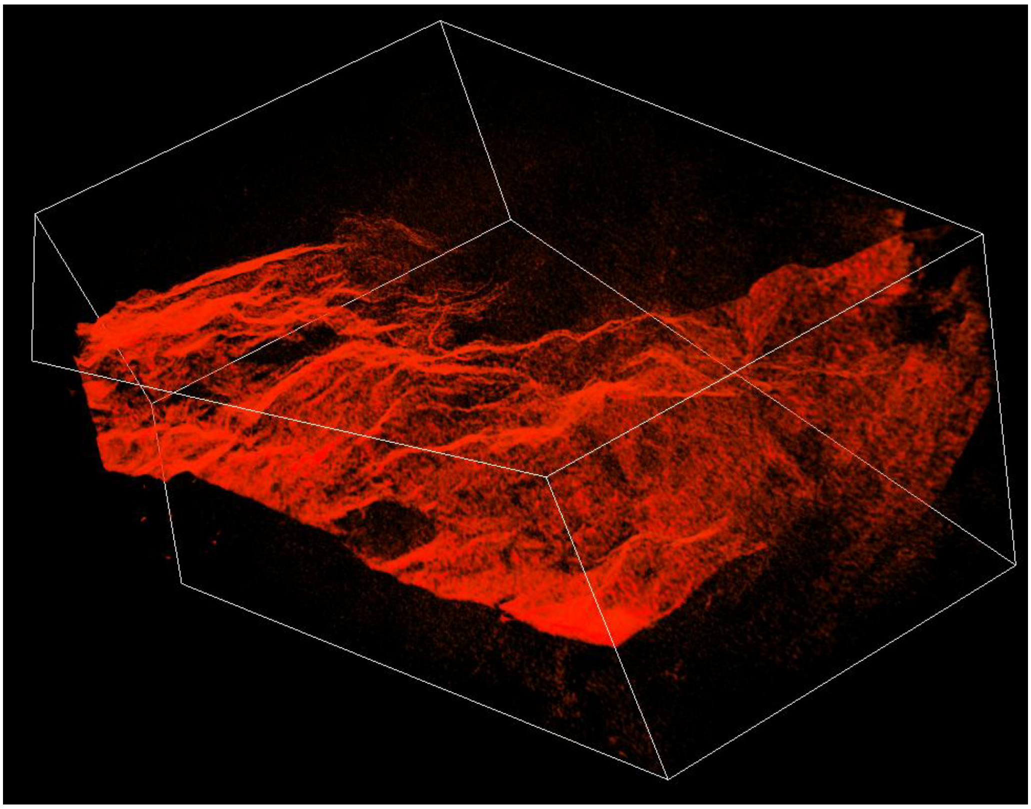

Figure 118. A 3D image of a segmented micro-fracture in muscovite granite. The sample used in imaging is shown by lines.

It is evident from this image that in this case the fracture extends across the whole sample. It partly follows grain boundaries but not always. It was possible to evaluate the tortuosity of migration pathways in this fracture from the image. We were also able to simulate the flow of water along the fracture and thereby evaluate its permeability. It would also be interesting to analyse the flow lines from which channelling effects can be determined.

It is obvious that the results reported here form only the beginning of a deeper understanding of the pore and fracture structures that appear in bedrock, possibly together with a realistic evaluation of transport properties based on fluid flow simulations in the 3D reconstructions.

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2.4.5 Sorption of radionuclides in tunnel filling materials[26]

INTRODUCTION

Sorption in mechanistic modelling and transport modelling is assumed to be a reversible process that follows the law of mass balance. Results from sorption experiments in HYRL suggest that sorption is not always fully reversible (Huitti et al. 2001). Desorption and isotopic exchange have been found to be slow or irreversible in mica gneiss, granodiorite, and rapakivi. Similar results have been reported for the Äspö rock samples in which the mica content is high (Byegård et al. 1998).

Mechanistic modelling was approached using two different methods. The first one is to assume that the rock is a multimineral composite for which mean sorption properties are determined. This is known as the general composite model (GC). Another way is to determine the sorption properties of the minerals of interest, and to derive the sorption for a known mineral composition. This is known as the component additive model. With some rocks it is sufficient to determine the properties only for the dominant sorbing mineral and take only it into account in modelling.

The major sorbing minerals in the Olkiluoto rocks are micas, biotite, muscovite, and hornblende. Cordierite and chlorite can also be important if present. In the Olkiluoto rocks the most important sorbing mineral is biotite, because of its sorption capacity and abundance. Thus the component additive approach can be considered. In this work the sorption of Cs on biotite was studied in brackish and saline ground waters and simple electrolyte solutions. The aim is to develop a mechanistic model to describe the sorption of Cs onto Finnish granitic rocks.

[26] By Jarkko Kyllönen, HYRL.

EXPERIMENTS

Batch sorption experiments were performed on biotite that had been separated from drill cores from the Olkiluoto boreholes from depths of 200–500 m, Table 17. The core was crushed and biotite was separated from the sample by heavy liquid separation and magnetic separators. The samples were characterised with XRD and were found to be slightly altered. The major alteration product was chlorite. The chlorite content was analysed by means of the point counting method and the chemical composition was analysed with a microanalyser. The specific areas were determined by N_2-BET. An unaltered pegmatitic biotite was used as a reference material. Cation exchange capacities were determined with standard methods.

Table 17. The properties of the biotite samples.

Sample	KR2	KR9
Particle size	< 0.5 mm	< 0.5 mm
Surface-area (BET)	0.64 m^2/g	0.83 m^2/g
CEC/AgTU	0.013 meq/g	0.013 meq/g
CEC/NH_4Ac	0.022 meq/g	0.018 meq/g
Biotite	94%	90%
Chlorite	6%	10%

Cs sorption capacities were determined in Allard ground water simulant as a function of Cs concentration. The mass balance of sorption was studied with NaCl, KCl, $MgCl_2$, and $CaCl_2$ with and without radioactive tracers. Ionic strengths were between 1 x 10^{-1} and 1 x 10^{-4} M. An atomic absorption spectrometer (AAS) was used to analyse the solutions of the inactive experiments.

Biotite KR9 was equilibrated with NaCl, KCl, and $CaCl_2$ in a column setup to occupy all cation exchange sites with a single element. Equilibration solutions were sampled and analysed with an inductively coupled plasma mass spectrometer (ICP-MS). Cs sorption experiments were performed on equilibrated biotites in order to determine selectivity coefficients in a binary system. These experiments were also performed with and without tracers. The inactive solutions were analysed with ICP-MS. The sorption isotherms are shown in Figures 119–121.

The experimental data are used to determine selectivity coefficients for the cation exchange reactions of Na, K, and Ca to Cs. These selectivity coefficients are then normalised to give selectivity coefficients for the cation exchange reactions of K, Ca, Cs to Na. These are used to model the sorption of Cs to biotite using Phreeqc (Appelo &

Parkhurst 2002). Batch reactions are modelled using a simple cation exchange model. It was found that at least two different cation exchange sites are needed to model the behaviour of Cs. One site with high capacity and a relatively low selectivity coefficient and another site with low capacity and high selectivity are needed. This is in good accordance with the literature; see, for example, Steefel et al. (2003), Bradbury & Baeyens (2000). Selectivity coefficients according to the Gaines and Thomas convention for the cation exchange of Na, K, and Ca to trace level Cs are presented in Table 18.

Table 18. Selectivity coefficients for Na, Ca, and K to Cs cation exchange reactions at trace Cs concentrations.

Reaction	**logK**
Na to Cs	3.8
Ca to Cs	8.3
K to Cs	1.6

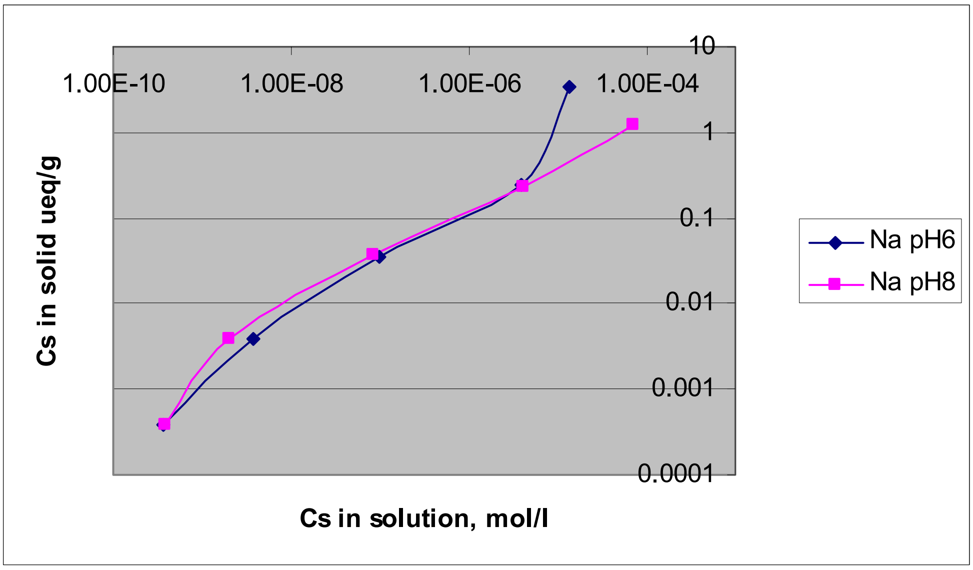

Figure 119. Sorption isotherm of Cs in 0.1 M NaCl, pH-buffered to 6 and 8.

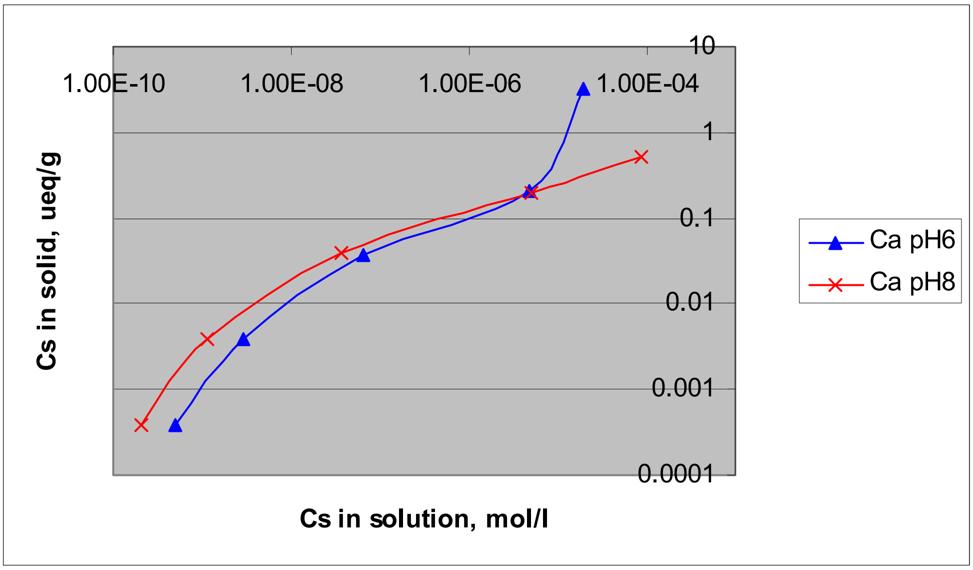

Figure 120. Sorption isotherm of Cs in 0.1 M $CaCl_2$, pH-buffered to 6 and 8.

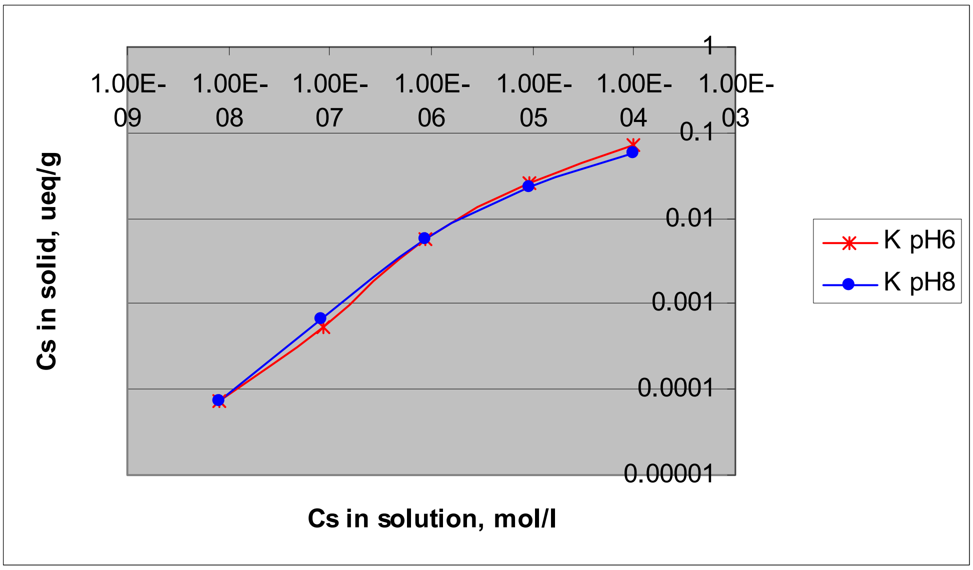

Figure 121. Sorption isotherm of Cs in 0.1 M KCl, pH-buffered to 6 and 8.

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2.4.6 NEA TDB and NEA Sorption Forum[27]

NEA TDB

A Thermodynamic Data Base project (NEA/TDB) was started about 20 years ago (1984) under the auspices of OECD/NEA. Finland has been involved in NEA/TDB since the very beginning. All the project phases have been joint undertakings of several international nuclear organisations all along. The current third phase (TDB III 2003–2006) of the project ends in February 2006 and has been financed and managed by 13 organisations, one of which has been Posiva Oy. In a later phase of TDB III two more organisations joined in. The KYT programme participated in the project during 2002–2003.

The importance of the TDB project is closely connected to the assessment of the safety of a radioactive waste repository as it is essential to be able to calculate the eventual radionuclide migration into the environment. Numerical simulation and/or the modelling of processes affecting the behaviour of radionuclides in various systems are data needed in the safety assessment work. The purpose of the TDB project has been to make available a comprehensive, internally consistent, internationally recognised, and quality-assured chemical thermodynamic database of selected chemical elements. This database should meet the specialised modelling requirements for the safety assessments of radioactive waste disposal systems. Nine volumes of thermodynamic data have been published thus far on compounds and complexes of U, Np, Pu, Am, Tc, Se, Ni, and Zr with selected organic ligands (oxalate, citrate, EDTA, isosaccharinic acid). The most recently published volumes are on Zr (Vol. 8 in 2005) and Volume 9 (in 2005). TDB III should still finalise data on Th, Sn, and Fe. Additionally, a group of experts has been

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reviewing the literature in order to find out whether scientific guidelines for the evaluation of thermodynamic data for solid solutions could be assigned.

NEA SORPTION FORUM

Following Phase I of the NEA Sorption Project (1997–1999), Phase II was started and the final workshop of the project was held in October 2005. Phase I was directed towards the demonstration of the capabilities of thermodynamic sorption modelling for guiding radioelement distribution coefficient investigations. Phase II was a comparative exercise, with the aim of interpreting well-characterised datasets for radionuclide sorption using different modelling approaches. The test cases and reporting of the results (NEA 2005) were carried out by a technical direction team (TDT) and a management board (MB). The Finnish participants were Martti Hakanen (University of Helsinki, MB), Markus Olin (VTT, TDT and modelling team), and Jarmo Lehikoinen (VTT, modelling team). The funding for the Finnish participation came from Posiva Oy.

PHASE 2 OF THE NEA'S SORPTION MODELLING FORUM – MEMBERSHIP OF THE PROJECT'S TECHNICAL DIRECTION TEAM AND PERFORMANCE OF THREE TEST CASES

General

Phase 2 was organised as both a model comparison and a modelling demonstration project. Twenty modelling teams from twelve countries were selected by the participating NEA agencies to submit modelling results for the project. Altogether seven test cases, divided into three classes, were prepared and sent to the modelling teams:

Group 1 – Simple substrates: Single oxide minerals

TC1: neptunium (V) sorption on hematite under the influence of carbonate.

TC2: selenium (VI) and selenium (IV) sorption on goethite.

TC3: uranium (VI) sorption on quartz under the influence of carbonate (two studies).

Group 2 – Substrates of intermediate complexity: clay minerals and bentonite

TC4: nickel sorption on montmorillonite and natural bentonite under varying conditions (four studies).

TC5: neptunium (V) sorption on montmorillonite under the influence of carbonate (two studies).

Group 3 – Complex substrates: soils and sediments

TC6: uranium sorption on weathered schist materials from the Koongarra deposit in Northern Australia.

TC7: cobalt sorption on subsurface materials including separated mineral components from South Carolina, USA, including the effects of humic substances.

Technical direction team (TDT)

A technical direction team (TDT) was appointed to evaluate the existing database, develop test cases for sorption modelling, and carry out the subsequent analysis and interpretation of modelling outcomes. VTT's M. Olin was a member of the TDT, which worked from the year 2000 to 2004, had four meetings (in Paris, San Francisco, Helsinki and Zürich), and arranged a 3-day workshop held in October 2002 in San Lorenzo de El Escorial, Spain, involving modelling teams from every participating organisation. The final report of the project was published in 2005 (Davis et al. 2005), and the final meeting for Phase 2 was held in October 2005 in Paris.

Test cases

VTT (J. Lehikoinen & M. Olin) was one of the twenty modelling teams and, of seven test cases, VTT participated in three: TC1, TC4, and TC5. For the modelling of the test cases, the TDT recommended a working time of about two weeks per test case, which aimed to test the possibility of doing good enough work with a cost level still at an acceptable level – for example, to cost-effectively support PA work. VTT's three test cases were modelled according to this guideline, and the model results were completely comparable to other groups' work, the results of which were presented at the El Escorial meeting in autumn 2003, although it was obvious that at least some groups had expended much more effort on the modelling. Instead of the increased amount of fitting per test case, a more fruitful approach might be a database of surface reactions, electrostatic models, and corresponding parameter values, similarly to aqueous chemistry. VTT's model results sent to the TDT for final analysis were published in Davis et al. (2005).

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2.5 Biosphere studies

Biosphere studies have an important role in safety case considerations as the safety criteria for the first few thousand years are based on dose limits. In addition to human radiation protection, the protection of flora and fauna is also necessary. Within the KYT programme, biosphere studies were focused on the radiation protection of life forms other than human, carried out in the form of international collaboration.

2.5.1 Environmental radiation protection[28]

At present the risk assessment and management of radionuclides entering the environment are generally based on human health considerations alone. However, in the

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last ten years there has been increasing awareness of the need to be able to demonstrate the protection of fauna and flora against ionising radiation. Several international, regional, and national programmes, notably those within the IAEA, ICRP, and EU, are involved in the development of standards and criteria, assessment methods, and managerial approaches for protection against environmental radiation.

The Finnish Nuclear Act includes a general provision that it has to be ensured that nuclear energy is safe for humanity and the environment. Furthermore, the Government Decision on the safe disposal of spent fuel provides that the potential impacts of disposal on species of flora and fauna are to be examined. The provision is further elaborated in Guide YVL 8.4. on the long-term safety of spent nuclear fuel with the statement that the disposal of spent fuel should not affect species of flora and fauna detrimentally. This is to be demonstrated by assessing typical radiation exposures for terrestrial and aquatic populations in the disposal environment and to compare them with the levels of exposure that, according to prevailing best scientific knowledge, would not cause a decline in biodiversity or other significant detriment to any living population.

As the current regulatory framework for the safe disposal of spent fuel provides for dose assessment, both the operator and the regulator have to develop their skills in the field. A dose calculation tool, MHDC, based on advanced Monte Carlo simulations, was earlier developed in the Research Department of the STUK. During this reporting period the tool was tested by comparing it with well-documented cases found in the literature. The results were in good agreement with published cases. The greatest differences were found with high energies and very small species, but there too the deviation was less that 50%. The tool was also applied to a new case by calculating doses for perch in a Finnish lake. The results were published in Oksanen et al. (2003).

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3. Strategic alternatives in spent fuel management[29]

The study of strategic alternatives in spent fuel management is necessary, although Finland is purposefully following the guidelines in the Decision-in-Principle (DiP) for the disposal facility at Olkiluoto. There is a need to have knowledge of the state of the art in nuclear waste management technology outside the domain of direct geological disposal that is the Finnish approach. Additionally, there is a need to keep abreast of the international discussion. This strategic knowledge will be of the utmost importance if the current spent fuel management plans cannot, for some reason, be realised.

INTRODUCTION

There are no real alternatives to the disposal of high-level nuclear waste or spent nuclear fuel elements in safe geological repositories. However, the long-term storage before the disposal and, especially, advanced fuel cycles with effective partitioning and transmutation (P&T) may make it easier to fulfil the long-term safety requirements and especially to decrease the required size of a repository. As a consequence these options may have an impact on the waste management strategy (Anttila et al. 1999).

The long-term storage option has not been studied in the KYT research programme. Therefore, in this chapter only the impact of advanced fuel cycles on nuclear waste management will be discussed. The text is based mainly on research carried out in the project "Suomalainen erotus- ja transmutaatiotekniikan tutkimus (Finnish Research on Partitioning and Transmutation)"; see Annex A. Detailed discussions of advanced fuel cycles and especially of P&T can be found, for instance, in the references (NEA 2002, Ahlström et al. 2004, and IAEA 2005).

3.1 Advanced nuclear fuel cycles and waste management

3.1.1 Classification of the nuclear fuel cycles

Nuclear fuel cycles can be divided into three main classes, which can be called open, partly closed, and fully closed cycles.

An open fuel cycle is based on natural or low-enriched uranium. The spent nuclear fuel is first stored for a few dozen years and is then disposed of in a repository.

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In partly closed fuel cycles the uranium-based spent fuel is reprocessed, i.e. the spent fuel is chemically partitioned in a few separate product streams. In a well-established PUREX process there are three product streams: uranium, plutonium, and high-level waste (HLW), which contains fission products and minor actinides (neptunium, americium, and curium). The streams are very pure. Only about 0.1–0.2% of uranium and plutonium goes into HLW. The basic PUREX process could be modified or extended quite easily. As a consequence, there are several partly closed fuel cycle options.

A fully closed fuel cycle would utilise the basic fuel material (uranium and later maybe also thorium) as effectively as possible. Therefore, all the spent fuel would be reprocessed and reused to such an extent as is reasonable from the economic point of view. Minor actinides and even some fission products might be mixed with uranium and plutonium in the fuel fabrication process. The other option is to burn them either as separate targets in normal reactors or in special transmutation plants.

The simplest fully closed fuel cycle would be one based on advanced fast reactors, which could use fuel elements made up of all the actinides, separated and recycled as a group in a reprocessing plant. In this Group Actinide Extraction (GANEX) process (CEA 2005) the fission products would be disposed of either as such or separated into two or more streams. New (depleted or natural) uranium would be added to the fuel cycle in order to keep the inventory of fissionable nuclides at a high enough level.

3.1.2 Partitioning and transmutation

Partitioning refers to any of the chemical and/or metallurgical technologies which are used to separate various element groups from the high-level nuclear waste stream. Transmutation is elementary nuclear conversion through single neutron absorption.

When used as a unified term, partitioning and transmutation (P&T) can be defined as a process, "(the) main objective of (which) is to eliminate or at least substantially reduce the amount of such long-lived radionuclides that has to go to a deep geological repository for final disposal" (Ahlström et al. 2004). This definition relates P&T closely to nuclear waste management. In fact, the term is often restricted to mean only those special facilities, e.g. accelerator-driven systems (ADS), which, more or less outside the normal fuel cycles, process and burn (mainly) minor actinides and maybe some long-lived fission products (at present Tc-99 and I-129 seem to be the only fission products feasible for transmutation).

However, the definition given above can also be interpreted in such a way that all partially and fully closed fuel cycles are P&T processes, because they reduce the amount of actinides in high-level waste and spent fuel. At present, it seems obvious that in the future P&T will be an integral part of advanced fuel cycles. The construction of special P&T facilities further into the future cannot be ruled out.

The partitioning of spent nuclear fuel or high-level waste could also be carried out without subsequent transmutation. It could be used to make possible optimised treatment for each separated element group. For instance, the storage and disposal of caesium and strontium separately would decrease the thermal load of HLW appreciably during the first 100 years of cooling. Curium is also a good candidate for special treatment, because after about 100 years most of it will have decayed into various plutonium isotopes. Partitioning followed by transmutation and/or conditioning (P&T/C) may be a more accurate description than P&T for the present orientation of R&D regarding advanced fuel cycles.

3.1.3 Research and development of advanced fuel cycles

In general, the more advanced a nuclear fuel cycle is, the more sophisticated some or all of its phases will be. To achieve a fully closed nuclear fuel cycle that is both economically competitive and safe, many problems remain to be solved (DoE/SC 2005). There is a clear need for more accurate and comprehensive nuclear data and computer codes to be used in the simulations of advanced reactors. However, the greatest obstacles may be related to the development of durable enough fuel and structure materials, as well as effective reprocessing methods for high-burnup and very radioactive fuel elements. All the fuel cycle facilities, especially the reactors, should be cheap enough to build, maintain, and dismantle.

The research and development of advanced reactors and fuel cycles is a very costly effort. New research facilities, which are needed to test and compare various options, are expensive to construct and experimental work as a whole demands large funding, too. Therefore, the national funding should be adequate and international cooperation should be organised in an effective way. All relevant research areas should be covered adequately and the duplication of work should be avoided.

At present, the United States (AFCI 2005), Japan, and France are the leading countries regarding R&D work on advanced reactors and fuel cycles, but China, India, and South Korea are allocating more and more resources to this field. Each country has its own priorities, which may complicate international co-operation. However, the establishment of the commercially-oriented Generation IV International Forum (GIF) and the more research-oriented INPRO project organised by the International Atomic Energy Agency (IAEA) are good examples of recent efforts to gather human and financial resources together. Several other multi- and bilateral co-operation projects have also been started.

In the European context, research carried out within the framework programmes of the European Union is important to the member counties, assuming that the national funding is large enough for meaningful participation. However, up till now, very little funding has been devoted to studies on advanced reactors and fuel cycles, the only exception being R&D on Partitioning and Transmutation (P&T), which will be discussed in more detail later. However, the European Union is now officially a partner of GIF, which may lead to increased funding for these kinds of studies.

From the Finnish point of view the Nuclear Energy Agency (NEA) of the Organisation for Economic Cooperation and Development (OECD) has played an important role as a source of knowledge on research and development on P&T and, in general, on advanced fuel cycles. The NEA working parties and expert groups have also offered an opportunity for limited cooperation with experts from other member countries.

In many countries, for instance in Sweden and the United States of America, R&D on P&T and advanced fuel cycles are also funded in order to train new nuclear experts.

3.1.4 European R&D on partitioning and transmutation

The European Union had already started to provide financial support for research and development on P&T at the beginning of the 1990s. However, within the third and fourth Framework programmes the funding was quite small. In 1998, the research ministries of France, Italy, and Spain decided to establish an advisory group on the use of accelerator-driven systems (ADS) for nuclear waste management. Later, Austria, Belgium, Finland, Germany, Portugal, Sweden, and the EU, through the Joint Research Centre, joined the group. In practice, the so-called technical working group (TWG) carried out the main work and produced a roadmap for an ADS demonstration programme to be performed in 12 years (see Ahlström et al. 2004, pp. 38–44). To a large extent, the recommendations of the TWG still guide the EU research and development on P&T.

Within the fifth framework programme the EU funded P&T research and development with 29 million euros. Thirteen projects were carried out within the so-called ADOPT (Advanced Options on P&T) network, which had 16 partners. The research projects were divided into four clusters: partitioning, fuel and transmutation, technological support, and basic (data) studies. The largest project of ADOPT, preliminary design studies for an experimental ADS (PDS-XADS), was not included into any cluster. The ADOPT projects have produced a large number of articles, but their main reports are not yet available for scientists in countries outside the network.

The EU-funded P&T research and development work continues within the sixth framework programme, almost as before. However, it was decided to establish only two large integrated projects, one for partitioning, EUROPART, and the other for transmutation, EUROTRANS. There is also a smaller research project called RED-IMPACT, the aim of which is to study the impact of P&T and advanced fuel cycles on waste management. The total EU funding for these projects is about 30 million euros.

Effective participation in the EU projects is possible only if the national funding is adequate. Therefore, Finland has not been able to participate in the European P&T research, even if Finnish experts have been involved in the preparatory phases of some projects. To some extent, limited access to the results of that work can be achieved, for instance though OECD/NEA committees and other personal relations.

3.2 NEA studies on P&T and advanced fuel cycles

3.2.1 NEA studies on scientific issues of P&T

OECD/NEA has already conducted several studies on advanced nuclear fuel cycles, including P&T systems, during the last 10–15 years. The NEA Nuclear Science Committee (NSC) has been responsible for scientific aspects of the subject, while the NEA Nuclear Development Committee has organised so-called system studies, which have been aimed at comparing various reactor or fuel cycle options with each other. The committees have always cooperated quite efficiently.

After relatively long consideration, the NEA Nuclear Science Committee established a Working Party on Scientific Issues in Partitioning and Transmutation (WPTT) in June 2000. About ten member countries decided to participate actively in the work of the new group. Some other countries, including Finland, chose to contribute only in a rather restricted way.

The WPTT itself established four subgroups, which covered the main fields of P&T: accelerator utilisation and reliability; chemical partitioning; fuels and materials, and the physics of transmutation systems. The main goal of each group was to establish a network of experts which would be responsible for writing a state-of-the-art (SOA) report and also for arranging scientific meetings.

The original work plan of the WPTT was very ambitious and the subgroups wanted to enlarge the scope of their studies. As a result, the Working Party could not finish its work during its original three-year mandate (2001–2004). After a one-year extension, the NSC decided to replace the WPTT with a new Working Party on Scientific Issues of

Fuel Cycle (WPFC) and to move the transmutation physics to another new Working Party (WP on Scientific Issues of Reactor Systems, WPRS). The new groups should take care of the completion of the WPTT. Up till now, three SOA reports have been published (NEA 2005a, NEA 2005b, and NEA 2005c).

Finland participated mainly in the work of the transmutation physics subgroup of the WPTT. The so-called MUSE benchmark, based on measurements at the French MASURCA experimental facility, was solved successfully in a very short period of time (Leppänen 2002 and 2003).

3.2.2 NEA system studies on advanced nuclear fuel cycles

The NEA Nuclear Development committee has now completed three main system studies, the main goal of which was the assessment of the pros and cons of partitioning and transmutation. In each case an expert group consisting of national experts and representatives of IAEA and EC was established. More than ten countries participated in the studies. Finland was involved in each phase, even if the Finnish contribution was always rather limited as a result of the lack of resources.

The first system study (NEA 1999) was carried out in 1996–1998. It was a review of progress in the separation of long-lived actinides and fission products, the various options for transmutation, and the potential benefits of P&T for waste management. However, some important issues, for instance fully closed fuel cycles and accelerator-driven systems (ADS), were not addressed.

The second NDC P&T system study in 2000–2002 (NEA 2002) focused on the roles and relative merits of fast critical reactors and accelerator-driven subcritical systems (ADS) in advanced fuel cycles. For that purpose several fuel cycle schemes were specified and analysed in great detail. The mass flows, including waste streams, were calculated or estimated for each case. The status of relevant technologies was also discussed in a very detailed way.

NDC started its third P&T system study in 2003. The NEA Radioactive Waste Management Committee (RWMC) decided to support the study, because the focus of the study was to be on nuclear waste management issues.

The mandate of the expert group defined the following three goals for the study:

- assessing the characteristics of radioactive waste arising from advanced nuclear fuel cycle options,

- assessing the performance of repositories using source terms for waste from advanced fuel cycles,
- identifying new options for waste management and disposal.

The expert group, consisting of experts from twelve countries and the two above-mentioned international organisations, started its work by defining ten fuel cycles (some with a variant), divided into three classes as follows:

- Cycles based on current industrial technology and its extensions
    - open fuel cycle (the reference case)
    - one-time recycling of plutonium in a pressurised water reactor (PWR)
    - one-time recycling of plutonium and neptunium in a PWR
    - DUPIC cycle (use of processed PWR spent fuel in a Candu reactor)
- Partially closed fuel cycles
    - multiple recycling of plutonium in a PWR
    - multiple recycling of plutonium and americium in a PWR
    - multiple recycling of plutonium and americium in a PWR and a fast reactor (FR)
- Fully closed fuel cycles
    - multiple recycling of transuranics in a FR
    - all actinides burnt in the so-called double strata system with an ADS
    - recycling of all actinides in a FR.

For the advanced fuel cycles several fuel and irradiation target types, as well as many reprocessing / partitioning techniques, were assumed. Some of the systems to be assessed were very complicated and many of them are still in an early phase of development. Even if the NDC group was able to get valuable data on the waste streams of advanced fuel cycles from an NSC group of reprocessing experts and other outside help, some of its input data are still only indicative. With this caveat, the expert group was able to calculate or estimate the masses, compositions, activities, and heat loads of the waste streams in all cases.

The NDC expert group also carried out a performance assessment for four repository types (clay, granite, salt, and tuff) for one fuel cycle of each cycle class. As a final step, an economic analysis of the fuel cycles was performed. Many of the unit costs are not yet known accurately enough. Therefore, the model used in the calculations of the fuel cycle costs included an option for an uncertainty and sensitivity analysis.

The report of the latest NDC expert group is still unpublished. However, some of its main conclusions can be presented here (Cavedon et al. 2005). The most important and at the same time unsurprising result is that only with fully closed fuel cycles will it be possible to achieve a large reduction in the consumption of natural uranium, the volume of conditioned high-level nuclear waste, or the size of the repository. On the other hand, the advanced nuclear fuel cycles would cost more than an open fuel cycle, but the difference in the total production costs might not be very large (less than 20%).

All three NDC system studies have analysed nuclear fuel cycles in equilibrium. Advanced reactors and fuel cycles should be utilised for a long period of time (maybe a few hundred years) if their potential benefits are to be realised. In other words, advanced nuclear fuel cycles require a long-term commitment to nuclear energy for their benefits to materialise.

3.3 Advanced reprocessing methods

Advanced reprocessing methods are needed to establish safe and economic advanced fuel cycles. Almost all the new technologies based on either hydrometallurgical (wet, aqueous) or pyrochemical (dry) processes are still under development and so far have been tested only on the laboratory scale (Paajanen & Harjula 2003, 2004, 2005, Zilliacus & Anttila 2003).

All present industrial-scale reprocessing plants are using a process called PUREX, which was developed for extracting plutonium from irradiated uranium fuel more than fifty years ago. The process is very efficient and selective. The uranium and plutonium streams are very pure and only about 0.1% of the plutonium and uranium goes into the high-level waste stream. Additionally, the volume of secondary waste has been decreased appreciably.

One of the main goals of P&T research and development is to extend or refine the PUREX process to produce more product streams. At present, it might be possible to separate at least all the actinides (uranium, plutonium, neptunium, americium, and curium), caesium and strontium, technetium and iodine from spent nuclear fuel. Not all the necessary processes are yet efficient enough, however. The main problem may be the separation of americium and curium from lanthanides and the subsequent separation of americium and curium.

In the United States of America, a variation of the PUREX process called UREX is under development (AFCI 2005, pp. 5-6–5-10). The main difference between the processes is that in UREX plutonium and neptunium are separated together in order to reduce the proliferation risk. The UREX process can be extended to include a few extra separation steps. The UREX versions are known as UREX+1, UREX+2, UREX+3, etc.

As mentioned above, very recently research and development of a process called Group Actinide Extraction (GANEX) has been started in the United States of America, France, and Japan. The goal of GANEX is to separate all actinides as a group for continuous recycling.

The PUREX process and its variations may not be able to treat new types of fuel which are planned for use in advanced reactors. The spent fuel of advanced fuel cycles may also be so radioactive that chemicals of hydrometallurgical processes will not survive in such conditions. Therefore, pyrochemical processes are now one of the priority areas of R&D on advanced fuel cycles (NEA 2005a, b, c).

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4. Other nuclear waste studies

The KYT programme mainly focused on studies of spent nuclear fuel management. Nevertheless, there has been a project on reactor waste studies dealing with the waste from the purifying process in nuclear power plants. In addition, there has been a limited but continuous study on the costs of the Finnish nuclear waste management programme.

4.1 Reactor waste studies

4.1.1 C-14 inventories in ion exchange resins to be bituminised[30]

The ion exchange resins which have been used in the purifying processes of nuclear power plants have to be characterised before disposal. The overall aim of this project was to define the current C-14 activity inventories in ion exchange resins to be bituminised at the Olkiluoto nuclear power plant. The focus was on developing and testing a method for C-14 analysis from dried resin samples. The development and testing were done at the Department of Radiochemistry, University of Helsinki. Special work by a radiochemistry student was later guided to perform the C-14 analyses at Olkiluoto.

A new method was developed, tested, and compared to the previously used method (Salonen and Snellman 1986). This was used as a reference method when the new acidifying equipment and C-14 gas-trapping unit (SDEC) were introduced at Olkiluoto. The method used to determine C-14 activity in dried ion exchange resins was tested with four different types of resins used at Olkiluoto. The activity of absorbed C-14 can be measured either by LSC directly from an alkaline solution or from a carbonate suspension. In the earlier method used at Olkiluoto, the C-14 was precipitated along barium carbonate and the precipitation measured with LSC. In the new method the solution is directly measured with LSC.

The method and testing part of this project are reported in Ervanne and Hakanen (2005). The student's special work is reported in Lunden (2004). The method and the results are now used at the Olkiluoto nuclear power plant in C-14 inventories.

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[30] By Heini Ervanne, HYRL.

Lunden, A. 2004. ^{14}C-määritys Olkiluodon bitumoitavissa hartseissa. Working Report of TVO VLJ-3/04. (In Finnish)

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4.2 Costs of nuclear waste management[31]

The management of all nuclear waste arising from existing nuclear reactors in Finland, including operative reactor waste, the decommissioning waste of nuclear power plants (NPPs), and spent nuclear fuel, is estimated to cost around 2,000–2,500 million euro. The lower limit assumes 40-year life times for the reactors, and the upper limit assumes 50 years for the Loviisa reactors and 60 years for the Olkiluoto reactors. Spent fuel management represents more than half of the costs. The costs of the waste from the new reactor under construction at Olkiluoto are not included in the figures.

Nuclear waste management is a particularly long-term project extending over dozens of years, which means that technical and economic preparations must be made in advance for operations that will be realised in the far future. For instance, the decommissioning of NPPs and the closing of disposal tunnels in a spent nuclear fuel repository are activities which are already in sight, but will not take place for the next few years. In order to make sure that the required funding is available when needed, the State Nuclear Waste Management Fund has been established. On the basis of the Nuclear Energy Act, the fund is administered by the Ministry of Trade and Industry (KTM).

The basic principle of the fund is that at all times it can cover the liability, i.e. all future management costs of currently existing nuclear waste. At the end of 2004 the money in the fund covered 100% of the liability. The power companies are entitled to borrow back 75% of their contributions against securities.

To make sure that the remaining liabilities are covered, the power companies, i.e. Fortum Heat and Power Oy and Teollisuuden Voima Oy, are obliged to set aside money into the State Nuclear Waste Management Fund. The power companies must annually present cost estimates for the future management of their nuclear wastes to the KTM. The cost estimates are based on the power companies' latest technical plans, and they also include the decommissioning of NPPs. The authorities review the cost estimates and, on the basis of the review, the KTM stipulates the power companies' contributions, as well as when the contributions are required. The need for contributions depends, for

[31] By Kari Rasilainen, VTT.

instance, on the accumulation rate of nuclear waste and on the nuclear waste management activities carried out so far.

In order to support the KTM in its work, the KYT research programme systematically compiles data on cost estimates from domestic and international sources and analyses the data thus obtained. Nuclear waste management provision needs are reviewed annually in statements prepared to meet the needs of the KTM. The statements on the plans of the power companies are based on a critical review of the cost estimates submitted to the KTM. The statements were prepared within the project "Scientific and technical basis of nuclear waste management", see Annex A.

5. Communication[32]

5.1 Domestic contributions

Domestic communication can be considered as normal communication inside the research programme and between the research programme and the outer world. The most important method of communication inside the research programme was the KYT website (http://www.vtt.fi/kyt), with links to the websites of all participating organisations and links to recent publications. The website had a slightly more compact English version.

An important forum for internal communication was a series of thematic workshops organised by various KYT projects. Although they were mainly for domestic communications, almost all also featured as speakers one or more foreign experts who were collaborating with the organising project. In addition to normal workshops, there were two special workshops for project presentations. A summary of the thematic KYT workshops is shown in Table 19.

Several Steering Group meetings were held annually. Although these were for the most part administrative meetings, there were several project presentations by the project managers, thus serving the purpose of internal communication. A special kind of communication method was the use of electronic information bulletins whenever a reason emerged. The information bulletins are shown on the KYT website.

Nuclear waste-related research in the European Union is organised via the Euratom Treaty. The Euratom research programme (called the Framework Programme, FP) is quite important for Finnish research organisations. Therefore, a national Support Group has been formed by the Ministry of Trade and Industry (KTM) to act as the forum of discussion between the research institutes, authorities, and actors in the Finnish nuclear waste management programme. The co-ordinator of the KYT programme has acted as the secretary of the national Support Group since 2004.

[32] By Kari Rasilainen, VTT.

Table 19. Summary of the thematic workshops in the KYT programme during 2002–2005. The presentations given at the workshops are shown on the KYT website.

Thematic workshop	**Comment**
Final seminar of JYT2001 research programme and presentation of KYT programme	28.10.2002, co-ordination by Kari Rasilainen, VTT
Migration chemistry of radionuclides	21.8.2003, co-ordination by Martti Hakanen, HYRL
Groundwater chemistry during the next ice age	27.8.2003, co-ordination by Juhani Suksi, HYRL
Presentation of Finnish contributions in the DECOVALEX III project	6.10.2003, co-ordination by Esko Eloranta, STUK
Presentation of the results obtained at the Palmottu natural analogue study site	26.3.2004, co-ordination by Lasse Ahonen, GTK
Presentation of partitioning and transmutation (P&T)	30.9.2004, co-ordination by Markku Anttila, VTT
Presentation of the results of the GEOCHEM project (Radionuclide behaviour in granitic rock: development of geochemical modelling tools for safety evaluation)	29.4.2005, co-ordination by Nuria Marcos, TKK
KYT-FUNMIG workshop on migration-related issues	19.10.2005, co-ordination by Marja Siitari-Kauppi, HYRL

5.2 International contributions

International communication is, for the most part, established communication among researchers from different countries that are planning or preparing for nuclear waste disposal. For instance, numerous series of conferences have been going on for years as regular meetings of specialists, and the role of these is to act as nodes in a network of information and experience exchange.

In an effort to reach foreign audiences, the KYT website has a slightly more compact English version. A specific effort to reach audiences outside the research programme was the preparation of a brochure in English that links the KYT programme to the Finnish nuclear waste management programme. The brochure is shown on the KYT website. As a further effort to communicate with foreign audiences, the KYT programme has prepared annually an abstract for the Radioactive Waste Management Abstracts (RWMA) published by the IAEA.

As research is international by nature, several projects of the KYT programme took place in collaboration with international projects and organisations. The KYT programme participated in the DECOVAEX III project during 2002–2003 and in its successor DECOVALEX-THCM during 2004–2005. Chapter 2.1.1 above gives a general description of the two DECOVALEX projects, Chapter 2.2.5 gives a description of the bentonite-related coupled modelling work, and Chapter 2.3.2 gives a description of the modelling work done in the area of rock mechanics.

The International Atomic Agency (IAEA) started the coordinated research project (CRP) 'The use of selected safety indicators (concentration; fluxes) in the assessment of radioactive waste disposal' in 2000; the project was completed in 2005. The KYT programme participated in the project, providing it with Finnish natural safety indicators; see Chapter 2.1.2 above for details. The final report of the CRP has been published in the TECDOC series of the IAEA.

As a separate extension to the IAEA safety indicator project, the KYT programme had an international (Finland, Russia, UK) collaboration project, with the main aim being to develop reactive transport modelling expertise in Finland. It has been acknowledged that our expertise in this area needs improving. Studies were carried out for uranium migration in granitic rock, e.g. using data from the Palmottu natural analogue site. Chapter 2.1.3 gives more technical details of the results.

It may be noted that the contents of the biosphere project of the KYT programme were derived from, in addition to Finnish safety criteria for spent fuel disposal, the environmental radiation protection work being done within international organisations, most notably the IAEA. Besides the IAEA, radiation protection work is done within the International Commission on Radiological Protection (ICRP) and within the EU.

At the beginning of the research programme (2002–2003), i.e. in the time of independent sponsors, the KYT programme participated in the international research project on permafrost. The experimental studies were focused on the Lupin Mine at Canada; see Chapter 2.3.6 for details.

The KYT programme has not formally participated in the current Framework Programme of Euratom, FP6. Notwithstanding, some communication has taken place as the co-ordinator of the KYT participated in the preparation work for the nuclear waste management part of Framework Programme 7 (to be started in 2007), organised by the

European Commission in 2004. In addition, several project managers within KYT are participating in FP6 projects under a separate contract, e.g. FUNMIG (Fundamental Processes of Radionuclide Migration) and NF-PRO (Understanding and Physical and Numerical Modelling of the Key Processes in the Near-Field and their Coupling for Different Host Rocks and Repository Strategies).

5.2.1 OECD/NEA collaboration

The most important international forums of communication have been the regular working groups within the OECD Nuclear Energy Agency (OECD/NEA). Annual country reports have been produced both for the 'Radioactive Waste Management Committee' (RWMC) and for the 'Integration Group for the Safety Case' (IGSC). These two working groups are quite useful in obtaining a structured picture of nuclear waste management programmes in different countries, as well as in presenting the Finnish situation to an influential expert audience. The latest country reports presented in the working groups are shown on the KYT website.

There are some thematic expert groups in the NEA in which the KYT programme has participated. The NEA TDB group (Thermochemical Database Project) has been going on for 20 years and during 2002–2003 the KYT programme participated in its work. Another relevant thematic expert group is the Sorption Forum (OECD/NEA Sorption Project), in which the research programme also participated during the first phase of the KYT programme (2002–2003). Chapter 2.4.6 gives more details of the work of these two expert groups.

The Nuclear Development Committee of the OECD NEA (NDC) organised a two-year (2003–2004) project on the 'Impact of Advanced Fuel Cycle Options on Waste Management Policies'. Partitioning and transmutation (P&T) is included in some of the advanced fuel cycles. The project is a partial continuation of the previous P&T work organised by the NDC. The KYT programme participated in the project with a limited budget. The final report of the project has been delayed and the current deadline is the beginning of 2006. Chapter 3 gives more details on the P&T work and its link to nuclear waste management.

6. Conclusions[33]

General

The nuclear waste management programme in Finland has so far managed to keep to its overall schedule, set in 1983. The next major milestone will be the construction licence application by Posiva Oy for the Olkiluoto spent nuclear fuel disposal facility in around 2012. The systematic progress of the nuclear waste management programme is largely due to the clear division of responsibilities between the main actors in Finnish nuclear waste management. This in turn is largely due to the clarity of the Nuclear Energy Act that controls all nuclear activities in Finland.

The KYT programme was originally started in 2002 as a public research programme funded by individual actors in Finnish nuclear waste management. The Nuclear Energy Act was amended in 2004 to ensure the level of funding for public research programmes. The amended Nuclear Energy Act emphasised the research needs of the Finnish authorities, but all organisations involved in the research programme can obviously benefit from the public results.

From the contents point of view the research in KYT was divided into two broad categories. The main category included studies promoting long-term safety assessments of the geological disposal of spent fuel. The second category included strategic studies of nuclear waste management.

Long-term safety of spent fuel disposal

Methodology

Participation in the DECOVALEX III project and its successor DECOVALEX-THMC provided us with a chance to study coupled thermo-hydro-mechanical (THM) processes in a nuclear waste repository system. The latter project, which is still ongoing, endeavours to include coupled chemical processes into the system. The understanding of coupled processes is essential in assessing the performance of a disposal system.

A safety case of a nuclear waste repository includes a quantitative safety analysis but it may also include other safety factors. Introduction of natural safety indicators is an attempt to utilise more transparent safety factors. The reason for looking for alternative factors is that safety analyses involve advanced mathematics and assertions that are not evident for non-professionals. Therefore, concentrations and mass flows of certain

[33] By Kari Rasilainen, VTT.

compounds in Finnish waters, when measured and calculated, respectively, may provide an independent safety indicator complementing the mathematical safety assessments. The studies within KYT on this topic were part of a larger project by the International Atomic Energy Agency (IAEA).

A conceptually sound and detailed modelling of radionuclide migration in fractured granitic rock requires the inclusion of reactive transport which couples hydrology with geochemistry. This research area needs to be enhanced in Finland, and the application of the Russian SONE code in such modelling was a cost-effective way to increase reactive modelling expertise in Finland.

Release

Experimental copper corrosion analysis indicated that the general corrosion of copper in compacted bentonite wetted with highly saline groundwater virtually stops within ca. 100 h. The corrosion was measured with an on-line corrosion probe developed in the KYT programme. The observation was interpreted as indicating that during the initial transient phase oxygen trapped in the bentonite is presumably consumed and subsequent corrosion occurs. It was considered that traditional weight loss coupons which give an integral measure of corrosion during the whole exposure may be misleading and give too high corrosion rates.

One project had the aim of assessing the potential combined creep and corrosion effects on the copper overpack under foreseen repository conditions. The expected service life of about 100,000 years of a copper canister continues to pose technical challenges for life prediction, because of the difference of up to four orders of magnitude between the service life and conventional range of testing times. Reduced creep life due to small scale defects was observed when testing across the weld root in the radial direction, in comparison with the base material or axial cross-weld performance of the copper cylinder. It was concluded that extended testing is necessary in order better to assess life under the combined effects of creep and corrosion.

Relatively little attention has been paid to the nature of copper corrosion products and even less to the possible interaction of radionuclides with the corrosion products. A literature survey was thus made to clarify the interactions of copper corrosion products, especially nantokite, with uranium in saline and reducing environments. It was found that in such an environment uranium occurs mainly as pitchblende or uraninite and copper as copper sulphides. Furthermore, not a single mineral compound containing Cu, U, and Cl was found in the literature.

A literature survey was also made on the influence of freeze-thaw cycle on the permeability of bentonite and bentonite mixtures. It revealed that the resulting change of hydraulic conductivity in compacted bentonite would probably be smaller than that in bentonite mixtures. It was proposed that this conclusion should be tested experimentally.

A fully coupled thermo-hydro-mechanical (THM) model for an unsaturated swelling porous medium has been derived. Small-scale HM and THM laboratory experiments and large-scale mock-up and in situ experiments for Febex bentonite have been simulated and the results were compared with the experimental observations. The large-scale experiments and the modelling of moisture content showed the importance of geometrical details. The brick wall-like structure of the Febex buffer, consisting of thousands of compacted bentonite blocks with construction gaps in between, appears to lead to transient behaviour. Furthermore, the simulations predict an unsaturated steady state for the condition of prevailing large temperature differences of the large scale tests. The THM behaviour of a hypothetical repository of Canadian design has been simulated within the international DECOVALEX-THMC project. For this purpose the model was applied to simulate the behaviour of MX-80 bentonite (reference bentonite in Finnish plans) and granite. A doctoral thesis on these topics is being prepared.

Bedrock and groundwater

The rock mechanical design of a hard rock repository for spent nuclear fuel is a demanding task, because a high level of safety against rock failure is needed, while the amount of rock reinforcement should be minimised. Because the repository will be located in competent rock at great depth the failure of intact rock can be a critical factor. Thus understanding the failure of brittle rock under low confinement is a key factor for the stability of the repository during operation. Brittle behaviour of rock and spalling-type rock failure were realised using special testing geometry and axial strain as a control variable. The test results can best be utilised as case studies when evaluating the applicability of different modelling methods for the prediction of brittle failure in rock.

A conceptual model for time-dependent crack growth and creep deformation was formulated and successfully implemented in a numerical code within the international DECOVALEX project. The numerical results achieved were shown to be accurate. The time-dependent model appears to work well for tensional loading. The improved FRACOD code can accurately simulate the failure of core samples in laboratory tests. An important achievement was the capability to model realistically brittle axial and lateral response during the failure process. A fundamental observation was made: constant-load experiments showed that Mode II (shear fracture) time-dependent fracture occurs at ambient pressure in laboratory conditions. This has never been shown before.

The relationship between bedrock fracturing and hydrogeology was studied in map interpretation and field experiments. Usually the first step in determining possible water-conducting fracture zones is lineament interpretation, as dense fracturing is a potential highway of groundwater flow. A systematic methodology for lineament interpretation was applied to the Kopparnäs site. The approach is based on separate analysis of mutually independent aerogeophysical datasets and topographic data, and their integration and linking into a coherent set of lineaments. One of the interpreted major lineaments was taken as the target of detailed hydrogeological studies, consisting of drilling, geophysical, and hydrogeological studies of the borehole. Video imaging was compared with the drill core log. Geophysical borehole logging was supported by fluid logging. Many hydraulically active zones were discerned in the borehole, and a clear correlation with geophysical resistivity logs was observed. An interesting observation was the correlation between anomalies in self-potential logs and observed hydraulically active fractures. Additionally, a systematic study of the geophysical borehole logging of the Outokumpu deep borehole (2,500 m) was carried out. The results were compared with available hydraulic tests and a methodology to discern hydraulically active borehole sections based on borehole geophysics was developed. Two master's theses (tech.) were done within the project.

A methodology for the interpretation of geochemical signals in the bedrock was developed in order to couple these signals to U mobilisation events. A conceptual model was formulated and tested for a redox front in the rock matrix surrounding a water-conducting fracture. Oxic water is thought to penetrate the surrounding matrix and oxidise it to an extent controlled by the duration of the oxic pulse, diffusion kinetics, and the abundance of oxygen consuming compounds in the matrix. The existence of a redox front can further be supported by the occurrence of Fe and Mn oxy-hydroxides as these will be formed in the oxidised part of the matrix. The dominant role of geochemical conditions in controlling radioactive disequilibria in groundwater was established when the role of direct alpha recoil was shown to be insignificant in strongly flowing systems. Based on this observation a new ^{234}U vs. ^{238}U method was outlined to interpret $^{234}U/^{238}U$ activity ratios in groundwater. The method was tested with an extensive experimental material from Finland and Sweden, and the measured activities of the U isotopes grouped surprisingly well along trend lines that appeared to characterise the sites. The method was also tested for its ability to show signs of underground excavations, e.g. mines or underground research facilities, and the results were encouraging.

Laboratory studies carried out so far have not proven conclusively the origin of gases in the bedrock, or their connection to seismic creep. The reflections of the seepages in the echo-sounding profiles obtained in the KYT programme imply that part of the gas is derived from the bedrock itself, and the formation of pockmarks seems to be related to fracture zones. Groundwater samples from deep drill holes at Olkiluoto consisted of high volume of gases (methane), which supports the bedrock origin. To reach better and more reliable conclusions, further research with reliable gas sampling techniques and broader analyses should be done. As such, tectonic movements and high hydraulic gradients are recognised in safety analyses as major threats to repository safety in glacial scenarios.

One of the key matters concerning permafrost in safety case considerations is to explore whether the out-freezing process can result in the formation of a saline groundwater layer at the base of the permafrost in crystalline bedrock. Highly saline waters might have undesired effects on the function of the bentonite buffer and increase canister corrosion. The available data from the Lupin Mine do not provide any evidence of highly saline waters below the permafrost. However, there is not enough information to unequivocally rule out the saline freeze-out front hypothesis. It seems that deep permafrost prevents any significant recharge of surface waters. Even in the disturbed mining environment, where a strong artificial hydraulic gradient exists (mine drainage) and a through talik is located next to the mine, the infiltration of recent waters is insignificant. This is, of course, partly due to the poor hydraulic connections, which prevent or slow down the potential flow.

Besides earthquakes, the glacial isostatic adjustment and related land uplift are the most significant indications of ongoing bedrock deformation in Fennoscandia. Seismic activity in Finland is low and heterogeneously distributed and the earthquake density maximums and the areas of postglacial faults have a spatial correlation. Detailed geodetic surveys indicate that crustal deformation occurs unevenly. The bedrock in Finland is so fractured that the deformation is distributed over a number of structures and deformations and displacements along individual structures are very small and difficult to resolve. Postglacial faults are re-activated old faults and the areas of postglacial faulting are still the most seismically active areas in Fennoscandia. The association of seismicity with glacial rebound suggests that in areas experiencing diminishing rebound, the level of seismicity decreases over time. Therefore, palaeoseismicity and geological modelling have to be combined to predict the likely incidence, magnitude, and frequency of future earthquake activity.

The Palmottu natural analogue study has been a major interdisciplinary effort, with funding from the Finnish public sector's nuclear waste research programmes and from the EU. The studies included different branches of geosciences, colloid and microbe studies, and matrix diffusion studies as part of wider migration studies. A very detailed hydrogeological site characterisation programme was carried out at Palmottu. The work, including the comprehensive research, testing, and development of field study methods, resulted in the construction of a hydrogeological model of the site, which was the primary input for the transport models developed. One of the major findings of the site study was that the extensive groundwater chemical data could be used as an additional support to the hydrogeological model. In addition, bedrock redox conditions of the Palmottu site were studied in depth, using the best available measuring methodologies. The results of the Palmottu analogue study provide valuable information for safety case consideration of nuclear waste disposal.

Radionuclide transport studies

The laboratory-scale tracer experiments clearly show the difficulty of seeing the response of a slow process such as matrix diffusion in a dynamic experiment including flowing groundwater. Two different experimental configurations, a rock core column experiment and a fracture experiment, were modelled using consistent parameterisation. It turned out that the channelled groundwater flow is difficult to control experimentally and this makes the modelling a demanding task. The variability observed in successive tests using the same experimental configuration underlines the importance of the flow field in the assessment of the transport and retention of the solutes. So far the modelling has supported the classic matrix diffusion model used in performance assessments, thus adding confidence to the current understanding of the process.

In the studies of pore space geometry in crystalline rock, effective techniques were developed to visualise and analyse the network of interconnected pores and fractures. These techniques included the ^{14}C-PMMA method, confocal microscopy combined with impregnation with fluorescent resin, and high-resolution x-ray tomography. The ^{14}C-PMMA method was extended from previous laboratory tests to in situ tests performed as an international collaboration at the Grimsel rock laboratory in Switzerland. Confocal laser scanning microscopy (CLSM), when combined with impregnation with fluorescent resin which allows imaging to a depth of about 300 micrometers, complements the ^{14}C-PMMA method as it can provide three-dimensional information and has a clearly better resolution. It can thus be used to improve our understanding of interactions between the pore structure and the pore water. The same is true of x-ray tomography which provides really three-dimensional images of the structure of rock with a resolution down to one micrometer. With the best resolution the imaged area has a linear dimension of two millimetres, but with a poorer resolution the imaged size can be extended to the centimetre scale. X-ray tomography was shown to be able to provide the three-dimensional structure of individual fractures in rock, as well as the amount of uranium minerals in rock samples. The detailed data of pore geometry can make a good input to the discussion concerning the role of matrix diffusion in radionuclide transport. The task remaining is to develop a comparably detailed model that can utilise the pore data to their full potential.

Sorption studies in tunnel filling materials emphasised the importance of coupling experimental work with modelling. In particular, the importance of reactive modelling of radionuclide transport became evident. The work included Cs sorption on biotite in different waters.

During 2002–2003 the KYT programme participated in the OECD NEA TBD project and the NEA Sorption Forum. The purpose of the still ongoing Thermodynamic Data Base project (TDB) is to make available a comprehensive, internally consistent, internationally recognised and quality-assured chemical thermodynamic database of selected chemical elements for the use of geochemical modellers. The second phase of the OECD NEA Sorption Forum focused on sorption model testing and demonstration, something that is highly relevant from the point of view of overall migration. A Finnish team was among the 20 teams modelling the total of 7 test cases, and it calculated three cases, one Ni case and two Np cases.

Biosphere

Biosphere studies were focused on environmental radiation protection. A dose calculation tool, MHDC, based on advanced Monte Carlo simulations, had earlier been developed at STUK. During 2003 the tool was tested by comparing its results with well-documented cases found in the literature. The results obtained by MHDC were in good agreement with the published cases.

Strategic alternatives in spent fuel management

There are no real alternatives to the disposal of high-level nuclear waste or spent nuclear fuel in safe geological repositories. However, long-term storage before the disposal and, especially, advanced fuel cycles with effective partitioning and transmutation (P&T) may make it easier to fulfil the long-term safety requirements and especially to reduce the required size of a repository. As a consequence these options may have an impact on the waste management strategy. The KYT programme has participated in the expert groups within the OECD NEA studying advanced nuclear fuel cycles, including P&T. The studies will be published by the NEA.

Other nuclear waste studies

Although the KYT programme has mainly focused on studies of spent nuclear fuel management, there was one project on reactor waste dealing with the waste from the purifying process in a nuclear power plant. The overall aim was to define the current ^{14}C activity inventories in ion exchange resins to be bituminised at the Olkiluoto nuclear power plant. In addition, there has been a limited but continuous study on the costs of the Finnish nuclear waste management programme. The management of all the nuclear wastes arising from the existing four nuclear reactors in Finland, including operative reactor waste, the decommissioning waste of nuclear power plants, and spent nuclear fuel, is estimated to cost around 2,000–2,500 million euro.

Annex A: Project descriptions

Projects belonging to the Finnish Research Programme on Nuclear Waste Management KYT (2002–2005)

1. Co-ordination of the KYT research programme
2. Finnish research on partitioning and transmutation
3. Scientific and technical basis of nuclear waste management
4. DECOVALEX studies in KYT
5. IAEA Coordinated Research Project, The use of selected safety indicators (concentrations, fluxes) in the assessment of radioactive waste disposal
6. Radionuclide behaviour in granitic rock: Development of alternative geochemical modelling tools for safety evaluation
7. Coupled modelling of bentonite buffer
8. Creep and combined corrosion and creep of canister copper
9. Copper corrosion analysis
10. Interaction between corrosion products of radionuclides and copper in saline and reducing environment
11. The effect of freezing and thawing of clay on the hydraulic conductivity of bentonite and bentonite mixtures
12. Post-glacial bedrock movement
13. Strength of crystalline rock in long-term loading
14. Time-dependent fracturing of rock mass and modelling of long-term stability of a repository for spent nuclear fuel
15. Time-dependent fracturing of rock mass and modelling of EDZ of a repository for spent nuclear fuel
16. Bedrock fracturing and hydrogeology
17. Summary of studies at Palmottu
18. Geochemical signs of ice ages in bedrock
19. Geochemical behaviour of a filled fracture
20. Behaviour of rare earth metals in comparison with uranium in a glacial scenario
21. Water-Rock-Interaction – U-REE
22. Gas studies at the Olkiluoto sea area
23. Permafrost studies
24. NEA TDB II, NEA Sorption Project Phase II
25. Fracture flow and radionuclide transport – tracer experiments and characterisation of flow routes
26. Determination of sorption reversibility in Olkiluoto bedrock
27. Radionuclide migration in tunnel filling materials and in bedrock
28. Planning of KULKE project
29. Retention processes in transport along a fracture
30. In-situ study of rocks: Characterization of pore space geometry by ^{14}C-PMMA impregnation
31. In situ study of rocks: Long term diffusion project
32. Characterisation of pore space in rock by tomographic methods
33. Environmental radiation protection
34. C-14 inventories in ion exchange resins to be bituminized from Olkiluoto

NAME OF THE PROJECT Co-ordination of the KYT research programme		
ORGANISATION VTT Processes	**PROJECT MANAGER** Kari Rasilainen	**PROJECT DURATION** 2002–2005

VOLUME (person-months)	**FUNDING (1000 €)**							
	KTM	**STUK**	**POSIVA**	**TVO**	**VYR**	**OWN FUNDING**	**OTHER, e.g. EU**	**TOTAL**
18	122				144			266

RESEARCH PARTNERS

FINNISH ORGANISATIONS	**FOREIGN ORGANISATIONS**	**PROJECTS IN OTHER RESEARCH PROGRAMMES**
KYT organisations	-	-

GENERAL AIMS

The aim of the project was to provide technical co-ordination of the KYT programme. A framework programme was compiled as the general objective. Practical co-ordination was done by technically supporting the Steering Group in preparing annual public calls for project proposals, in evaluating the proposals, and in communicating the results. The Steering Group prepared each year a funding recommendation for the whole research programme. Specific tasks in co-ordination were to compile annual research plans and annual reports for the whole research programme. These reports had the aim of following the implementation of the framework programme. The co-ordination project also compiled every year three interim reports on the progress of individual projects in relation to their specific aims. Further tasks in co-ordination were related to communication that were realised via a website, topical scientific seminars, and information bulletins.

MAIN ACHIEVEMENTS

- compilation of the framework programme (in Finnish and in English) for KYT together with the Steering Group. The framework programme was updated when the Nuclear Energy Act was amended in 2004
- supporting technically the annual project call, evaluation, and communication procedures
- compilation of annual research plans and annual reports for the whole research programme
- compilation of three annual interim reports for the whole research programme
- organisation of eight thematic workshops together with the host projects
- KYT website
- preparing a brochure in English of the research programme
- acting as the secretary of the Steering Group in 22 meetings
- compilation of the final report of the KYT programme together with the research projects
- compilation of the draft framework programme (in Finnish) for the next research programme (KYT2010) together with the Steering Group.

SCIENTIFIC STAFF

D.Sc.(Tech) Kari Rasilainen Co-ordinator

NAME OF THE PROJECT Finnish research on partitioning and transmutation (P&T)		
ORGANISATION VTT, HYRL	**PROJECT MANAGER** Markku Anttila	**PROJECT DURATION** 2002–2005

VOLUME (person-months)	**FUNDING (1000 €)**							
	KTM	**STUK**	**POSIVA**	**TVO**	**VYR**	**VTT OWN FUNDING**	**OTHER, e.g. EU**	**TOTAL**
24	94				74	44		212

RESEARCH PARTNERS

FINNISH ORGANISATIONS	**FOREIGN ORGANISATIONS**	**PROJECTS IN OTHER RESEARCH PROGRAMMES**
VTT, HYRL	OECD/NEA, KTH	EMERALD/SAFIR

GENERAL AIMS

The project was aimed at increasing national capabilities and knowledge on the field of P&T.

MAIN ACHIEVEMENTS

A capability to carry out reactor physics calculations related to burning of minor actinides in thermal reactors; participation in the OECD/NEA working and expert groups; state-of-the-art reports on advanced reprocessing methods and treatment of secondary wastes.

SCIENTIFIC STAFF

MSc(Tech)	Markku Anttila	Project manager, Senior Research Scientist / VTT
LicTech	Jaakko Leppänen	Research Scientist/ VTT
MSc	Riitta Zilliacus	Senior Research Scientist /VTT
Phil. Lic	Airi Paajanen	Research Scientist/HYRL
PhD	Risto Harjula	University Lecturer /HYRL

NAME OF THE PROJECT
Scientific and technical basis of nuclear waste management

ORGANISATION	PROJECT MANAGER	PROJECT DURATION
VTT Processes	Kari Rasilainen (2002–2004) Jarmo Lehikoinen (2005)	2002–2005

VOLUME (person-months)	FUNDING (1000 €)							
	KTM	STUK	POSIVA	TVO	VYR	OWN FUNDING	OTHER, e.g. EU	TOTAL
21	135				185			320

RESEARCH PARTNERS

FINNISH ORGANISATIONS	FOREIGN ORGANISATIONS	PROJECTS IN OTHER RESEARCH PROGRAMMES
GTK, HYRL, TKK/KAL, VTT/RTE	-	-

GENERAL AIMS

The aim of the project was to develop methods and expertise for technical and economic assessment of spent nuclear fuel management in support of Finnish authorities. This broad aim was divided into smaller subtopics for performance assessment methodology, nuclear fuel cycle collaboration, cost estimates, paleohydrogeological studies in safety case, and review of the Kd model in performance assessment.

MAIN ACHIEVEMENTS

- participation in relevant expert groups dealing with performance assessments and safety case within the OECD/NEA, e.g. RWMC (Radioactive Waste Management Committee) and IGSC (Integration Group for the Safety Case)
- participation in the Finnish and international OECD/NEA collaboration in the area of advanced nuclear fuel cycles, including partitioning and transmutation (P&T)
- annual reviews for the Ministry of Trade and Industry (KTM) of the cost estimates of the whole Finnish nuclear waste management by the nuclear energy producing power companies
- paleohydrogeological studies in the area of uranium-series disequilibria (USD) in order to develop a feasible interpretation methodology for observed USD data in rock matrix and in flowing groundwater especially in relation to past glacial cycles
- review of the basis and applicability of the widely used Kd model for sorption in near-field and far-field studies of a spent fuel repository (to be published in 2006).

SCIENTIFIC STAFF

MSc(Tech)	Jarmo Lehikoinen	Project manager (2005)
DSc(Tech)	Kari Rasilainen	Project manager (2002–2004)
DSc(Tech)	Timo Vieno	Performance assessment methodology
LicTech	Henrik Nordman	Performance assessment methodology
MSc(Tech)	Tiina Koljonen	Cost estimates

<table>
<tr><td colspan="9">NAME OF THE PROJECT
DECOVALEX studies in KYT</td></tr>
<tr><td colspan="2">ORGANISATION
STUK</td><td colspan="3">PROJECT MANAGER
Esko Eloranta</td><td colspan="4">PROJECT DURATION
1999–2003 (DECOVALEX III), years 2002–2003 in KYT period
2004–2005 (DECOVALEX THMC)</td></tr>
<tr><td>VOLUME (person-months)</td><td colspan="8">FUNDING (1000 €)</td></tr>
<tr><td></td><td>KTM</td><td>STUK</td><td>POSIVA</td><td>TVO</td><td>VYR</td><td>OWN FUNDING</td><td>OTHER, e.g. EU</td><td>TOTAL</td></tr>
<tr><td>1 (STUK)</td><td></td><td>142</td><td></td><td></td><td>50</td><td></td><td></td><td>192</td></tr>
<tr><td colspan="9">RESEARCH PARTNERS</td></tr>
<tr><td colspan="3">FINNISH ORGANISATIONS
TKK, Fracom Oy</td><td colspan="3">FOREIGN ORGANISATIONS
Univ. Uppsala</td><td colspan="3">PROJECTS IN OTHER RESEARCH PROGRAMMES
-</td></tr>
<tr><td colspan="9">GENERAL AIMS
To support the national research teams at TKK, Fracom Oy, Uppsala University
To pay the funding fees to SKI.</td></tr>
<tr><td colspan="9">MAIN ACHIEVEMENTS
From the general point of view main achievements are immaterial, like the gaining of deeper understanding of coupled processes, and increasing the expertise on national level.</td></tr>
<tr><td colspan="9">SCIENTIFIC STAFF (at STUK)
DSc(Tech) Esko Eloranta Project manager</td></tr>
</table>

<table>
<tr><td colspan="10">NAME OF THE PROJECT
IAEA Coordinated Research Project, The use of selected safety indicators (concentrations, fluxes) in the assessment of radioactive waste disposal</td></tr>
<tr><td colspan="3">ORGANISATION
STUK</td><td colspan="3">PROJECT MANAGER
Karl-Heinz Hellmuth</td><td colspan="4">PROJECT DURATION
2000–2003</td></tr>
<tr><td>VOLUME (person-months)</td><td colspan="9">FUNDING (1000 €)</td></tr>
<tr><td></td><td>KTM</td><td>STUK
370</td><td>POSIVA</td><td>TVO</td><td>VYR</td><td>OWN FUNDING</td><td>OTHER, e.g. EU</td><td colspan="2">TOTAL
370</td></tr>
<tr><td colspan="10">RESEARCH PARTNERS</td></tr>
<tr><td colspan="3">FINNISH ORGANISATIONS
STUK, GTK, VTT</td><td colspan="4">FOREIGN ORGANISATIONS
Univ. of St. Petersburg
Univ. of Bern
Univ. of Reading</td><td colspan="3">PROJECTS IN OTHER RESEARCH PROGRAMMES
-</td></tr>
<tr><td colspan="10">GENERAL AIMS
Develop a database of measured natural concentrations and fluxes of target substances (elements, radioactive isotopes and particular chemical compounds) in and between different solid and aqueous materials, from different geographical areas, showing the spatial variability in these concentrations from the regional to the site scale focussed on an agreed suite of elements (U, Th, K, Rb, Sn, Cs, Cu, Ra and Rn).</td></tr>
<tr><td colspan="10">MAIN ACHIEVEMENTS
Database of measured natural concentrations and fluxes of target substances (elements, radioactive isotopes and particular chemical compounds) in and between different solid and aqueous materials, from different geographical areas, showing the spatial variability in these concentrations from the regional to the site scale focussed on an agreed suite of elements (U, Th, K, Rb, Sn, Cs, Cu, Ra and Rn).</td></tr>
</table>

NAME OF THE PROJECT
Radionuclide behaviour in granitic rock: Development of alternative geochemical modelling tools for safety evaluation

ORGANISATION	PROJECT MANAGER	PROJECT DURATION
TKK	Nuria Marcos	2004

VOLUME (person-months)	FUNDING (1000 €)							
	KTM	STUK	POSIVA	TVO	VYR	OWN FUNDING	OTHER, e.g. EU	TOTAL
					70			70

RESEARCH PARTNERS

FINNISH ORGANISATIONS	FOREIGN ORGANISATIONS	PROJECTS IN OTHER RESEARCH PROGRAMMES
TKK, GTK, Univ. of Helsinki	Univ. of St. Petersburg Univ. of Reading	-

GENERAL AIMS

To develop an assessment methodology for a spent fuel repository based on sound geochemical and hydro-geological principles that effectively utilises site characterisation data. It should adopt best international practice with respect to modelling protocols and the use of fundamental data/concepts.

MAIN ACHIEVEMENTS

Characterization of rock properties and U mineralization at the Palmottu site. Verification of observations of U minerals and rock mineral alteration products in nature by coupled modelling. Simulation of the post-glacial evolution of the Palmottu groundwater-rock system. Setting the time frames of hydrogeochemical influences by independent dating based on U series nuclides and putting the Palmottu results into a larger regional context.

NAME OF THE PROJECT
Coupled modelling of bentonite buffer

ORGANISATION	PROJECT MANAGER	PROJECT DURATION
TKK Institute of Mathematics	Petri Jussila	2004–2005

VOLUME (person-months)	FUNDING (1000 €)							
	KTM	STUK	POSIVA	TVO	VYR	OWN FUNDING	OTHER, e.g. EU	TOTAL
24					142			142

RESEARCH PARTNERS

FINNISH ORGANISATIONS	FOREIGN ORGANISATIONS	PROJECTS IN OTHER RESEARCH PROGRAMMES
-	-	Decovalex-THMC

GENERAL AIMS

Derivation of fully coupled thermo-hydro-mechanical (THM) model for unsaturated swelling porous medium.

Calibration of the model against Febex bentonite.

Simulation of small scale laboratory tests and large scale experiments for Febex bentonite.

Participation to the Task A of the international Decovalex-THMC project.

MAIN ACHIEVEMENTS

The fully coupled THM model for unsaturated swelling porous medium has been derived and calibrated. The modelling approach is published in two separate articles in a refereed scientific journal. The first article describing the general model and the TH application was accepted 3/2005. The second article describing the THM model with swelling is under consideration for publication - the third revised version was submitted 1/2006.

Small scale HM and THM laboratory experiments for Febex bentonite and the larger scale mock-up and in-situ experiments have been simulated with comparisons to the observations.

In Task A of Decovalex THMC the THM behaviour of a hypothetical repository of Canadian design has been simulated. For this purpose the model is applied to simulate the behaviours of MX-80 bentonite and granite. The results are published in one individual paper and in two joint papers in the GeoProc2006 conference in Nanjing 5/2006.

A doctorate thesis covering the above mentioned bentonite issues is prepared.

ACADEMIC DEGREES

Doctor of Technology	Petri Jussila (to be completed in 2006)	Thermomechanics of swelling unsaturated porous media – Compacted bentonite clay in spent fuel disposal

SCIENTIFIC STAFF

MSc	Petri Jussila	Project manager

NAME OF THE PROJECT Creep and combined creep and corrosion of canister copper								
ORGANISATION VTT			**PROJECT MANAGER** Pertti Auerkari			**PROJECT DURATION** 2004–2005		
VOLUME (person-months)	**FUNDING (1000 €)**							
	KTM	**STUK**	**POSIVA**	**TVO**	**VYR**	**OWN FUNDING**	**OTHER, e.g. EU**	**TOTAL**
14					86	11	66 (SKI)	163

RESEARCH PARTNERS

FINNISH ORGANISATIONS	FOREIGN ORGANISATIONS	PROJECTS IN OTHER RESEARCH PROGRAMMES
-	-	-

GENERAL AIMS

- to determine the long term creep and creep damage behaviour of copper including welds
- to assess experimentally the effects of combined creep and corrosion under simulated repository conditions.

MAIN ACHIEVEMENTS

- extended creep data base for copper (max > 30 000 h) and evaluation of creep damage and small scale defects in friction stir welds
- predictive model for creep of copper (parent and welds), and revised creep life prediction based on creep modelling
- new feature testing facility for combined creep and corrosion testing of welded copper, and first testing results on effects of combined creep/corrosion environment.

SCIENTIFIC STAFF

LicTech	Stefan Holmström	Feature testing, modelling
MSc(Eng)	Jorma Salonen	Metallography, damage evaluation
LicTech	Pertti Nenonen	Electron microsopy analysis
MSc(Eng)	Juha Veivo	Creep testing & analysis
MSc(Eng)	Anssi Laukkanen	Numerical analysis (FEA)

NAME OF THE PROJECT
Copper corrosion analysis

ORGANISATION	PROJECT MANAGER	PROJECT DURATION
VTT	Timo Saario	2002–2003, 2005

VOLUME (person-months)	FUNDING (1000 €)							
	KTM	STUK	POSIVA	TVO	VYR	OWN FUNDING	OTHER, e.g. EU	TOTAL
7		40			40		23	103

RESEARCH PARTNERS

FINNISH ORGANISATIONS	FOREIGN ORGANISATIONS	PROJECTS IN OTHER RESEARCH PROGRAMMES
Cormet Oy	SKI, Sweden	-

GENERAL AIMS

To develop monitoring tools for corrosion of copper under disposal conditions. Using the tools developed to verify experimentally the estimates made on the extent and rate of the general corrosion of copper under disposal conditions.

MAIN ACHIEVEMENTS

The general corrosion rate of copper as measured in closely simulated Olkiluoto-type final disposal environment with fully compacted bentonite was shown to be clearly lower than the acceptable maximum rate. The scientific monitoring tools developed in this work are available for possible use in field measurements in the future and have already been further applied in other research projects.

SCIENTIFIC STAFF

Ph.D.	Iva Betova	Data analysis
Ph.D.	Martin Bojinov	Experiments, data analysis
Dr.	Timo Saario	Project manager, sensor development
Dr.	Petri Kinnunen	Sensor development

NAME OF THE PROJECT		
Interaction between corrosion products of radionuclides and copper in saline and reducing environment		
ORGANISATION TKK	**PROJECT MANAGER** Nuria Marcos	**PROJECT DURATION** 2003

VOLUME (person-months)	FUNDING (1000 €)							
	KTM	STUK	POSIVA	TVO	VYR	OWN FUNDING	OTHER, e.g. EU	TOTAL
4		13						13

RESEARCH PARTNERS

FINNISH ORGANISATIONS	FOREIGN ORGANISATIONS	PROJECTS IN OTHER RESEARCH PROGRAMMES
-	-	-

GENERAL AIMS

To investigate the behaviour of copper corrosion products in relation to uranium in a saline groundwater environment.

MAIN ACHIEVEMENTS

A literature survey of the possible coexistence of nantokite (CuCl) and uranium in saline groundwater conditions. No natural compounds or minerals were found in the literature in which Cu, U, and Cl exist at the same time.

SCIENTIFIC STAFF

DSc(Tech) Nuria Marcos Project manager

<table>
<tr><td colspan="9">NAME OF THE PROJECT
The effect of freezing and thawing of clay on the hydraulic conductivity of bentonite and bentonite mixtures</td></tr>
<tr><td colspan="3">ORGANISATION
VTT Communities and Infrastructure</td><td colspan="3">PROJECT MANAGER
Seppo Saarelainen</td><td colspan="3">PROJECT DURATION
2002</td></tr>
<tr><td>VOLUME (person-months)</td><td colspan="8">FUNDING (1000 €)</td></tr>
<tr><td></td><td>KTM</td><td>STUK</td><td>POSIVA
24</td><td>TVO</td><td>VYR</td><td>OWN FUNDING</td><td>OTHER, e.g. EU</td><td>TOTAL
24</td></tr>
<tr><td colspan="9">RESEARCH PARTNERS</td></tr>
<tr><td colspan="3">FINNISH ORGANISATIONS
Posiva</td><td colspan="3">FOREIGN ORGANISATIONS
-</td><td colspan="3">PROJECTS IN OTHER RESEARCH PROGRAMMES
-</td></tr>
<tr><td colspan="9">GENERAL AIMS
The aim of the project was to do a literature survey on the effect of freezing and thawing of clay on the hydraulic conductivity of bentonite and bentonite mixtures. The results were expected to help estimate the effect of possible permafrost on the function of bentonite barrier in the spent fuel repository.</td></tr>
<tr><td colspan="9">MAIN ACHIEVEMENTS
Saarelainen, S. 2002. Influence of freeze-thaw on the permeability of bentonite and bentonite mixtures. literature study (in Finnish, English abstract), Posiva Work Report 2002-31, 44 p. + app. 2 p.</td></tr>
<tr><td colspan="9">SCIENTIFIC STAFF
D.Sc.(Tech) Seppo Saarelainen Project manager</td></tr>
</table>

NAME OF THE PROJECT								
Post-glacial bedrock movements								
ORGANISATION Geol. Surv. Finland (GTK)			**PROJECT MANAGER** Paavo Vuorela			**PROJECT DURATION** 2003		
VOLUME (person-months)	**FUNDING (1000 €)**							
	KTM	**STUK**	**POSIVA**	**TVO**	**VYR**	**OWN FUNDING**	**OTHER, e.g. EU**	**TOTAL**
3	26							26

RESEARCH PARTNERS

FINNISH ORGANISATIONS	FOREIGN ORGANISATIONS	PROJECTS IN OTHER RESEARCH PROGRAMMES
GTK	-	-

GENERAL AIMS

Purpose of the KYT-project “Post Glacial Bedrock Movements” was to summarise the current knowledge of the postglacial deformation of bedrock in Finland. These are divided into recent earthquake activity, postglacial faulting, glacial isostatic rebound and current horizontal crustal deformation.

MAIN ACHIEVEMENTS

Clearly, the glacial isostatic adjustment and related land uplift horizontal motions in addition to earthquakes are the most significant indications of the ongoing bedrock deformation in Fennoscandia, and these are closely connected. Maximum vertical uplift rates based on the GPS measurements are about 11 mm/yr and horizontal motions are up to 2 mm/yr. Tectonic component is about 10% of the land uplift (or 1 mm/yr). Horizontal motions are directed outward from area of the fastest uplift. Horizontal tectonic motions are also less than 1 mm/yr.

Seismic activity in Finland is low and heterogeneously distributed and the earthquake density maximums and the areas of postglacial faults have a spatial correlation. Detailed geodetic surveys indicate that crustal deformation occurs unevenly. However, the bedrock in Finland is so fractured that the deformation is distributed over a number of structures and that deformations and displacements along individual structures are very small and difficult to resolve. Fault intersections can form a locked area where stresses large enough to trigger intraplate earthquakes can build up. In the absence of intersections, the pre-existing faults can creep at a lower stress threshold.

In Fennoscandia, plate-boundary tectonic stresses drive the regional compressive stress field, but to account for the current level of seismicity the glacial isostatic adjustment has a very important role. Brittle crust is near the point of failure, and, consequently, small changes, like glacial rebound related, (0.1 Mpa) in the state of stress can nucleate earthquakes are sufficient to reactive optimally oriented pre-excising weaknesses.

Stress orientations inferred from the strain measurements of the first order triangulation network and seismological stress data shows a) the dominating ridge-push/mantle drag related compression and, b) evidence on significant local variations of the surface stress field influenced by the orientation of major fracture zones.

Postglacial faults are re-activated old faults and the areas of postglacial faulting are still the most seismically active areas in Fennoscandia. The association of seismicity with glacial rebound suggests that in areas experiencing diminishing rebound, the level of seismicity decreases over time. Therefore, palaeoseismicity and geological modelling have to be combined to predict the likely incidence, magnitude and frequency of future earthquake activity.

SCIENTIFIC STAFF

Lic. Tech	Paavo Vuorela	Project management
PhD	Juhani Ojala	Tectonics and bedrock deformation
MSc.	Aimo Kuivamäki	Postglacial faults t

<table>
<tr><td colspan="10">NAME OF THE PROJECT
Strength of crystalline rock in long-term loading</td></tr>
<tr><td colspan="3">ORGANISATION
Helsinki University of Technology</td><td colspan="3">PROJECT MANAGER
Juha Antikainen</td><td colspan="4">PROJECT DURATION
2002–2003</td></tr>
<tr><td>VOLUME (person-months)</td><td colspan="9">FUNDING (1000 €)</td></tr>
<tr><td></td><td>KTM</td><td>STUK</td><td>POSIVA</td><td>TVO</td><td>VYR</td><td>OWN FUNDING</td><td>OTHER, e.g. EU</td><td>TOTAL</td><td></td></tr>
<tr><td>7</td><td>43</td><td></td><td></td><td></td><td></td><td>7</td><td></td><td>50</td><td></td></tr>
<tr><td colspan="10">RESEARCH PARTNERS</td></tr>
<tr><td colspan="3">FINNISH ORGANISATIONS
-</td><td colspan="3">FOREIGN ORGANISATIONS
-</td><td colspan="4">PROJECTS IN OTHER RESEARCH PROGRAMMES
-</td></tr>
<tr><td colspan="10">GENERAL AIMS
The state-of-art in testing of brittle crystalline rock is reviewed as well as the present estimates about the loading conditions in underground disposal of spent nuclear fuel in crystalline rock. Different strength criteria and their parameters are reviewed and also the practical acquisition of the parameters by laboratory testing.</td></tr>
<tr><td colspan="10">MAIN ACHIEVEMENTS
According to the literature review, the loading conditions in underground disposal site can be predicted, at adequate accuracy, for time from present to possible recovery of disposal canisters before next glaciation. The understanding of the loading during a complete glaciation cycle should be still enhanced.

The material models most often utilized in rock mechanics are not able to reproduce all failure modes observed in brittle rock. Modelling of the formation of thin slabs under high stress and low confiment rock mass (spalling) requires either major manipulation of material model parameters or use of completely different material models. In practical part of this work laboratory tests were made for determination the parameters of CWFS (Cohesion Weakening Friction Strengthening) model. Because the behaviour predicted with CWFS model does not manifest itself in standard rock mechanics tests, a special testing geometry was adapted with specimen width/diameter ratio of two. The test results show expected brittle behaviour of rock. These results can be utilized by comparing different modelling results to the actual rock behaviour in tests.</td></tr>
<tr><td colspan="10">SCIENTIFIC STAFF
DTech. Juha Antikainen Project manager
M.Sc. Pekka Eloranta Research Scientist, rock mechanical testing</td></tr>
</table>

NAME OF THE PROJECT

Time-dependent fracturing of rock mass and modelling of EDZ of a repository for spend nuclear fuel (CREEP, 2005)

Time-dependent fracturing of rock mass and modelling of long-term stability of a repository for spent nuclear fuel (2004)

ORGANISATION	PROJECT MANAGER	PROJECT DURATION
FRACOM	Mikael Rinne	CREEP 2004–2006. Jointly DECOVALEX project will continue until September 2007.

VOLUME (person-months)	FUNDING (1000 €)							
	KTM	STUK	POSIVA	TVO	VYR	OWN FUNDING	OTHER, e.g. EU	TOTAL
24					150	32		182

RESEARCH PARTNERS

FINNISH ORGANISATIONS	FOREIGN ORGANISATIONS	PROJECTS IN OTHER RESEARCH PROGRAMMES
Fracom Oy, Helsinki University of Technology	GeoFrames GmbH Nick Barton and Associates	DECOVALEX-THMC

GENERAL AIMS

The CREEP project aims to apply fracture mechanics principles to predict time-dependent behaviour of fractured rock mass. The study is focused on the Excavation Disturbed Zone (EDZ) around a rock tunnel.

MAIN ACHIEVEMENTS

A conceptual model for time-dependent crack growth has been established. It is based on sub-critical crack growth theory. The concept has been successfully implemented in a numerical code, FRACOD^{2D}. The improved code has been used to simulate rock failure detected in short and long term laboratory tests. The preliminary simulations suggest that the time-dependent model works well under tensional and compressive loading.

Experimental testing methods to determine subcritical fracture growth parameters under Mode I (tensile) and Mode II (shear) conditions have been established. According to preliminary results the developed methods have a good potential to determine these parameters. The constant load experiments have shown that Mode II time-dependent fracturing behaviour exists under laboratory conditions. This has never been reported before. Strain-stepping tests have been made to study time-dependent deformation properties of stressed rock. The analysis is ongoing and the monitored time-dependent behaviour will be modelled to verify the developed numerical method.

SCIENTIFIC STAFF

LicTech	Mikael Rinne	Theoretical studies, modelling, analysis
PhD	Baotang Shen	Code improvement
PhD	Hee-Suk Lee	Modelling, analysis
DTech	Juha Antikainen	Laboratory testing procedures, analysis
PhD	Tobias Backers	Laboratory testing procedures, analysis
PhD	Nick Barton	Technical auditing

NAME OF THE PROJECT Bedrock fracturing and hydrogeology		
ORGANISATION Geological Survey of Finland	**PROJECT MANAGER** Lasse Ahonen	**PROJECT DURATION** 2004–2005 (2006)

VOLUME (person-months)	**FUNDING (1000 €)**							
	KTM	**STUK**	**POSIVA**	**TVO**	**VYR**	**OWN FUNDING**	**OTHER, e.g. EU**	**TOTAL**
29					200	5		205

RESEARCH PARTNERS

FINNISH ORGANISATIONS	**FOREIGN ORGANISATIONS**	**PROJECTS IN OTHER RESEARCH PROGRAMMES**
VTT/Communities and Infrastructure	-	-

GENERAL AIMS

The fundamental aim of the project is to examine the bedrock "structures" in hydrogeologist's eyes and, thus to support the efforts to model groundwater flow and radionuclide transport processes adequately. In more practical level, the project deals with the research activities required – and have been applied – to produce data for the geohydrological models, and background information for conceptualization of bedrock hydrogeology.

Initially, impetus for project came from several earlier site-specific studies and hydrogeological experience derived from them: Palmottu, Outokumpu, Lupin mine (Canada), Hyrkkölä, Pori, Ylivieska. Current project aims at an integration of the earlier data with hydrogeological information from on-going site-studies: Kopparnäs, Outokumpu deep hole, Klaukkala fracture zone.

This three-year project will continue in 2006.

MAIN ACHIEVEMENTS

- Application and demonstration of feasibility of systematic lineament interpretation methodology in bedrock hydrogeological studies
- A methodological study to document/classify fracturing in drill core and it's comparison with fracturing observed on the borehole wall (TV survey)
- Feasibility study and comparison of different geophysical borehole logs in locating hydraulically active fracture zones in boreholes
- Use of 'fluid-logging' as a method to characterize fracture flow conditions
- An examination of the block-mosaic concept and it's relevance to bedrock hydrogeology.

ACADEMIC DEGREES

Master of Sciences	Paula Jääskeläinen 2005	Water conductive structures in crystalline bedrock at the Kopparnäs site. Master of Sciences thesis (engineering). Helsinki University of Technology, Materials Science and Rock Engineering
Master of Sciences	Anna-Maria Tarvainen 2006	Identification of water conductive structures in Outokumpu deep borehole using borehole geophysical methods. Master of Sciences thesis (engineering). Helsinki University of Technology, Department of Civil and Environmental Engineering.

SCIENTIFIC STAFF

PhD	Lasse Ahonen	Project manager
MScTech	Paula Jääskeläinen	Kopparnäs studies
MScTech	Kimmo korhonen	Lineament studies, structural modelling
student	Anna-Maria Tarvainen	Outokumpu deep borehole studies
PhD	Jarkko Jokinen	Borehole geophysical studies
MSc	Antero Lindberg	Bedrock fracturing and fracture mineralogy
MSc	Veikko Hakkarainen	Quaternary geology, aerial photo interpretation

NAME OF THE PROJECT Summary of studies at Palmottu								
ORGANISATION Geological Survey of Finland			**PROJECT MANAGER** Juha Kaija			**PROJECT DURATION** 9/2003–9/2004		
VOLUME (person-months)	**FUNDING (1000 €)**							
	KTM	**STUK**	**POSIVA**	**TVO**	**VYR**	**OWN FUNDING**	**OTHER, e.g. EU**	**TOTAL**
2		20						20
RESEARCH PARTNERS								
FINNISH ORGANISATIONS -			**FOREIGN ORGANISATIONS** -			**PROJECTS IN OTHER RESEARCH PROGRAMMES** -		
GENERAL AIMS General aim was to improve the awareness of the results of Palmottu Natural Analogue Studies. Research conducted during different phases of the project was reviewed, and published in a summary report.								
MAIN ACHIEVEMENTS Palmottu bibliography up to 31.8.2004.								
SCIENTIFIC STAFF PhD Lasse Ahonen Editor of the report								

NAME OF THE PROJECT								
Geochemical signs of ice ages in bedrock								
ORGANISATION University of Helsinki, Laboratory of Radiochemistry			**PROJECT MANAGER** Juhani Suksi			**PROJECT DURATION** 2002–2005		
VOLUME (person-months)	**FUNDING (1000 €)**							
	KTM	**STUK**	**POSIVA**	**TVO**	**VYR**	**OWN FUNDING**	**OTHER, e.g. EU**	**TOTAL**
73		135	20		180		8	343

RESEARCH PARTNERS

FINNISH ORGANISATIONS	FOREIGN ORGANISATIONS	PROJECTS IN OTHER RESEARCH PROGRAMMES
VTT Processes, TKK, Geological Survey of Finland	GRS, FZK-INE, NRI-REZ	EU-FUNMIG

GENERAL AIMS

1) Identification of glacial melt water induced marks in rock and water using U series
2) Use of melt water marks in characterising groundwater flow system
3) Assessment of radionuclide migration under melt water influence.

MAIN ACHIEVEMENTS

1) Methods for studying melt water – rock interaction phenomena
2) Identification of melt water influence with the help of U decay series
3) A method for interpreting $^{234}U/^{238}U$-activity ratios in groundwater.

SCIENTIFIC STAFF

Dr	Juhani Suksi	Project coordination and research
MSc	Paula Juntunen	Research
MSc	Susanna Salminen	Research

NAME OF THE PROJECT Geochemical behaviour of a filled fracture								
ORGANISATION TKK		**PROJECT MANAGER** Nuria Marcos			**PROJECT DURATION** 2002			
VOLUME (person-months)	**FUNDING (1000 €)**							
	KTM	**STUK**	**POSIVA**	**TVO**	**VYR**	**OWN FUNDING**	**OTHER, e.g. EU**	**TOTAL**
		25						25
RESEARCH PARTNERS								
FINNISH ORGANISATIONS HYRL, VTT, GTK			**FOREIGN ORGANISATIONS** -			**PROJECTS IN OTHER RESEARCH PROGRAMMES** -		
GENERAL AIMS To study radionuclide migration at the interface between smectite-rock and smectite-water.								
MAIN ACHIEVEMENTS Collaboration with Finnish research partners in the area on natural analogues to study the penetration of glacial meltwaters into bedrock. The results support the formation of glacial scenarios in the safety assessment of spent fuel disposal.								
ACADEMIC DEGREES Doctor of Technology		Nuria Marcos 2002		Lessons from Nature – The behaviour of Technical and Natural barriers in the Geological Disposal of Spent Nuclear Fuel.				
SCIENTIFIC STAFF MSc		Nuria Marcos		Project manager				

NAME OF THE PROJECT		
Behaviour of rare earth metals in comparison with uranium in a glacial scenario		
ORGANISATION	**PROJECT MANAGER**	**PROJECT DURATION**
TKK	Nuria Marcos	2003, 2004

VOLUME (person-months)	FUNDING (1000 €)							
	KTM	STUK	POSIVA	TVO	VYR	OWN FUNDING	OTHER, e.g. EU	TOTAL
8		6			33	5		44

RESEARCH PARTNERS

FINNISH ORGANISATIONS	FOREIGN ORGANISATIONS	PROJECTS IN OTHER RESEARCH PROGRAMMES
HYRL, VTT, GTK	-	-

GENERAL AIMS

To increase knowledge of the behaviour of Rare Earth Elements (REE's) in relation that of U in a case in which glacial meltwater intrudes bedrock.

MAIN ACHIEVEMENTS

Collaboration with Finnish research partners in the area on natural analogues to study the penetration of glacial meltwaters into bedrock. Past redox changes in the bedrock have been studied by parallel interpretation of REE and uranium-series disequilibrium profiles in rock samples. Indications have been obtained that processes that cause uranium mobility also cause REE mobility to some extent.

SCIENTIFIC STAFF

DSc(Tech)	Nuria Marcos	Project manager

NAME OF THE PROJECT Water-Rock-Interaction – U-REE		
ORGANISATION TKK	**PROJECT MANAGER** Kirsti Loukola-Ruskeeniemi	**PROJECT DURATION** 6/2005–2/2006 (8 months)

VOLUME (person-months)	**FUNDING (1000 €)**							
	KTM	**STUK**	**POSIVA**	**TVO**	**VYR**	**OWN FUNDING**	**OTHER, e.g. EU**	**TOTAL**
6					37			37

RESEARCH PARTNERS

FINNISH ORGANISATIONS	**FOREIGN ORGANISATIONS**	**PROJECTS IN OTHER RESEARCH PROGRAMMES**
University of Helsinki, Laboratory of Radiochemistry	-	-

GENERAL AIMS

The aim of this study was to find out the effect of acidic groundwater and glacial melt waters on the release of uranium/REE phases.

MAIN ACHIEVEMENTS

Uranium is released from secondary phases during contact with acidic waters either from glacial melt water intrusion or meteoric waters. The release of REEs is coupled to the release of uranium only in association with calcite, but not in association with e.g. iron or phosphates.

ACADEMIC DEGREES

M.Sc. (tech.)	Mira Markovaara 2006	Mineralogical approach to the study of the release of U and REE in granitic rock samples

SCIENTIFIC STAFF

Under-graduate	Mira Markovaara	Student
Professor	Kirsti Loukola-Ruskeeniemi	Supervisor
Lecturer	Marja Siitari-Kauppi	Advisor
Consultant	Nuria Marcos	Advisor

NAME OF THE PROJECT								
Gas studies at the Olkiluoto sea area (Hydrogen/carbon isotope anomalies in the pore waters of the Holocene sediments in the Olkiluoto area, the Gulf of Bothnia, Baltic Sea)								
ORGANISATION STUK, GTK			**PROJECT MANAGER** Kaisa-Leena Hutri			**PROJECT DURATION** 2002–2003 (2004 outside KYT)		
VOLUME (person-months)	**FUNDING (1000 €)**							
	KTM	**STUK**	**POSIVA**	**TVO**	**VYR**	**OWN FUNDING**	**OTHER, e.g. EU**	**TOTAL**
		32						32
RESEARCH PARTNERS STUK, GSF								
FINNISH ORGANISATIONS STUK, GTK, Dating Laboratory and Finnish Museum of Natural History at the University of Helsinki			**FOREIGN ORGANISATIONS** -			**PROJECTS IN OTHER RESEARCH PROGRAMMES** -		

GENERAL AIMS

Aim of that study was to confirm the occurrence of methane in the submarine sediments (2002) and to reveal the origin of the observed gas/fluid release by means of sampling and chemical analyses, and to look whether there is a connection between the gas observations and bedrock fault creep (2003–2004).

MAIN ACHIEVEMENTS

Study confirmed the existence of methane in the sediments, but did not reliable show the origin of the gases.

Final Report: Hämäläinen, J. & Kotilainen, A., 2004. Sedimenttien kaasututkimukset Olkiluodon merialueella. Loppuraportti 1/2004. 29.1.2004.

SCIENTIFIC STAFF

Kaisa-Leena Hutri STUK	Project manager
Aarno Kotilainen GTK	
Eloni Sonninen University of Helsinki	Sample dating

NAME OF THE PROJECT Permafrost studies								
ORGANISATION Geol. Surv. Finland (GTK)			**PROJECT MANAGER** Timo Ruskeeniemi			**PROJECT DURATION** 2002–2003		
VOLUME (person-months)	**FUNDING (1000 €)**							
	KTM	**STUK**	**POSIVA**	**TVO**	**VYR**	**OWN FUNDING**	**OTHER, e.g. EU**	**TOTAL**
30			114			114	260	488

RESEARCH PARTNERS

FINNISH ORGANISATIONS	FOREIGN ORGANISATIONS	PROJECTS IN OTHER RESEARCH PROGRAMMES
Geological Survey of Finland, Posiva	SKB (Sweden), Nirex Ltd (UK), OPG (Canada), University of Waterloo (Canada)	-

GENERAL AIMS

In order to evaluate the long-term performance of a geologic repository, the influence of periglacial and glacial conditions must be considered. To be able to address the relevant questions with respect of the geologic disposal concept, the Permafrost Project was initiated in 2001. The fundamental idea was to investigate permafrost at a representative field site in order to evaluate the various hypotheses presented and reviewed in literature with the emphasis on the understanding of the cryogenic processes and hydrogeology in crystalline rock under deep permafrost conditions. Lupin gold mine in Arctic Canada was the first field research site of the project.

MAIN ACHIEVEMENTS

The existence of structural heterogeneity at the Lupin site is evidenced by (i) wide range in groundwater salinity (2.2 to 36.3 g/L) and gas differences; (ii) the heterogeneity of the hydraulic heads at the site; (iii) the apparent lack of hydraulic connection between boreholes during hydraulic testing; and (iv) the observed heterogeneity in gas flow from fractures versus fluid flow during down-hole video surveys.

One of the major implications of such a poorly connected system would be the *limitation on recharge* to depth or across the site. The results from the electromagnetic soundings suggest that the groundwater table is consistently deeper than the assumed base of the permafrost (540 m) for a distance of 2–3 kilometres away from the mine. In addition, the inflow of natural water into the mine is low, reflecting the low bulk permeability of the fractured rocks at this site (only about 53 000 m^3 of water is annually pumped out from the mine). The heterogeneity in salinity and the observed head measurements (with the possible exception of the non-tritiated fresh water sampled on the 1130 m level), would suggest that the connections to the assumed large talik structure (unfrozen bedrock) connected to a nearby major lake are poor.

Talik is an unfrozen section of ground within permafrost. In the Lupin area all shallow lakes will freeze down to the bottom during winter. The bottoms of larger lakes deeper than 2–3 m will remain unfrozen all year. These lakes have the potential to support an open talik. Still larger lakes are likely to conceal taliks extending through the deep permafrost. Their impact on hydrogeological regime depends on the geological setting beneath. The taliks within an intact bedrock would be hydrogeologically isolated “worm-holes”. However, some chains of lakes related to regional lineaments, may have partly interconnected unfrozen channels beneath. The most significant talik structure in the Lupin area would be related to the Lake Contwoyto which is only 1,5 km N from the mine. In principal the taliks provide routes for vertical flow, if a gradient exists.

Highly saline waters under high pressures have not been observed in or directly under the permafrost. The lack of such a large reservoir of high pressure, gas-rich saline fluid coupled with the structural heterogeneity discussed above constrains our conceptual model of the site. There are mine-induced contaminants moving slowly downwards in one large fault system. However, there appears to be a mixing or dilution of this saline, nitrate-rich contamination by a matrix fluid as seen in the results for a variety of stable isotopes (^{2}H, ^{18}O, ^{37}Cl) used to fingerprint the water and solutes. In addition, anomalies in some parameters such as sulphate concentrations in fluids from the permafrost zone may prove to be precipitates from the freeze out process.

Gases found in the deep groundwater systems below the permafrost are apparently thermogenic in origin. The site does have considerable metasedimentary rocks which were originally deep ocean sediments. The gas composition and age would therefore most likely be tied to the age of the metamorphic event (i.e. hundreds of millions to billions of years). In most cases, it is assumed that by association, the gases, matrix fluids and much of the saline groundwaters have similar evolutionary histories.

SCIENTIFIC STAFF

MSc.	Timo Ruskeeniemi	Project management, hydrogeochemistry
PhD	Lasse Ahonen	Hydrogeochemistry, hydrogeology
Lic. Tech.	Markku Paananen	Geophysics
MSc.	Juha Kaija	Hydrogeochemistry
MSc.	Antero Lindberg	Mineralogy and petrology
MSc. (tech.)	Jukka Lehtimäki	Geophysics

NAME OF THE PROJECT
NEA TDB II, NEA Sorption Project Phase II

ORGANISATION	**PROJECT MANAGER**	**PROJECT DURATION**
VTT Processes HYRL	Ulla Vuorinen (VTT) Jarmo Lehikoinen (VTT) Martti Hakanen (HYRL)	2002–2003

VOLUME (person-months)	**FUNDING (1000 €)**							
	KTM	**STUK**	**POSIVA**	**TVO**	**VYR**	**OWN FUNDING**	**OTHER, e.g. EU**	**TOTAL**
4			54					54

RESEARCH PARTNERS

FINNISH ORGANISATIONS	**FOREIGN ORGANISATIONS**	**PROJECTS IN OTHER RESEARCH PROGRAMMES**
HYRL, VTT, Posiva	NEA, ANSTO, ONDARF/NIRAS, RAWRA, ANDRA,IPSN, GRS/FZK/FZR, CRIEPI,JNC, ENRESA,NAGRA/PSI/HSK, BNFL, NIREX, NRC	-

GENERAL AIMS

The main aims of the TDB project is to make available a comprehensive, internally consistent, internationally recognised and quality assured chemical thermodynamic database on specific elements to meet the specialised modelling requirements for safety assessments of radioactive waste disposal systems centred around performance assessment (PA) technical needs, and also to maintain and update the existing database as well as the on-line services.

The main aim of the Sorption Project is to promote the use of thermodynamic sorption modelling in sorption data selection for PA of radioactive waste disposal.

MAIN ACHIEVEMENTS

The update on the chemical thermodynamics of Uranium, Neptunium, Plutonium, Americium and Technetium (Chemical Thermodynamics, Vol. 5, Elsevier 2003). Review work on Ni, Se, Zr and selected organic ligands to be published later (Vol. 6, 7, 8 and 9).

Book: NEA Sorption Project Phase II, Interpretation and Prediction of Radionuclide Sorption onto Substrates Relevant for Radioactive Waste Disposal Using Thermodynamic Sorption Models. (NEA No 5992).

SCIENTIFIC STAFF

Msc	Ulla Vuorinen	Project manager 2003 (TDB management board))
Msc(Tech)	Jarmo Lehikoinen	Project manager 2002, sorption modelling
MSc	Martti Hakanen	Project manager 2003 (Sorption Project management board)

NAME OF THE PROJECT

Fracture flow and radionuclide transport – tracer experiments and characterisation of flow routes (2002–2003 Posiva)
Determination of sorption reversibility in Olkiluoto bedrock (2002–2003 Posiva)
Radionuclide migration in tunnel filling materials and in bedrock (2004–2005 VYR)

ORGANISATION	PROJECT MANAGER	PROJECT DURATION
University of Helsinki, Lab. of radiochemistry	Pirkko Hölttä Martti Hakanen	2002–2005

VOLUME (person-months)	FUNDING (1000 €)							
	KTM	STUK	POSIVA	TVO	VYR	OWN FUNDING	OTHER, e.g. EU	TOTAL
90			210		320	15		545

RESEARCH PARTNERS

FINNISH ORGANISATIONS	FOREIGN ORGANISATIONS	PROJECTS IN OTHER RESEARCH PROGRAMMES
VTT Processes, GTK	-	-

GENERAL AIMS

Fracture flow and radionuclide transport studies aimed to examine the processes that cause retention in solute transport through rock fractures, especially focused on the matrix diffusion, and to estimate importance of the retention processes during transport in different scales and flow conditions.

Sorption experiments aimed to study cation exchange on biotite, mainly to determine the cation exchange selectivity coefficients of Na, K, Ca and Cs and to study the kinetics of Cs sorption reactions. Reaction parameter values are used in reactive transport modeling.

MAIN ACHIEVEMENTS

- Hydraulic characterization of the natural fracture, determination of the flow channel distribution for the tracer tests, and block–scale and rock–core column experiments.
- Conceptualization of advection-dispersion and matrix diffusion in a sufficient accuracy to reproduce the experimental results.
- Demonstration of the effects of the matrix diffusion on the slightly sorbing tracer breakthrough curves using two different experimental configurations.
- Knowledge and understanding of the transport and retention processes transferable to different scales.
- Determination of cation exchange selectivity coefficients for Sodium, Kalium, Calcium and Cesium and successful application in modeling batch reactions in minerologically well defined rock.
- Found that the order of selectivity is Cs>K>Na>Ca, selectivity of Cs is especially high when the concentration of Cs is below 1E-6 M.
- Found that among studied cations Kalium is the only one that can compete with Cesium even in relatively small concentrations.
- Found several different cation exchange sites in biotite according to the modelling.

ACADEMIC DEGREES

Doctor of Philosophy	Pirkko Hölttä 2002 (JYT2001)	Radionuclide migration in crystalline rock fractures – Laboratory study of matrix diffusion

SCIENTIFIC STAFF

Ph.D.	Pirkko Hölttä	Senior researcher, Fracture flow and radionuclide transport
M.Sc.	Jarkko Kyllönen	Researcher, Sorption in tunnel filling materials
M.Sc.	Martti Hakanen	Laboratory manager, Sorption in tunnel filling materials

NAME OF THE PROJECT Planning of KULKE project								
ORGANISATION HYRL			**PROJECT MANAGER** Martti Hakanen			**PROJECT DURATION** 2002		
VOLUME (person-months)	**FUNDING (1000 €)**							
	KTM	**STUK**	**POSIVA**	**TVO**	**VYR**	**OWN FUNDING**	**OTHER, e.g. EU**	**TOTAL**
1			5					5
RESEARCH PARTNERS								
FINNISH ORGANISATIONS HYRL, VTT			**FOREIGN ORGANISATIONS** -			**PROJECTS IN OTHER RESEARCH PROGRAMMES** -		
GENERAL AIMS Planning of co-ordinated radionuclide migration related Posiva-funded studies.								
MAIN ACHIEVEMENTS Plan for studies of radionuclides in bentonite (VTT) and in host rock (HYRL).								
SCIENTIFIC STAFF MSc Martti Hakanen Project manager								

NAME OF THE PROJECT Retention processes in transport along a fracture								
ORGANISATION VTT			**PROJECT MANAGER** Antti Poteri			**PROJECT DURATION** 2004–2005		
VOLUME (person-months)	**FUNDING (1000 €)**							
	KTM	**STUK**	**POSIVA**	**TVO**	**VYR**	**OWN FUNDING**	**OTHER, e.g. EU**	**TOTAL**
4					55			55

RESEARCH PARTNERS		
FINNISH ORGANISATIONS	**FOREIGN ORGANISATIONS**	**PROJECTS IN OTHER RESEARCH PROGRAMMES**
Laboratory of Radiochemistry, University of Helsinki	-	-

GENERAL AIMS

Examine the processes that cause retention in solute transport through rock fractures. The focus of the work is on the matrix diffusion.

MAIN ACHIEVEMENTS

The modelling indicated that matrix diffusion for tracer transport through a natural fracture can be studied in the laboratory scale. Solute transport is very sensitive to the variations in the flow system. However, successful experiments were performed for the natural fracture and for the rock columns. Effects of the matrix diffusion that were observed in both experiments are in good agreement with the corresponding modelling results.

SCIENTIFIC STAFF

MSc	Antti Poteri	Project Manager

NAME OF THE PROJECT

In-situ study of rocks: Characterization of pore space geometry by ^{14}C-PMMA impregnation (PSG)

ORGANISATION	PROJECT MANAGER	PROJECT DURATION
HYRL	Marja Siitari-Kauppi	2002–2004

VOLUME (person-months)	FUNDING (1000 €)							
	KTM	STUK	POSIVA	TVO	VYR	OWN FUNDING	OTHER, e.g. EU	TOTAL
48		92			70			162

RESEARCH PARTNERS

FINNISH ORGANISATIONS	FOREIGN ORGANISATIONS	PROJECTS IN OTHER RESEARCH PROGRAMMES
STUK, GTK, JYFL,TKK(GT),S&R	Uni Poitiers (HYDRASA), ERM, ANDRA Geotechnisches Institut NAGRA	-

GENERAL AIMS

To develop a new application based on the PMMA method in combination with quantitative petrography tools for in situ rock matrix characterisation.

MAIN ACHIEVEMENTS

The PMMA autoradiography technique is possible to use for rock matrix visualisation and characterisation in situ.

ACADEMIC DEGREES

PhD	Marja Siitari-Kauppi 2002	Development of ^{14}C-polymethylmethacrylate method for the characterisation of low porosity media. Application to rocks in geological barriers of nuclear waste storage
PhD	Dimitri Prét 2003	
PhD	Magalie Cassiaux 2004	

SCIENTIFIC STAFF

MSc	Maarit Kelokaski	Scientist, maternity leave 8.2004–4.2006
MSc	Laura Penttinen	Scientist, from 9.2004
PhD	Marja Siitari-Kauppi	Project manager

NAME OF THE PROJECT In situ study of rocks: Long term diffusion project (LTD)								
ORGANISATION HYRL			**PROJECT MANAGER** Marja Siitari-Kauppi		**PROJECT DURATION** 2005–			
VOLUME (person-months)	**FUNDING (1000 €)**							
	KTM	**STUK**	**POSIVA**	**TVO**	**VYR**	**OWN FUNDING**	**OTHER, e.g. EU**	**TOTAL**
16					87	10		97
RESEARCH PARTNERS								
FINNISH ORGANISATIONS JYFL, STUK,GTK, TKK		**FOREIGN ORGANISATIONS** Nagra, Geotechnical Institute, JNC,NRI, RAWRA, HYDRASA, ERM				**PROJECTS IN OTHER RESEARCH PROGRAMMES** -		

GENERAL AIMS

1. To perform a long term diffusion experiment in situ.

2. To measure the bedrock structures in different scales and to characterise the porewater content in low porosity rocks.

MAIN ACHIEVEMENTS

-

SCIENTIFIC STAFF

PhD	Marja Siitari-Kauppi	Project manager
MSc	Laura Penttinen	Scientist

NAME OF THE PROJECT Characterisation of pore space in rock by tomographic methods		
ORGANISATION University of Jyväskylä, Dept. of Physics	**PROJECT MANAGER** Jussi Timonen	**PROJECT DURATION** 2004-2005

VOLUME (person-months)	**FUNDING (1000 €)**							
	KTM	**STUK**	**POSIVA**	**TVO**	**VYR**	**OWN FUNDING**	**OTHER, e.g. EU**	**TOTAL**
28					178			178

RESEARCH PARTNERS

FINNISH ORGANISATIONS	**FOREIGN ORGANISATIONS**	**PROJECTS IN OTHER RESEARCH PROGRAMMES**
University of Helsinki, Lab. of Radiochemistry	University of Oviedo, Dept. of Geology	-

GENERAL AIMS

Characterisation from micrometre to centimetre scale of pore space, fractures and mineral components of rock samples using high-resolution x-ray tomography, confocal laser scanning microscopy and autoradiography.

MAIN ACHIEVEMENTS

Development of methods in x-ray tomography and related image analysis, and in confocal microscopy combined with impregnation with fluorescent resin, such that the aims of research can be addressed. First determinations were made of porosity, 3D structure of fractures, and contents of uranium minerals of rock samples by x-ray tomography, and of fractures and conducting pores by confocal microscopy in combination with autoradiography.

SCIENTIFIC STAFF

Prof.	Jussi Timonen	Project leader, all scientific aspects
Dr.	Markko Myllys	X-ray tomography
Dr.	Marja Siitari-Kauppi	Confocal microscopy and autoradiography
MSc	Ari Jäsberg	Image analysis
MSc	Tuomas Turpeinen	Image analysis
MSc	Mikko Voutilainen	X-ray tomography
	Tomi Lähdemäki	Confocal microscopy

NAME OF THE PROJECT Environmental radiation protection		
ORGANISATION STUK/Research Department	**PROJECT MANAGER** Kirsti-Liisa Sjöblom	**PROJECT DURATION** 2003

VOLUME (person-months)	FUNDING (1000 €)							
	KTM	STUK	POSIVA	TVO	VYR	OWN FUNDING	OTHER, e.g. EU	TOTAL
1		16						16

RESEARCH PARTNERS

FINNISH ORGANISATIONS	FOREIGN ORGANISATIONS	PROJECTS IN OTHER RESEARCH PROGRAMMES
STUK	-	EU 5th Framework project FASSET

GENERAL AIMS

The aim of the project was to further develop and test the non-human radiation dose modelling software.

MAIN ACHIEVEMENTS

- testing of the software NHDC (own by STUK) by comparing it with well documented cases found in the literature
- applying it to calculation of doses to perch in a Finnish lake (1 proceedings publication).

SCIENTIFIC STAFF

MSc	Kirsti-Liisa Sjöblom	Project Manager
PhD	Riitta Hänninen	Project Support
LicPh	Marko Oksanen	Testing, developing and application of the model

NAME OF THE PROJECT
C-14 inventories in ion exchange resins to be bituminized from Olkiluoto

ORGANISATION	PROJECT MANAGER	PROJECT DURATION
University of Helsinki, Department of Chemistry, Radiochemistry Laboratory	Heini Ervanne Martti Hakanen	2002

VOLUME (person-months)	FUNDING (1000 €)							
	KTM	STUK	POSIVA	TVO	VYR	OWN FUNDING	OTHER, e.g. EU	TOTAL
6			21	9				30

RESEARCH PARTNERS

FINNISH ORGANISATIONS	FOREIGN ORGANISATIONS	PROJECTS IN OTHER RESEARCH PROGRAMMES
TVO, Posiva	-	-

GENERAL AIMS

Main aim was to develop and test a suitable method for the C-14 inventories in ion exchange resins to be bituminized at Olkiluoto Nuclear Power Plant. The method was planned to be used in future with the C-14 collector unit at Olkiluoto. A special work of a student was conducted so that the assessment of C-14 activities in resin samples was made.

MAIN ACHIEVEMENTS

Developed and tested workable method and preliminary results from real resin samples are achieved. A special work of student in radiochemistry is completed. Method and results are published in Working Reports of TVO.

SCIENTIFIC STAFF

PhD	Heini Ervanne	Scientist
MSc	Martti Hakanen	Laboratory engineer
	Anne Lunden	Student

Annex B: List of most important publications

Long-term safety of spent fuel disposal

Performance assessment methodology

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Annex C: Organisation of the KYT research programme

The role of the Steering Group was to plan the research programme in general, set the focus of the research, and follow the quality of the results. There was annually an open call for project proposals. After the evaluation of the project proposals the Steering Group submitted a proposal to the Ministry of Trade and Industry (KTM) for an annual research plan of the programme.

The co-ordinator of the KYT programme was given the task of collating research plans, following the implementation of the plans, and reporting the progress of projects to the Steering Group. Furthermore, the co-ordinator acted as the secretary of the Steering Group.

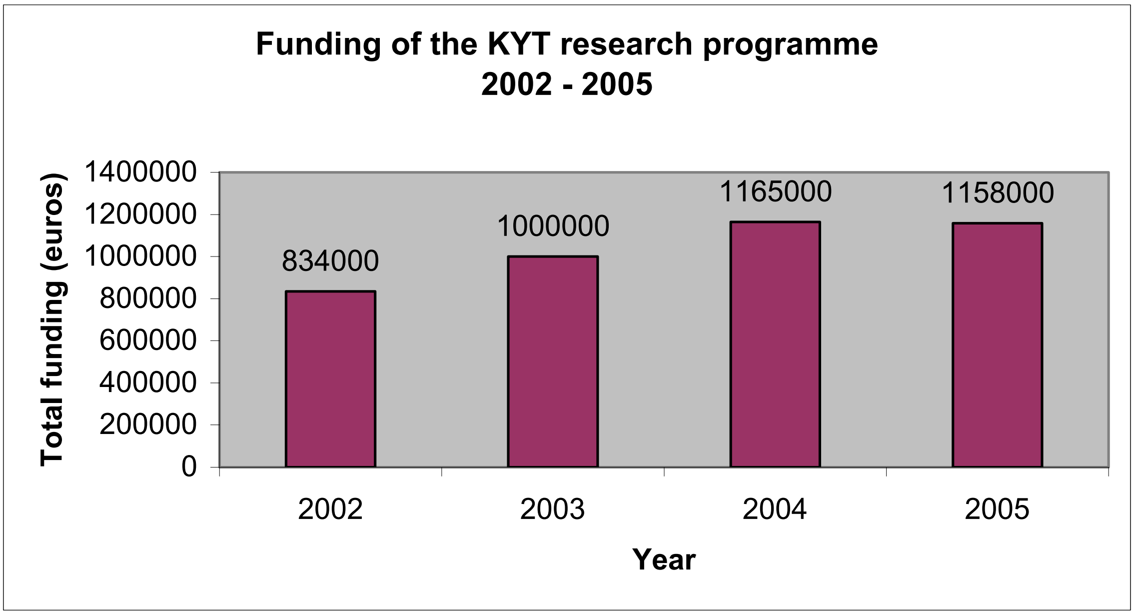
Steering group of the KYT research programme 2002–2005.

Member	Organisation
chief counsellor Anne Väätäinen **(chair 2002–2003)**	Ministry of Trade and Industry (KTM)
director Tero Varjoranta (2002–26.9.2002)	Radiation and Nuclear Safety Authority (STUK)
head of Nuclear Waste Section Esko Ruokola (26.9.2002–2005; **chair 2004–2005)**	Radiation and Nuclear Safety Authority (STUK)
senior inspector Kaisa-Leena Hutri	Radiation and Nuclear Safety Authority (STUK)
senior technology adviser Piia Moilanen (2002–2003; 3.9.2004–16.3.2005)	National Technology Agency of Finland (Tekes)
senior technology adviser Pia Salokoski (2004–3.9.2004; 16.3.2005–31.12.2005)	National Technology Agency of Finland (Tekes)
technology manager Reijo Munther	National Technology Agency of Finland (Tekes)
manager, research and development Eero Patrakka (2002–2004)	Teollisuuden Voima Oy
manager Jari Tuunanen (2005)	Teollisuuden Voima Oy
manager of operation support Anneli Reinvall	Teollisuuden Voima Oy
senior adviser Päivi Mäkinen (2002)	Fortum Power and Heat Oy
manager, technical development Harriet Kallio (2003–2005)	Fortum Power and Heat Oy
senior adviser Ilpo Kallonen	Fortum Nuclear Services Oy
project manager Margit Snellman	Saanio & Riekkola Oy
research director Juhani Vira	Posiva Oy
senior research scientist Kari Rasilainen **(secretary)**	Technical Research Centre of Finland (VTT)

Annex D: Funding of the KYT research programme

The annual funding of the KYT programme is depicted in the diagram below. During the first two years (2002–2003) the programme was funded by independent sponsors. In 2002 the most important sponsors were the Ministry of Trade and Industry (KTM) (200 000 €), the Radiation and Nuclear Safety Authority (STUK) (420 000 €), Posiva Oy. (192 000 €) and Teollisuuden Voima Oy. (22 000 €). In 2003 the sponsors were KTM (210 000 €), STUK (366 000 €), and Posiva (407 000 €). In 2003 the funding of the research institutes themselves was 17 000 €.

From 2004 on, after the amendment of the Nuclear Energy Act, funding was provided by the dedicated Nuclear Waste Research Fund established in the State Nuclear Waste Management Fund (VYR). In 2004 the funding of VYR was 1 041 000 €, external funding was 93 000 € and that of the research institutes 31 000 €. In 2005 the funding of VYR was 1 101 000 €, external funding 33 000 € and that of the research institutes 24 000 €.



Annex E: Evolution of the emphasis of the KYT research programme

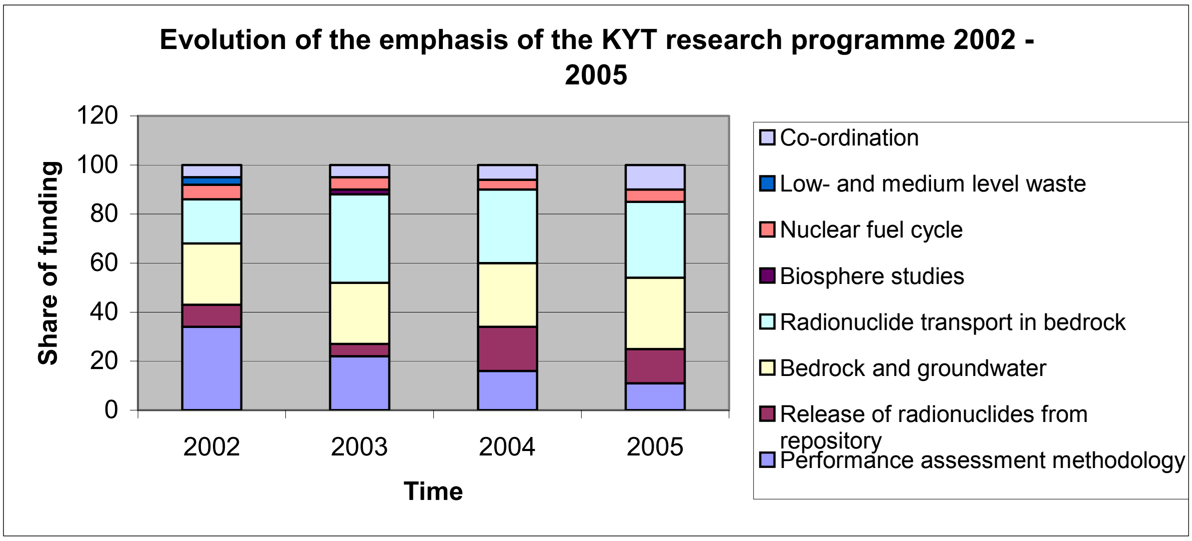
The evolution of various research areas in terms of relative funding is shown in the table and figure below. It can be seen that after four years of the research programme the most important research areas were Radionuclide transport in bedrock (31 % of funding), Bedrock and groundwater (29 % of funding), and Release of radionuclides from repository (14 % of funding).

Evolution of the research profile in the KYT programme during 2002–2005.

KYT research areas	**Year**							
	2002		2003		2004		2005	
	Projects	Share of funding (%)	Projects	Share of funding (%)	Projects	Share of funding (%)	Projects	Share of funding (%)
Long-term safety of spent fuel disposal								
Performance assessment methodology	3	34	4	22	3	16	2	11
Release of radionuclides from repository	2	9	3	5	3	18	3	14
Bedrock and groundwater	5	25	7	25	4	26	4	29
Radionuclide transport in bedrock	4	18	3	36	4	30	4	31
Biosphere studies	0	0	1	2	0	0	0	0
Strategic studies								
Nuclear fuel cycle	1	6	1	5	1	4	1	5
Low- and medium level waste	1	3	0	0	0	0	0	0
Co-ordination	1	5	1	5	1	6	1	10
Projects in total	17	100	20	100	16	100	15	100
Funding in total*	834 000 €		1 000 000 €		1 165 000 €		1 158 000 €	

* Includes funding by the State Nuclear Waste Management Fund, other external funding, and own funding by the resarch instutues

The funding shares allocated to different research areas are shown in the figure below.



Published by

Series title, number and report code of publication

VTT Research Notes 2337
VTT–TIED–2337

Author(s)
Rasilainen, Kari (Ed.)

Title

The Finnish Research Programme on Nuclear Waste Management (KYT) 2002–2005

Final Report

Abstract

The producers of nuclear energy are responsible for the safe handling, management, and disposal of their wastes, as well as for the costs arising, according to the Finnish nuclear energy legislation. National authorities supervise the nuclear waste management and issue regulations for this purpose. The research needs of the authorities have traditionally been supported by public research programmes. The four-year KYT programme (2002–2005) followed the earlier public nuclear waste research programmes that started as long ago as in 1989.

The main research area in KYT was in studies promoting the assessment of the long-term safety of the geological disposal of spent nuclear fuel. This wide area was divided into five sub-areas: (1) the methodology of safety assessment; (2) the release of radionuclides from the repository; (3) bedrock and groundwater; (4) radionuclide transport in bedrock, and (5) biosphere studies. The second research area covered strategic alternatives in spent nuclear fuel management. Studies in this area were focused on advanced fuel cycles, including e.g. partitioning and transmutation (P&T). In addition, there were limited studies on reactor wastes resulting from the operation of nuclear power plants.

KYT promoted Finnish know-how in nuclear waste management for the use of national authorities. It acted as a national forum for discussion and co-operation between the authorities, the nuclear industry, and the research community. Furthermore, it trained new experts in the field, as the most experienced nuclear waste specialists are approaching their retirement.

The next major milestone in the Finnish nuclear waste management programme is the construction licence application for the disposal facility for spent fuel, due around 2012. The licensing process increases the need for high-quality research on the technical safety of the facility. The successor to the KYT programme, called KYT2010, will be carried out from 2006 to 2010.

Keywords
nuclear waste management, Finland, repositories, safety, radionuclides, migration, bedrock, ground water, spent fuel, reactor wastes, environmental protection

ISBN
951–38–6786–2 (soft back ed.)
951–38–6787–0 (URL: http://www.vtt.fi/publications/index.jsp)

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The Finnish Research Programme on Nuclear Waste Management (KYT) was conducted during 2002–2005. KYT was co-ordinated by the Technical Research Centre of Finland (VTT) under the guidance of the Steering Group, which included representatives of all the actors in Finnish nuclear waste management, both the authorities and the nuclear industry. The primary role of KYT was to support the Finnish authorities.

The main research area in KYT was studies promoting the assessment of the long-term safety of the geological disposal of spent nuclear fuel. This wide area was divided into studies on the methodology of safety assessment, on the release of radionuclides from the repository, on bedrock and groundwater, on radionuclide transport in bedrock, and on biosphere issues. The second research area covered strategic alternatives in spent nuclear fuel management. The studies in this area were focused on advanced fuel cycles, including e.g. partitioning and transmutation (P&T).

KYT promoted Finnish know-how in nuclear waste management. It acted as a national forum for discussion and co-operation between the authorities, the nuclear industry, and the research community. Furthermore, it trained new experts in the field, as the most experienced nuclear waste specialists are approaching their retirement.

Tätä julkaisua myy	Denna publikation säljs av	This publication is available from
VTT	VTT	VTT
PL 1000	PB 1000	P.O. Box 1000
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Puh. 020 722 4404	Tel. 020 722 4404	Phone internat. + 358 20 722 4404
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ISBN 951–38–6786–2 (soft back ed.) ISBN 951–38–6787–0 (URL: http://www.vtt.fi/inf/pdf/)
ISSN 1235–0605 (soft back ed.) ISSN 1455–0865 (URL: http://www.vtt.fi/inf/pdf/)