



KYT2010 Finnish Research Programme on Nuclear Waste Management 2006–2010

Final Report

Publications of the Ministry of Employment
and the Economy
Energy and the Climate
26/2011



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KYT2010 Finnish Research Programme on Nuclear Waste Management 2006–2010 – Final Report			
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<p>The Finnish Research Programme on Nuclear Waste Management, KYT2010, was launched in 2006. The research period closed at the end of 2010.</p> <p>Key themes of the research programme include 1) strategic analyses of nuclear waste management, 2) assessment of the safety of final nuclear waste disposal and 3) sociological analyses. The assessment of the safety of final nuclear waste disposal comprises three sectors: technical barriers, bedrock and groundwater, and the release and transport of radionuclides.</p> <p>The roughly 40 or so research projects underway during the research period have primarily been concerned with assessing the safety of nuclear waste management. The State Nuclear Waste Management Fund allocated funding totalling approximately EUR 7 million to the research projects.</p> <p>The following research organisations have participated in the KYT2010 research programme: VTT – the Technical Research Centre of Finland, the Aalto University School of Science and Technology, University of Helsinki Laboratory of Radiochemistry, the Geological Survey of Finland, University of Jyväskylä, University of Eastern Finland Kuopio Campus, and Numerola Oy.</p> <p>Work in the research programme has been based on mutual cooperation and division of duties between the research programme steering group, two support groups, a coordinator and the research programmes. The research programme steering group has included representatives of the Radiation and Nuclear Safety Authority, Tekes – the Finnish Funding Agency for Technology and Innovation, Fortum Power and Heat Oy, Teollisuuden Voima Oyj, Posiva Oy and the Ministry of Employment and the Economy.</p> <p>Contact persons within the Ministry of Employment and the Economy: Energy Department/Jaana Avolahti, tel +358 10 606 4836</p>			
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Foreword

The Finnish Research Programme on Nuclear Waste Management, KYT2010, took place between 2006 and 2010. The research programme continued the coordinated nuclear waste management research programmes of Finland's public administration, launched in 1989. Precedents of the KYT2010 research programme were the following: publicly funded JYT nuclear waste research programme in 1989–1993, public administration's JYT2 nuclear waste research programme in 1994–1996, public administration's JYT2001 nuclear waste research programme in 1997–2001 and Finnish Research Programme on Nuclear Waste Management KYT in 2002–2005. Correspondingly, separate research programmes on reactor safety have been in progress.

This final report of the KYT2010 research programme presents the programme's objectives, organisation and research projects. The final report was edited by the Ministry of Employment and the Economy, with Jaana Avolahti as the contact person. Heikki Leinonen of Carrum Oy has participated in editing the entire report. Moreover, Marko Alenius (chapter 4.1), Esko Eloranta (chapter 6), Rainer Laaksonen (chapter 4.2), Katriina Labbas (chapter 5) and Paula Ruotsalainen (chapter 6) of the Radiation and Nuclear Safety Authority, and Ilpo Kallonen (chapter 4.3) of Fortum Power and Heat Oy have contributed to the editing of chapters 4, 5 and 6.

Project abstracts are compiled by those responsible for the individual research projects.

Helsinki, April 2011

*Ministry of Employment and the Economy
Energy Department*

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1 Introduction

The Finnish Research Programme on Nuclear Waste Management, KYT2010, was launched in 2006. The research period closed at the end of 2010.

The principles of the KYT2010 research programme are based on the Nuclear Energy Act (990/1987), according to which the aim of research activity is to ensure that the authorities have such sufficient and comprehensive nuclear engineering expertise and other facilities at their disposal that are needed for comparisons of the various ways and methods of carrying out nuclear waste management.

The Nuclear Energy Act emphasises the research needs of the authorities. Research programme contents strove to find research subjects considered key in terms of national expertise, which needed to be analysed due to their importance. Subjects that directly involve the preparation or implementation of nuclear waste management, or the inspection of such activities by the authorities, are excluded from the research programme, even though research serving the needs of authorities may simultaneously benefit parties bound by the obligation to manage nuclear waste. The research area was defined this way in order to ensure that participation in the research programme would not risk the independence expected from nuclear waste management actors (e.g. the Radiation and Nuclear Safety Authority STUK and Posiva).

The KYT2010 research programme has aimed at financing research that would strengthen the national competence base in the nuclear waste sector in terms of assessing safety from the strategic viewpoint, and in terms of reviewing social acceptability. Another objective was to contribute to ensuring the preservation and development of national scientific and technical expertise with regard to nuclear waste management. In this respect, the induction of a new generation of researchers into the topic area has been considered vital.

Moreover, the KYT2010 research programme has aimed at serving as a discussion and communication forum between authorities, organisations engaged in nuclear waste management and research institutions, thus seeking to create the preconditions for efficient utilisation of limited research resources in order to ensure a sufficiently diverse and interdisciplinary research team for individual research projects. Efficient information exchange has helped to avoid any overlapping research, as well as in coordinating international projects, for instance.

The KYT2010 Research Programme has also facilitated the implementation of projects based on joint funding by the National Nuclear Waste Management Fund (VYR) and other Finnish or international financiers. For instance, in most cases participation in EU projects requires national co-financing as well.

2 Research programme organisation

2.1 Objectives and their attainment

Key themes of the research programme include 1) strategic analyses of nuclear waste management, 2) assessment of nuclear waste final disposal safety and 3) sociological analyses. The assessment of the safety of nuclear waste final disposal comprises three sectors: engineered barriers, bedrock and groundwater, and the release and transport of radionuclides.

Guidelines specifying the research programme, compiled by the steering group on an annual basis, were introduced in 2007. First and foremost, the guidelines have specified the topic of assessing the safety of final nuclear waste disposal. The significance of engineered barriers was emphasised towards the end of the research period. Guidelines specified general objectives as well. For instance, in 2010, the project evaluation emphasised the objective of completing projects in progress prior to the end of the research period.

Research project assessment criteria have included 1) targeting and usability, 2) networking and integrability, 3) the educational impact, 4) scientific merit and 5) profitability shown in KYT projects or other contexts. The weighting coefficients of the assessment criteria are published in connection with the project search.

Targeting has been assessed in line with the objectives set out in the Nuclear Energy Act on the granting of research funding, and in relation to the annual guidelines of the research programme steering group. Usability has been assessed primarily in terms of the safety assessment of nuclear waste management, however it has been designed so as to facilitate the justification of other potential benefit to end-users in the assessment.

Networking and integrability have meant that research projects have been expected to network with actors in the field, to produce pooled, joint projects and integral entities.

The educational impact has taken account of both the quantitative (dissertations, theses, graduate theses) and the qualitative effect, which refers to creating expertise in Finland in key areas of expertise in nuclear waste management.

Publications and poster presentations etc. have been taken into account as having scientific value. Qualitative review has paid attention, i.e., to the type of research (experimental study, basic research, modelling), the degree of innovation (new arrangements for experiments, new techniques) and the extent (e.g. the scope of samples).

Productivity has assessed the progress of projects. Results achieved in other research contexts have been taken into account when assessing new projects.

A total of 44 research projects have been underway during the research period. They have been either separate and new projects or a continuance to projects previously carried out. Research projects have primarily been related to assessing the safety of nuclear waste management. The seven projects that have been underway throughout the research period have mainly been related to the release and transportation of radionuclides. In addition, a research project on partitioning and transmutation technology has been underway throughout the research period. Funding has been granted to sociological research since 2008.

The following number of research projects has been underway: 19 in 2006, 18 in 2007, 20 in 2008, 19 in 2009 and 24 in 2010. A list of research projects is attached.

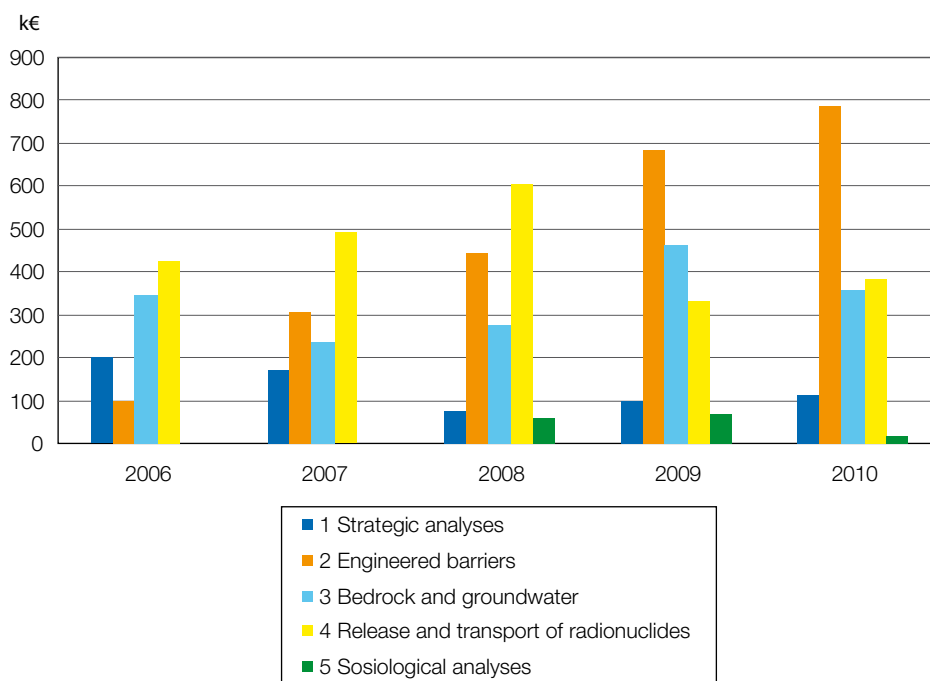
The State Nuclear Waste Management Fund allocated funding totalling approximately EUR 7 million to the research projects. Every year, more than one million euros were allocated to research.

Table 1. Distribution of VYR funding by topic.

Research area/EUR 1,000	2006	2007	2008	2009	2010
Strategic analyses	201	171	75	98	112
Engineered barriers	100	306	444	686	787
Bedrock and groundwater	345	236	276	461	358
Release and transport of radionuclides	425	493	606	330	384
Sociological analyses			60	70	20
Total	1071	1206	1461	1645	1661

At the beginning of the research period, 40 per cent of research funds were allocated to studying the release and transport of radionuclides, but towards the end of the research period, the focus transferred to engineered barriers.

Figure 1. Distribution of VYR funding by topic.



Projects within the research programme have published a total of 230 scientific articles or reports, etc. In addition, one database has been completed. The names of publications are reported in the annual reviews of the research programme ^{1 2 3 4}.

Table 2. Number of publications by topic 2006–2010.

Publications by topic/number	2006	2007	2008	2009	2010
Strategic analyses	6	10	4	0	8
Engineered barriers	3	7	15	25	13
Bedrock and groundwater	18	7	0	5	14
Release and transport of radionuclides	12	14	22	16	12
Sociological analyses			4	7	8
Total	39	38	45	53	55

1 The Finnish Research Programme on Nuclear Waste Management, KYT 2006–2010. Annual review 2006. (in Finnish only)

2 The Finnish Research Programme on Nuclear Waste Management, KYT 2006–2010. Annual review 2007. (in Finnish only)

3 The Finnish Research Programme on Nuclear Waste Management, KYT 2006–2010. Annual review 2008. (in Finnish only)

4 The Finnish Research Programme on Nuclear Waste Management, KYT 2006–2010. Annual review 2009. (in Finnish only)

A total of 27 theses have been completed through the research projects within the programme. Research projects have also reported on theses in progress, but these are not taken into account in the figures below.

Table 3. Number of theses by topic 2006–2010.

Theses by topic/number	2006	2007	2008	2009	2010
Strategic analyses	0	1	0	3	0
Engineered barriers	1	1	5	0	3
Bedrock and groundwater	1	0	0	2	1
Release and transport of radionuclides	2	0	4		3
Sociological analyses					
Total	4	2	9	5	7

The following research organisations have participated in the KYT2010 research programme: VTT – the Technical Research Centre of Finland, the Aalto University School of Science and Technology, University of Helsinki Laboratory of Radiochemistry, the Geological Survey of Finland, University of Jyväskylä, University of Eastern Finland Kuopio Campus, and Numerola Oy.

2.2 Research programme evaluation

The research programme evaluation was implemented between 29 October and 2 November 2007. The evaluation team reviewed research programme documents and interviewed members and deputy members of the research programme steering group and support group, other key persons and project managers of research projects. Results included general conclusions, replies to evaluation questions, challenges and recommendations. A separate evaluation report is published in the Ministry’s publication series.⁵

The research programme steering group reviewed evaluation results systematically in late 2007 and early 2008. In 2008, the steering group decided on a number of development measures related to procedures, research project monitoring, nuclear waste management training, centres of expertise and safety analysis methods.

Operating instructions⁶ were compiled and introduced for the selection of research projects. In 2008, a mentor programme was launched for the purpose of research project monitoring, comprising free-form visits with each project. The development of reporting has been postponed and will take place in subsequent programme periods.

5 Apted, M., Papp, T. & Salomaa, R. 2008. KYT2010 Review Report. Publications of the Ministry of Employment and the Economy, Energy and Climate 2/2008, 26 p.

6 Varjoranta, T. 2009. Operating instructions for the KYT2010 programme (in Finnish only). <http://www.ydinjatutkimus.fi/tiedostot>, 10 p. + appendices.

Steering group guidelines for the selection of research projects were introduced in 2007. In 2008, the guidelines specified research needs particularly related to alternatives in a more detailed manner than before.

National nuclear waste management education and training and centres of excellence were handled as special issues by working groups in 2008–2009. Nuclear waste management education and training possibilities and needs were mapped in an analysis conducted by a working group. Based on the initial analysis, the steering group recommended a further analysis, which comprised a tailored course in nuclear waste management, implemented as a pilot project. The centre of excellence analysis defined the organisation of nuclear waste management research.

To enhance interaction, thematic seminars and a mid-term seminar were organised.

2.3 Research programme administration

Work in the research programme has been based on mutual cooperation and the division of duties between the research programme steering group, two support groups, a coordinator and research projects. The research programme steering group has convened regularly, three or four times a year. Research programme support groups have convened once or twice a year.

The steering group has been responsible for research programme strategic policies and has served as the body which coordinates research programme administration and the general research guidelines. The steering group has also guided research programme planning and has monitored the quality of research results. The group has compiled annual recommendations to the Ministry on targeting of VYR funding to nuclear waste management research.

The support group has conducted detailed annual assessments of the project proposals and prepared an annual draft financing proposal for the steering group on the basis of its evaluation. The support group has monitored the progress of research projects. Separate support groups have been managing technical-scientific and sociological research.

In 2006–2008, Carrum Oy, and in 2009–2010, the Ministry of Employment and the Economy have been in charge of research programme coordination.

Representatives of the Ministry of Employment and the Economy, the Radiation and Nuclear Safety Authority, Tekes – the Finnish Funding Agency for Technology and Innovation, and organisations in the nuclear waste management industry were appointed as members of the research programme steering group. Tero Varjoranta (Radiation and Nuclear Safety Authority STUK) has been the chairman of the research programme. Other steering group members are Pia Salokoski (Tekes), Harriet Kallio (Fortum Power and Heat Oy, until 10 January 2008), Sami Hautakangas (Fortum Power and Heat Oy, from 10 January 2008), Jari Tuunanen (Teollisuuden Voima Oyj, until 10 January 2008), Veijo Ryhänen (Teollisuuden Voima Oyj, from 10 January

2008), Juhani Vira (Posiva Oy), Anne Väätäinen (Ministry of Employment and the Economy, until 22 August 2006), and Jaana Avolahti (Ministry of Employment and the Economy, from 22 August 2006).

Deputy members were Kaisa-Leena Hutri (Radiation and Nuclear Safety Authority STUK), Piia Moilanen (Tekes), Ilpo Kallonen (Fortum Power and Heat Oy), Anneli Reinvall (Teollisuuden Voima Oyj), Tiina Jalonen (Posiva Oy) and Jorma Aurela (Ministry of Employment and the Economy).

The steering group has appointed support group members from the Radiation and Nuclear Safety Authority STUK, Ministry of Employment and the Economy, Fortum Power and Heat Oy, Posiva Oy and Teollisuuden Voima Oyj.

2.4 Contacts

2.4.1 Seminars

For information exchange within the KYT2010 Research Programme, a total of eight seminars were arranged, while research programme themes were handled at six thematic seminars. They focused on one topic at a time, and presented relevant viewpoints from the perspective of research institutions and end-users of results. The research programme and all its topics and research projects were handled at the mid-term of the programme period and at seminars arranged at the end. Two of these seminars were held.

Several stakeholders were informed about the seminars, which were open to all interested parties. The seminars, usually attended by 30–60 people, attracted participants from outside the research programme, too. Table 4 shows a list of the seminars.

Table 4. KYT2010 research programme seminars.

Seminar topic	Time	Location
Bedrock hydrogeology	17 November 2006	Geological Survey of Finland, Espoo
Engineered barriers	8 May 2007	Helsinki University of Technology, Espoo
Radionuclide transport in bedrock	15 February 2008	University of Helsinki, Laboratory of Radiochemistry
The Long Term Diffusion (LTD)	8 May 2008	University of Helsinki, Laboratory of Radiochemistry
Programme period mid-term seminar	26 September 2008	Diana Auditorium, Helsinki
Bentonite and filling material	29 September 2009	VTT – the Technical Research Centre of Finland, Espoo
Canister copper	29 April 2010	Aalto University, School of Science and Technology, Espoo
Final seminar of the programme period	18 March 2011	Sähkötalo Auditorium, Helsinki

2.4.2 Mentor activity

To facilitate the monitoring of research projects, mentor activity was introduced as part of the research programme in 2008. Each research project had a team of mentors, to which authorities and nuclear waste management organisations appointed representatives. The team monitored and steered the progress of the project by maintaining contacts with those implementing the project, and by visiting the research institutions. The aim is to hear the opinions, views and wishes of researchers and to highlight the needs and wishes of end-users in terms of the contents and focus of the projects.

In 2008, 13 mentor visits were made to various research institutions. In 2009, the number of visits was 16, and there were 22 in 2010.

2.4.3 Other contacts

The research programme website (www.ydinjatutkimus.fi) has been the primary medium for contacts and communication. All material published by the research programme is available on the website.

The research programme has published annual plans and annual reviews every year. Interim reports have been compiled in 2006–2008 (3 per year). Bulletins have been prepared to communicate the decisions made in the steering group meetings.

3 Strategic analyses of nuclear waste management 2006–2010

In the research programme, strategic analyses refer to analyses related to the alternative methods of implementing nuclear waste management, the nuclear fuel cycle or ensuring expertise. General safety issues, for instance those related to radiation protection viewpoints, also form part of the analyses in question.

The objective of analysing alternative methods of nuclear waste management has been to support the reliability of nuclear waste management implementation. The analyses have also aimed at ensuring the competence base in implementing nuclear waste management, and monitoring the debate on alternatives and their development. International discussion on regional fuel cycle centres and enhancing the efficiency of nuclear non-proliferation control and reprocessing has characterised the research programme period. Issues related to the basic policies of nuclear fuel cycle and nuclear waste management have also included analyses related to future options, such as partitioning and transmutation or long-term storage.

The research programme has financed four projects, of which one has been running throughout the research period. Other projects lasted one or two years. Strategic analyses have augmented know-how on nuclear waste management and optional ways of implementing it. The projects have monitored international development or participated in international programmes. A separate project on ensuring expertise in nuclear waste management has also been in progress. In connection with this project, a pilot course was implemented to secure special training in nuclear waste management.

The projects have cooperated in particular with organisations based in Finland and have collaborated as a network. Key actors in the industry, and authorities, contributed to implementing the pilot course.

4 Research into engineered barriers in the 2006–2010 research programme

Research into engineered barriers in the research programme is mainly related to assessing the safety of disposal. A canister, a buffer made of bentonite, and bentonite raw material have been studied as engineered barriers. The research programme has also engaged in other research related to engineered barriers.

4.1 Research on final disposal canisters

The long-term safety of the final disposal concept must be based on safety operations achieved through barriers that complement each other, and the final disposal canister is the most important single barrier in the KBS-3 concept. The most important safety function of the final disposal canister is long-term isolation of spent nuclear fuel from the environment. Government decree 736/2008 on the safety of disposal of nuclear waste and the Radiation and Nuclear Safety Authority's draft YVL guides D.3 and D.5 impose requirements i.e. on the design, manufacturing, safety functions, performance targets and the long-term safety of the final disposal canister.

With regard to the long-term safety of spent nuclear fuel disposal, the final disposal canister's mechanical durability and chemical resistance are both key targets of analysis and research. Several projects related to the long-term integrity and safety analysis of the final disposal canister have been in progress in the KYT2010 research programme: three projects studying the mechanical durability of the final disposal canister, and four related to its chemical resistance.

Government decree 736/2008 and the draft YVL guide D.5 state that compliance with the requirements concerning long-term radiation safety, and the suitability of the disposal method and disposal site, shall be proven through a safety case. The Government decree states for instance the following on the reliability of the safety case: the input data and models utilised in the safety case shall be based on high-quality research data and expert judgment. In the view of the authorities, projects related to the mechanical and chemical resistance of the final disposal canister, implemented in the KYT2010 research programme, have contributed to producing independent high-quality research data that can be utilised in inspection and assessment tasks conducted by authorities.

4.2 Research into bentonite related to the buffer and tunnel filling

The final disposal concept KBS-3, chosen for implementation in Finland, is based on the safety principle of defence-in-depth, formed by barriers complementing each other, required by safety regulations.

The canister is the most important barrier, protected and isolated from the environment by another barrier, a buffer made of bentonite. Government decree 736/2008 on the safety of disposal of nuclear waste and the Radiation and Nuclear Safety Authority's draft YVL guide D.5 specify the requirements set by authorities on safety functions and performance targets for the buffer.

The primary objectives of the buffer are, first, to protect the canister mechanically and chemically, providing containment and yielding minor rock movements, and protecting it from external chemical stress, or harm caused by microbial activity. On the other hand, the aim of the buffer is to provide containment by isolating the canister from its environment and to reduce and slow-down the transport of radionuclides through the buffer to ground level through the surrounding rock and bedrock, caused by any unlikely damage to the canister.

The buffer forms part of a multiple barrier system. Its interaction and collaboration, on the one hand with the canister and on the other, with the rock surrounding the deposition hole and the tunnel filling material, must also be managed in terms of the materials and technical properties at different times. In addition to the buffer, bentonite or other types of swelling clay will probably be utilised in the filling materials of the final disposal tunnel, and other sealing structures of excavated facilities.

Posiva, STUK and research communities and experts in the field have identified significant needs for further information related to the mineralogical and technical properties of bentonite. In order to prove that the performance targets of the buffer can be met, these issues must be analysed. The KYT programme has also focused on extensive analysis of and research into these aspects. The Radiation and Nuclear Safety Authority, too, has participated in defining the objectives of the programme and the monitoring of projects to a varying extent.

Work performed within the KYT research programme has produced, and will continue to produce, information suited to the needs of the authorities. This information is parallel and supplementary, applied and contains basic research. In connection with this research related to bentonite, new experts have also been trained on the topic, national networks have been forged and national research capacities have been developed. In the next programme, more extensive work will continue.

4.3 Other research objectives related to engineered barriers

In research into disposal safety, the disposal of low and intermediate level waste has been included in the research programme. One of the aims has been to predict the behaviour of reinforced concrete structures serving as sealing structures.

The research programme has included one project on this topic. It has created competencies in applying and developing models for the interaction of reinforced concrete ageing mechanisms. These models can be utilised in assessing the ageing of reinforced concrete structures in the disposal conditions of operating waste.

5 Bedrock and groundwater in the 2006–2010 research programme

Research into bedrock and groundwater is related to the assessment of disposal safety in the research programme. Long-term safety of nuclear waste final disposal must, in accordance with safety requirements, be based on functional safety achieved through barriers that complement each other.

Bedrock serves as a natural barrier, if the performance targets set for bedrock are met. In the immediate vicinity of the final disposal canister, the bedrock must be stable and compact, and the flow of groundwater must be low. The host rock surrounding final disposal facilities must have low flow of groundwater, and reducing and favourable groundwater chemistry in other respects, too. Aspects indicating unsuitability as a final disposal site may include abnormally high rock stresses in relation to the strength of the rock and exceptionally adverse groundwater characteristics, such as the lack of reducing buffering capacity. The final disposal depth must be selected favourably with regard to long-term safety, taking account of the geological characteristics of bedrock and changes in hydraulic conductivity, groundwater chemistry and the trends in rock stress ratio due to depth.

The prospective final disposal site shall contain sufficiently large, solid rock volumes that facilitate the construction of the final disposal facilities. When constructing the final disposal facilities, any such structures and other characteristics of the host rock that may be significant in terms of groundwater flow, rock movements or other aspects influencing long-term safety must be defined and classified.

When excavating and constructing final disposal facilities, the bedrock must retain, as far as possible, all characteristics that are vital with regard to long-term safety. When implementing the final disposal of spent fuel, the methods adopted for construction must be those that limit any disturbance due to excavation in the host rock of the final disposal repository.

The need of authorities to include bedrock and groundwater research projects in the KYT2010 research programme is based on the Government decree 736/2008 on the safety of disposal of nuclear waste, and YVL guides. Projects in the KYT2010 research programme concerning bedrock and groundwater research have been justified from the viewpoint of authorities.

These bedrock and groundwater research projects in the KYT2010 research programme have generated new competence in nuclear waste research and theses are to be expected, too, in line with the objectives of the framework programme. From the viewpoint of authorities, projects in the KYT2010 research programme were to produce independent information that could be utilised in the assessment of work performed by licence applicants and licence holders. Project results can also be utilised in the implementation of final disposal.

6 Release and transport of radionuclides in the 2006–2010 research programme

Research into the release and transport of radionuclides is related to the assessment of disposal safety in the research programme. According to safety requirements, the long-term safety of nuclear waste disposal shall be based on safety functions achieved through mutually complementary barriers so that a deficiency of an individual safety function or a predictable geological change will not jeopardise long-term safety (Government decree 736/2008; YVL D.5). According to the KBS-3 concept, bedrock forms a natural barrier to the transport of nuclides. Safety functions are specified for the bedrock, related i.e. to groundwater chemistry and restricting the movement of radioactive substances by retarding nuclides on rock surfaces and in pores (YVL D.5).

Analysis of the release and transport of radionuclides is integral to the assessment of disposal safety. Analyses are related to scenarios of nuclear waste containers in the final disposal facility being damaged so that radionuclides are released from them to the buffer and the host rock. Government decree 736/2008 on the safety of disposal of nuclear waste contains separate provisions on the maximum values for radiation impacts on ground level permissible in such situations, over a period of time extending over several millennia. This period of time is one during which radiation exposure caused to humans can be assessed with a sufficient degree of reliability. Certain nuclide-specific maximum limits are specified for release into the environment after this period of time. These are based on long-term average values of the quantities of radioactive materials released. These values were specified following the principle that at a maximum, radiation impacts caused by final disposal can be equivalent to those caused by natural radioactive materials in the earth's crust and on a large scale, the radiation impacts remain insignificantly low. The Radiation and Nuclear Safety Authority separately specifies the maximum values for each radionuclide, listed for instance in the Radiation and Nuclear Safety Authority's draft YVL Guide D.5.

Specification of the quantities of radioactive substances released through barriers into the environment from waste to be placed in disposal, and the resulting radiation doses forms a key part of the safety case.

The KYT₂₀₁₀ research programme included almost 15 projects which gave, directly or indirectly, results and conclusions related to the release and transport of radionuclides. These are significant not only nationally but internationally, too, because certain projects have conducted pioneering research. These projects have enabled Finnish researchers to join international research projects. For authorities,

the results provide versatile reference data on the conditions and properties of crystalline bedrock. The projects have generated new expertise, acquired new equipment and trained new experts in the field by way of theses. Research projects in the KYT2010 research programme on the release and transport of radionuclides have been justified from the viewpoint of authorities.

7 Sociological analyses in the 2006–2010 research programme

The purpose of sociological analyses in the research programme is to support licensing procedures in Finnish nuclear waste management, one component of which is the issue of the acceptability of nuclear waste management and a final disposal facility for spent nuclear fuel in particular. Since 2008, sociological research related to nuclear waste management has constituted a focus area in terms of content that has been highlighted in the guidelines prepared by the research programme steering group.

Between 2008 and 2010, the research programme financed a sociological research project that monitored changes in attitudes towards final disposal, and analysed needs and methods of information acquisition regarding final disposal of nuclear waste in Eurajoki and neighbouring municipalities. The research programme project also covered the views of inhabitants of neighbouring municipalities to Eurajoki, while previous studies have primarily focussed on the views of Eurajoki inhabitants.

The results lend themselves to use as part of decision-making and comparative assessments, as largely similar studies, commissioned by other parties, have been conducted simultaneously. Sociological analysis contributes to enhancing international knowledge of how Finland has managed to proceed with the final disposal of nuclear waste, and how extensively accepted these operations are, nationally and locally. Sociological research is internationally significant and networked and results are published in international scientific publications in the field.

8 Abstracts of projects

8.1 Finnish research project on partitioning and transmutation

Markku Anttila, VTT

ORGANIZATION

The project received the VYR funding during the whole KYT2010 period (2006-2010). It was based on co-operation between VTT and HYRL (The Radiochemistry Laboratory of the Department of Chemistry of the University of Helsinki). The main goals of the project were to follow the progress of the international R&D on partitioning and transmutation and to increase our own expertise on some specific subfields. Within the project VTT focused on the reactor physics of transmutation systems and HYRL studied partitioning (separation) techniques, especially the treatment of secondary wastes.

ANALYSIS OF TRANSMUTATION SYSTEMS

To perform reliable assessments of the benefits of various transmutation options it is necessary to be able to calculate the material balances of the systems reliably enough both during the introductory phase of a new cycle and at a more or less stable equilibrium phase. These data are needed when the benefits of advanced fuel cycles are assessed, especially when the transition from one fuel cycle to another is studied.

The reactor physics of fast transmutation systems

VTT has tried to develop a capability to analyze fast reactors during the past few years. The ERANOS code was initially developed when the so-called European Fast Reactor was designed in the 1990s. VTT acquired first the ERANOS2.0 version from OECD/NEA Data Bank and then made a license agreement with CEA, under which VTT received the latest version of the code (ERANOS2.2) with its updated data libraries. Most of the ERANOS related work at VTT has been carried out outside the project, but the project has supported the completion of a special assignment and a diploma thesis. At VTT there are some other computer codes, which could be used for analyses of fast reactors. Two codes based on the Monte Carlo technique, Serpent and MCNP5/MCNPX, are perhaps their best examples. Serpent can be used to produce group constants for three-dimensional calculations of the cores.

Within the project two literature reviews related the reactor physics of subcritical systems were carried out. The first one concerned the computer codes used for the calculations of subcritical reactors and the second one the methods, which could

be used for the measurements of subcriticality. Both questions have been studied extensively in the R&D programs of the accelerator-driven subcritical system (ADS). Regarding the computer codes, the practical and largely accepted starting point is that the codes developed assuming the criticality like ERANOS can reliably be used for analysing subcritical systems, even if all the assumptions are not exactly fulfilled. There are also accurate enough methods in order to calculate the external neutron source of the ADS. During the past two decades much effort has been devoted to improve nuclear data needed for describing the interaction between high-energy protons and the target material and the subsequent behaviour of energetic neutrons produced within the spallation source.

The theoretical basis, as well as practical procedures, for the measurements of the reactivity of subcritical systems was developed already in the 1950s. However, the methods were not suitable as such for operating ADSs. The theoretical assumptions were no more valid and some of the measurement procedures could not be applied in ADS cores. With extensive R&D activities good progress has been achieved, but due to the lack of large enough test facilities the validation of the methods is difficult.

Minor actinide management in the LWRs

The so-called minor actinides (MA = neptunium, americium and curium) have a large impact on the radioactive properties of the spent nuclear fuel and especially of high-level nuclear waste after a cooling period of 60–80 years. From the reactor physics point of view, it is obvious that a fast reactor can burn minor actinides more effectively than a LWR. However, a large-scale introduction of fast reactors will not happen in a near future. Therefore, quite many studies have been performed for assessing the possible role of the LWRs in the minor actinide management. Calculations show clearly that also the LWRs can burn at least neptunium and americium. Curium is a much more difficult case, because its isotopes can be transformed by the neutron capture into more difficult radionuclides. Preliminary analyses carried out at VTT with CASMO and SIMULATE-3 codes produced similar results. To be able to get more accurate results for fuel cycle analyses VTT has decided to acquire a special SIMULATE-3 version from Studsvik Scandpower, Inc.

ANALYSIS OF THE NUCLEAR FUEL CYCLES

During the past decade several computer codes have been developed for time-dependent analyses of nuclear fuel cycles. Such tools are necessary for instance, when the transition from one fuel cycle to another should be designed and maybe optimized. Some of the new codes are quite complicated including even modules, which can be used to assess the impact of social factors, proliferation resistance etc. A literature survey was carried out in the project. Based on its conclusions and some earlier decisions (ERANOS) negotiations were started with CEA in order to acquire the COSI6 code, which has been developed by CEA and Areva. COSI6 has been widely used in international fuel cycle studies.

PARTITIONING

Review of R&D on partitioning techniques

During the first two years of the KYT2010 program HYRL continued to follow and assess the progress of international R&D programs on partitioning. The amount of secondary waste and the methods for the treatment of secondary waste were the focus of the studies. First, pyrochemical processes and then in 2007, the so-called group extraction methods were studied.

New methods for separation of actinides and lanthanides

Based on literature reviews carried out in project and other existing expertise HYRL started a small-scale experimental study on Adsorption and Desorption of ^{141}Ce , ^{152}Eu and ^{241}Am for Metal Antimonates.

Partitioning of radionuclides from used fuel for transmutation generates liquid wastes which contain the residue of fission products and TRU. These can be removed by using selective inorganic adsorbents. Metal antimonates (MeSb) are hydrous mixed metal oxides which capacity of adsorption is based on chemical nature and quantity of Me (ratio Me/Sb). In the laboratory of radiochemistry have been synthesized several MeSb-compounds. From these compounds was chosen a representative number of MeSbs. An adsorption of ^{152}Eu and ^{241}Am for MeSbs was tested in nitric acid solution. Many MeSb-compounds had a very high distribution factor (K_d) which quantifies the adsorption. For further tests were selected tungsten doped antimony silicates (WSbSi) and antimony silicate (SbSiO). Adsorption and desorption of ^{152}Eu and ^{241}Am for WSbSi and SbSiO were tested by determining the K_d -value and a discrimination factor ($K_d(\text{Am})/K_d(\text{Eu})$) on an acidic pH area and in mineral acids and in organic acids which commonly used complex forming reagents in partitioning. Cerium, ^{141}Ce , which represents light lanthanides, was tested by SbSiO.

WSbSi and SbSiO had very high K_d -values (10^5 – 10^6 mL/g) for ^{141}Ce , ^{152}Eu and ^{241}Am on the pH area from 1 to 3. Also all organic complexing agents had a concentration area where their adsorption was close to a detection limit. In a WSbSi-column test was observed very high a processing capacity for ^{152}Eu and ^{241}Am in a trace concentration. Based on the results WSbSi and SbSiO are excellent materials for secondary liquid wastes management. WSbSi takes also cesium and strontium. In a chromatographic separation of ^{152}Eu and ^{241}Am by WSbSi and SbSiO, the best solution was 2 M ortho-phosphoric acid and 0.3–0.4 M nitric acid. The organic acids had deficient desorption and the separation capacity of nuclides from WSbSi and SbSiO although there were differences between some discrimination factors. Cerium behaved almost identical with americium. In the case of a repository, the poor desorption of Ln/Am-activity from antimony silicates is benefit for a sorption material in management of radioactive waste.

EXCHANGE OF INFORMATION

One of the main goal of the project was to inform the Finnish stakeholders of the status and progress of international R&D programs on partitioning and transmutation. In 2006 a seminar was arranged at VTT, where Dr. Janne Wallenius from the Royal Institute of Technology gave a presentation with the title “Status of R&D programs on Generation IV reactors and transmutation.” In the same year a short overview of an extensive NEA study on advanced fuel cycles was written. Afterwards the exchange of information has been carried out mostly in national seminars. Expertise of the project has been utilized also in a few studies funded by Posiva Oy.

8.2 Scientific and technical basis of nuclear waste management (TEPE)

Kari Rasilainen, VTT

The project was active from 2006 to 2007 and it was part of an earlier started long-term work on the development on safety analysis methodology for nuclear waste management. Technically speaking, the project focussed on (1) international and national collaboration aiming at developing safety analysis methodology for spent fuel disposal, (2) national collaboration advancing the use of palaeohydrogeological research results in the safety case of spent fuel disposal, (3) cost risk assessment of nuclear waste management, and (4) general radiological studies of nuclear fuel cycles.

MAIN RESULTS OF THE PROJECT

Subproject 1

The subproject has participated in the work of two relevant nuclear waste management expert groups of the OECD/NEA, namely RWMC (Radioactive Waste Management Committee) and IGSC (Integration Group for the Safety Case). Our work has covered meeting-related activities, participation in the preparation of the annual country reports for the RWMC, and attending relevant workshops organised by the expert groups. In Finland the subproject has participated actively in the work of nuclear waste safety section (now called nuclear waste safety committee) of the Nuclear Safety Advisory Body (YTN, now called Nuclear Safety Advisory Commission). The subproject has furthermore participated actively in the national user group meetings of the EU project ERICA (Environmental Risk from Ionising Contaminants: Assessment and Management).

Subproject 2

The subproject has participated actively in the domestic collaboration aiming at utilising the palaeohydrogeological research in safety case. Main participants in this collaboration have been VTT, the University of Helsinki, Laboratory of Radiochemistry and the Helsinki University of Technology (TKK, now called Aalto University).

The main aim of the subproject was to consider how USD studies (Uranium Series Disequilibrium) done for crystalline bedrock can be utilised in the Finnish safety case for spent fuel disposal. There is namely a “knowledge zone” between primary observations and safety case in which understanding of the redox-behaviour of bedrock is accumulating. It is this understanding that can be used in the components of the safety case dealing with, e.g. bedrock, mass flows, processes, and alternative safety arguments. Individual USD observations must be integrated in this understanding before they can be utilised in the safety case.

Measured USD data (e.g. activity ratio U-234/U-238) for water samples taken at different sites were used in an effort to test their applicability in characterising the respective flow systems (Suksi et al. 2006). Studies of water samples taken at Palmottu uranium deposit indicated that direct alpha-recoil of U-234 probably is not enough to alone explain the disequilibria observed (Rasilainen et al. 2006). Alpha-recoil is a physical release mechanism and the conclusion means that the release mechanism that could explain the observed disequilibria must be chemical in nature (assuming that the possible release mechanisms are either physical or chemical). Alpha-recoil will undoubtedly take place but more abundant U-234 release would require a chemical release process and the measured radioactive disequilibrium in groundwater represents both physical and chemical release.

In collaboration with the University of Helsinki, Laboratory of Radiochemistry (HYRL) the subproject has prepared a laboratory experiment to mimic the redox change induced uranium release in bedrock caused by oxygen-rich glacial meltwater.

Subproject 3

The subproject applied Monte Carlo simulation in studying cost risk characteristics of nuclear waste management. A model was constructed for cost risk assessment and preliminary simulations were done (Forsström 2006). The results obtained link costs with uncertainty thus increasing understanding of risk characteristics and advancing the estimation of liabilities. The main aim of the subproject was to test and demonstrate the usefulness of the risk analysis method in cost risk estimation.

The subproject also covered the annual statement of VTT for the Ministry of Industry (KTM), now called Ministry of Employment and the Economy (TEM), on the cost estimates that nuclear waste producers had to present annually for the ministry.

Subproject 4

The subproject has participated in strategic projects dealing with the radiological impacts of various nuclear fuel cycles, international and national. A report "Developed nuclear fuel cycles and the nuclear waste management" (Anttila & Rasilainen 2007) has been written in collaboration with Markku Anttila (VTT). The report briefly reviews a working group report on nuclear fuel cycles by the OECD/NEA (OECD 2006) with focus on parts interesting from Finnish point of view. The interesting parts discuss safety assessments of high-level wastes from various nuclear fuel cycles that are hypothetically disposed of in crystalline bedrock. These safety assessments have subsequently been compared with those for spent fuel disposal in crystalline bedrock.

THE MEANING OF THE RESULTS FOR NUCLEAR WASTE MANAGEMENT RESEARCH

The meaning of the results depends on the subproject. In subproject 1 we could communicate the topics in the OECD/NEA discussion to Finland and, vice versa,

communicate the situation in Finnish nuclear waste management to the expert groups of the OECD/NEA. In subproject 2 we paved the way for a proper way to integrate results from palaeohydrogeological research, e.g. those of USD studies, into safety case. In subproject 3 a model for cost risk estimation of nuclear waste management was developed. In subproject 4 an OECD/NEA working group report was reviewed with emphasis on topics relevant from Finnish point of view.

METHODS USED

The methods used were meetings and research, including mathematical modelling.

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8.3 Review on environmental remediation techniques of uranium mining and milling tailings

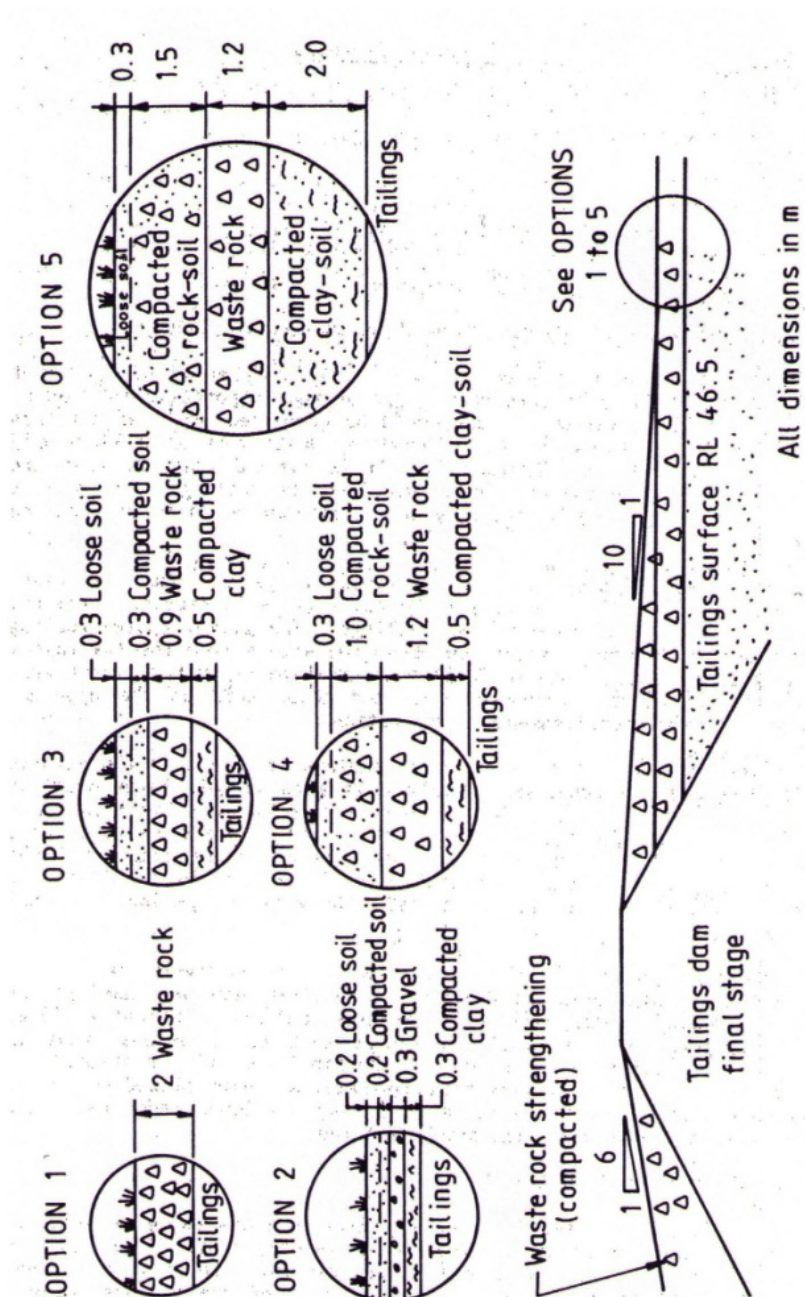
Vesa Suolanen, VTT

ESSENTIAL RESULTS

Based on known or planned solutions, remediation techniques for uranium mine wastes – host rock and milling tailings – were considered in the research project.

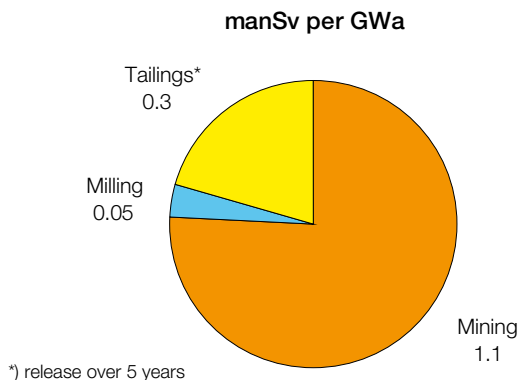
The main isolation methods of milling tailings are overground, underground or immersion in water disposal. A cave excavated underground or exploitation of e.g. an abandoned pit, could be acceptable solution, based on probably long-term stable expected conditions. Processing of milling tailings and possibly additional isolation of pit surfaces have to be prepared before starting disposal activities of tailings into a pit. The milling tailings have to be solidified by a proper industrial method and dispose further into a facility or a pit from which there is no connection to organic nature. In all cases, erosion of tailings material has to be prevented, and on the other hand, also tailings dissolution and transfer to surface waters or into surface soil (Figure 2).

Figure 2. Tailings isolation options by soil, clay and gravel (OECD).



Occupational individual effective doses from uranium mining activities are on the same level as normal background radiation dose rate, i.e. around 3 mSv per year per worker. Normalised effective dose to the public is around 1 manSv/GWa (Figure 3).

Figure 3. Collective doses to the public from uranium mining, milling and tailings per annual energy production of a 1000 MW nuclear power plant (manSv/GWa) (OECD/NEA & IAEA). The corresponding fuel amount used is around 200 tU/year.



SIGNIFICANCE AND EXPLOITATION OF RESULTS

In the study, applicability of various remediation methods for mine wastes have been considered, particularly how radiation doses to the public could be cost-effectively minimised on the long-term. Selected processing methods for milling tailings and waste disposal option have to be based on strategically sustainable solutions.

USED RESEARCH METHODS

The study has been performed as applied literature research work. As essential reference material publications from International Atomic Energy Agency (IAEA), from Nuclear Energy Agency of Organisation for Economic Cooperation and Development (OECD/NEA) and from United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) were used.

8.4 Coordination of the nuclear waste management university education and further training

Mira Markovaara-Koivisto, Aalto University

FOCAL RESULTS OF THE RESEARCH

Aim of the project was to make a draft plan of the training contents for a national-level nuclear waste education and to make a summary of the potential content providers and of funding alternatives.

Work started on 11.6.2010 with a work group meeting steered by the Ministry of Employment and the Economy (TEM). The working group members were Jaana Avolahti (TEM), Kaisa-Leena Hutri (STUK), Marjatta Palmu (Posiva Oy), Mira Markovaara-Koivisto and Jussi Leveinen (Aalto University). It was decided that the education plan is reported in written form as part of the project report.

Representatives or organizations, which were potential to provide content to the course were invited to the expert working group. Sami Hautakangas represented Fortum Power and Heat Oy, Juhani Hyvärinen represented Fennovoima Oy, Anssu Ranta-aho represented Teollisuuden Voima Oyj, Kari Rasilainen represented VTT and Mikael Rinne represented Aalto University. They were told in more detail the purpose of education, its intended content, and of a shorter pilot course, which could be organized possibly already in 2010.

Framework for the contents of the pilot course was set by the draft training plan, but the precise content was determined according to the subjects of lectures, they had to offer on such short notice. From these lectures a two days course was compiled to satisfy all the parties. VTT offered to provide ca. 4-hour guided tour of the Otaniemi laboratory facilities and the nuclear research reactor.

Number of participants attending the pilot course on nuclear waste management was limited to 21 participants because of the limited space on the guided tour to VTT. The course was advertised within the organizations of the content providers, and 20 persons from the registered 21 attendees were able to attend the course.

The course had to be divided into two separate days because of scheduling of the guest speakers. The first day of the lecture was held at the Aalto University on November 23, 2010. The tour to VTT's laboratories and the nuclear research reactor was held on December 1, 2010 from 13.30 to 17.00 hrs. The second lecture day was on December 2, 2010. At the end of the second lecture day the participants filled a course questionnaire.

In the future, this course is planned to be held annually at Aalto University. The aim is to extend the course duration to 5 days. In this way all the topics listed in the draft training plan can be covered. The course will be run in parts of shorter duration to facilitate participation on the course.

During the project the future funding opportunities of the education were discussed:

- KYT2014-program
- Production of the course content jointly according to the YK-model by different actors in nuclear waste management
- Charging the coordination costs directly from nuclear power companies
- A course fee, which is proportional to the in-kind lectures produced by each participant's home organization.

SIGNIFICANCE AND EXPLOITABILITY OF THE RESULTS

During the project a 2.5-day pilot course on the basics of nuclear waste management was held. During the course's organization and course days, the course coordinator Mira Markovaara-Koivisto collected experiences on organizing such a course in practice. The participants filled a feedback questionnaire concerning the course's success. These lessons will be exploited during the next course that introduces the nuclear waste management sector to the future experts.

During the project 20 new experts in the field of nuclear waste management got familiar with the basics of nuclear waste management, organizations in the field and related legislation. During the course, the participants got to know each other, the lecturers and organizations they represented. At the end of the course a mind map was handed out to the participants to support networking. The mind map included all the participants, lecturers, the members of the working group, and their integration in the field of nuclear waste management.

An important result in the project was the setting up the planning working group from the experts in the field and taking into account their opinions when considering the contents of the training. As the course is held annually, the contents of the course may be modified, and the working group can gain more members. Also the list of the lecture providing organizations must be extended as the contents of the course expands. In addition, the course must be informed broader in order to obtain all of the new experts to participate the education.

RESEARCH METHODS

The project did not use any validated research methods in the planning, but the draft training planning was carried out by a collaborative expert group. The function of the draft plan was tested by implementing a pilot course and the pilot course was evaluated by using a structured and tailored feedback questionnaire that was adopted from Aalto University's course feedback form.

8.5 Long-term integrity of copper overpack

Juhani Rantala, Jorma Salonen, Pertti Auerkari, Stefan Holmström, VTT
Tapio Saukkonen, Aalto University

Changes in base material and FSW welds of OFP copper have been investigated after low temperature (150–175°C), low stress (35–120 MPa) creep experiments. The observed changes in the multiaxial (CT) specimens tested up to 30 000 h (3.4 years) at 175°C appear to be largely restricted to widening recovery zones at stressed grain boundaries and to increasing grain boundary cavitation (Fig. 4) that had first emerged after 15 000 h of testing at the natural (joint) notch tip. The cavitation damage appears to be related to the combined local strain and stress state in front of the notch/crack tip. The results from CT tests and earlier tests with nominally uniaxial but defective specimens suggest that multiaxiality is important in controlling and limiting creep life. In comparison, fast evolving intergranular creep damage, crack branching and low ductility was confirmed in pure (OFHC) copper in CT specimens tested up to 10 400 h (1.2 years), while much higher creep ductility has been retained in OFP copper so far.

The longest continuing uniaxial creep test (150°C/120 MPa) for OFP copper has exceeded 75 000 h (8.5 years). For damage modelling it is of interest that interrupted testing of the longest uniaxial specimen has also shown distributed microcracking (Fig. 5). The observed effect of small scale natural weld (FSW) defects suggests increasing notch weakening with increasing time to rupture (decreasing stress). The test results continue to support creep modelling and have been used for life assessment with the latest expected temperature history of the canister at different assumed stress levels, using a combined Wilshire and LCSP creep model for OFP copper (Fig. 6). The resulting predicted allowable (constant) stress level for a lifetime of 100 000 years would be about 140 MPa for the copper overpack.

In the combined corrosion and creep testing with welded CT specimens immersed in aerated simulated Olkiluoto groundwater at 90°C, heavy general corrosion up to a depth of about 2 mm was observed already after 4400 h of testing (Fig. 7). In comparison to this, no significant indications of localised corrosion have been observed. Further work is suggested to clarify the temperature dependence of reported stress corrosion under reducing groundwater conditions.

The results appear to carry important implications relevant for the final disposal conditions. The earlier assumption of low stress level in repository conditions may not hold as very high stresses are also foreseen by the SKB design report for the copper overpack even after long times. It is suggested that the vessel stress analysis is repeated using the VTT creep model for an independent verification of the stresses especially in the weld region, and the effect of multiaxiality is clarified in terms of claimed notch strengthening versus observed life shortening.

Figure 4. Grain boundary cavity density in the OFP copper CT-specimen with friction stir weld at 175°C/35 MPa reference stress; up to 15000 h the cavity size was below the limit of detection.

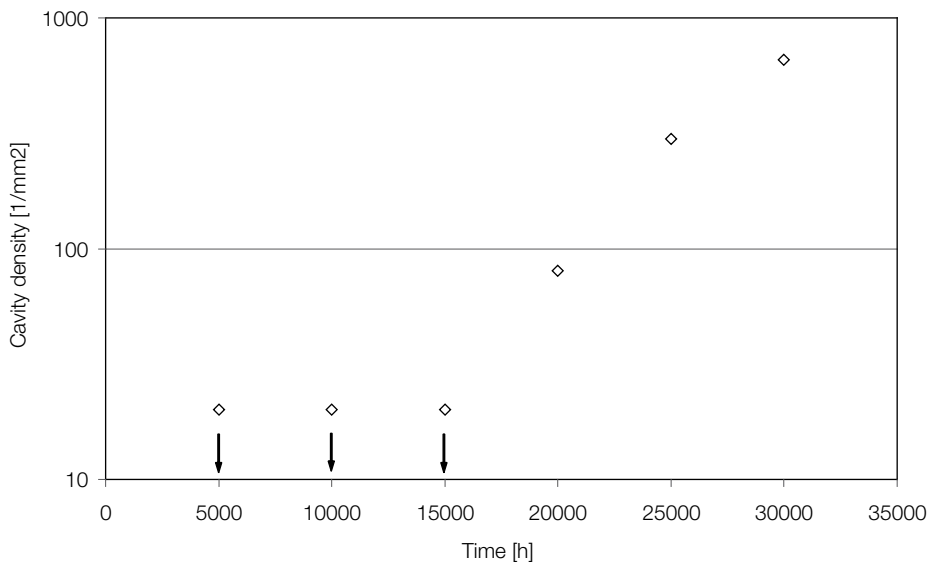


Figure 5. Surface cracks in the uniaxial specimen after 75134 h at 150°C.

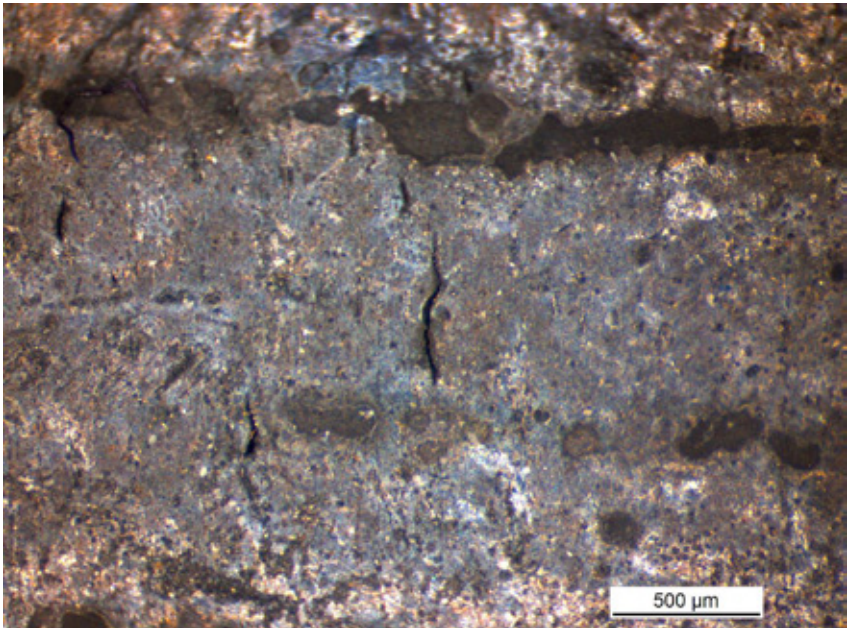


Figure 6. The temperature history of the copper canister and the corresponding predicted life fraction at three stress levels in final disposal conditions using the dry buffer assumption; dashed line is a conservative estimate for temperature history used in life prediction.

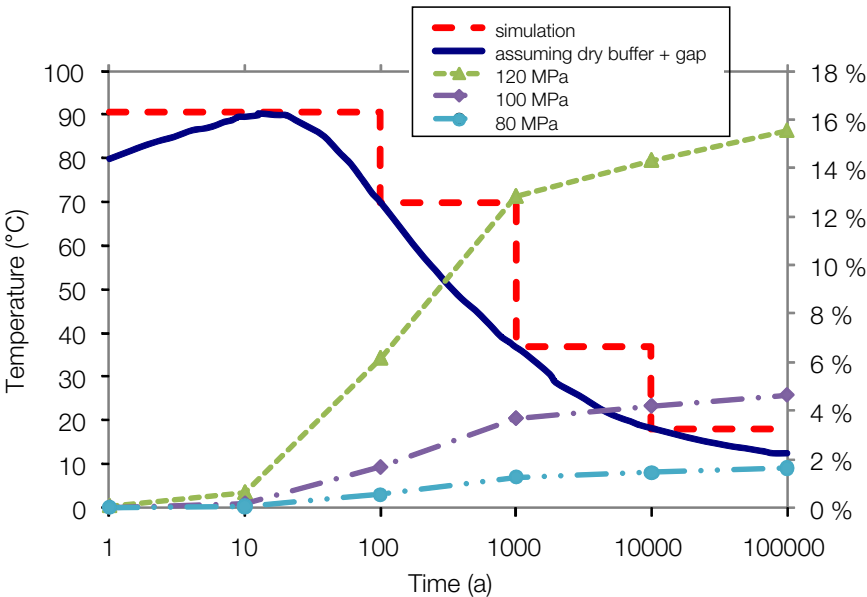


Figure 7. Extensive general corrosion in the CT specimen after 4400 h (90°C) in aerated simulated Olkiluoto groundwater.



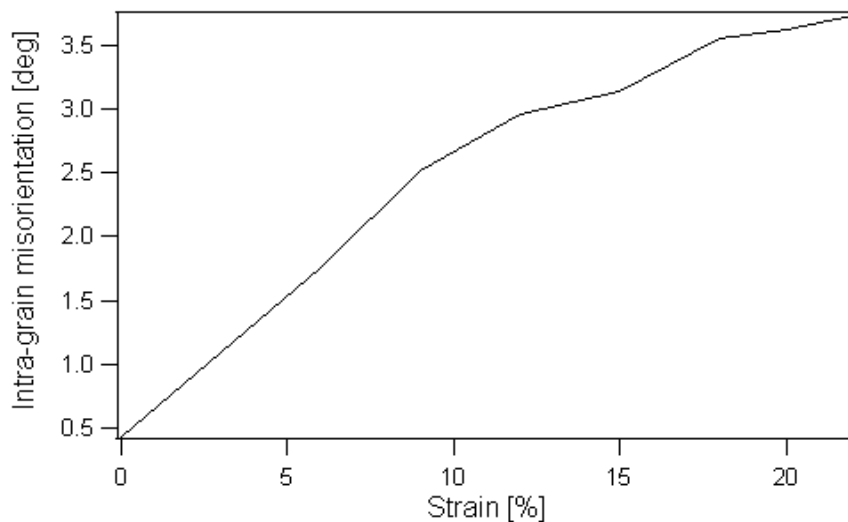
8.6 Deformation Mechanisms of Nuclear Waste Canisters

Kati Savolainen, Aalto University

MAIN RESULTS

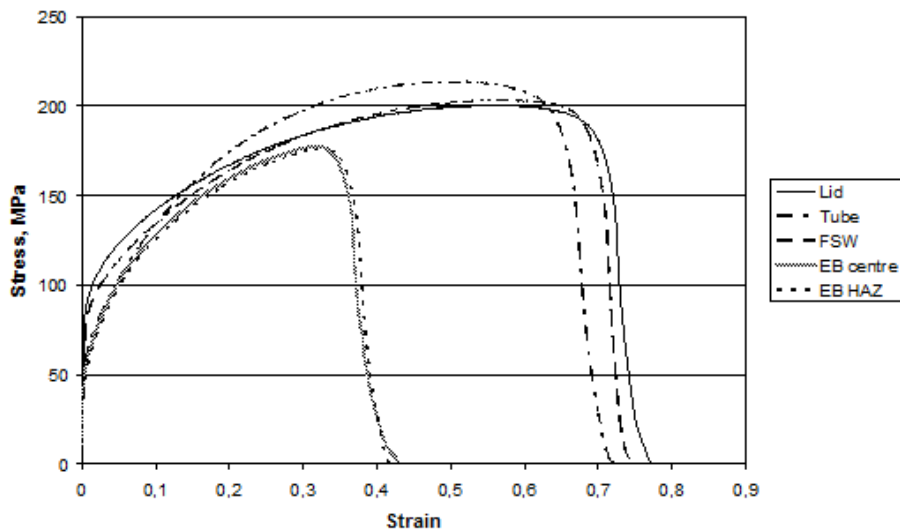
Calibration curve for determining the degree of local deformation using FE-SEM/EBSD equipment has been completed (Figure 8).

Figure 8. Calibration curve for determining the degree of local deformation in Cu-OFB.



Localization of plastic deformation in the different parts of the copper nuclear waste canister (forged lid material, extruded tube material, electron beam weld EB, and friction stir weld FSW) has been studied using large samples which contain the entire weld joint and optical strain measurement system. It was noticed that the strength and elongation to fracture of the FSW welds were similar to those of the base materials, while they were considerably lower in the EB weld. The EB weld samples fracture either at the centre of the weld due to the large grain size or at the HAZ between the weld and the lid due to the large grain size gradient. The FSW weld samples fractured at the line of entrapped oxide particles. Stress-strain curves of the samples are seen in Figure 9.

Figure 9. Stress-strain curves of the different parts of the spent nuclear fuel canister.



In-situ testing with FE-SEM/EBSD/EDS equipment has been done to study the localization of deformation on the microstructural level. It was noticed that due to technical reasons the majority of the tests should be done using light optical microscopy and optical strain measurement system and use the in-situ electron microscopy for specific measurements needed for publications. The results are used in most of the articles and reports of the project.

The effect of hydrogen on the mechanical properties of the copper canister was studied by tensile tests with hydrogen charged specimens as well as internal friction and hydrogen thermal desorption spectroscopy (TDS) methods. Figure 10 shows the typical TDS spectra of hydrogen charged Cu-OFP specimens tested under different conditions. Figure 11 shows the stress-strain curves of as-supplied and hydrogen charged Cu-OFP specimens. The results show that testing of hydrogen charged specimens in air and under applied electrochemical cathodic potential leads to similar elongation to fracture – ductility of copper is not lowered due to the effect of hydrogen. However, the stress-strain curves show that the deformation of copper occurs at a lower stress level (reduced flow stress) due to the hydrogen effect.

Figure 10. TDS spectra of hydrogen charged Cu-OFP copper specimens tested under different conditions. The total amount of hydrogen has been measured with the LECO method (weight ppm).

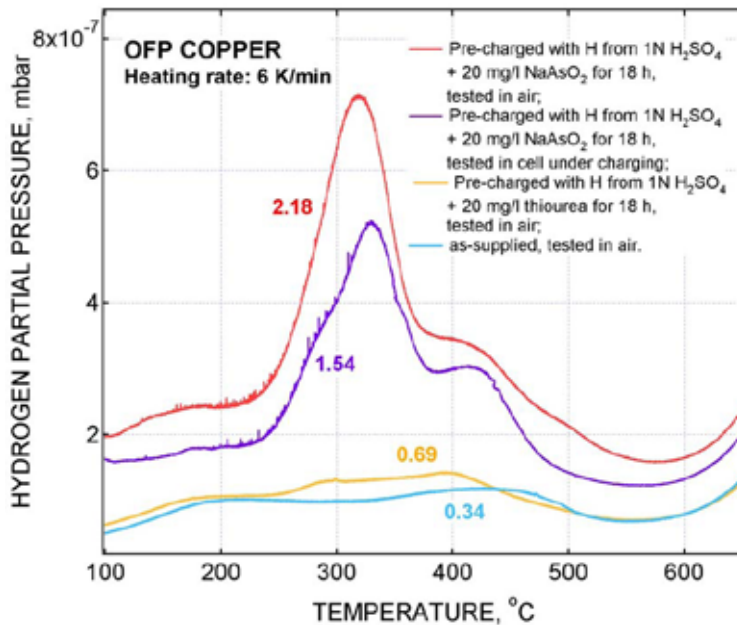
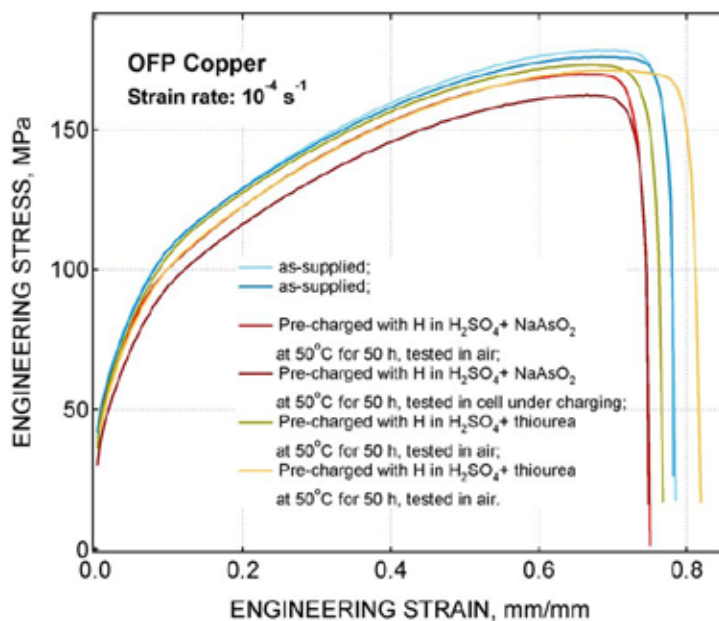


Figure 11. Stress-strain curves of the as-supplied and hydrogen charged Cu-OFP specimens.



THE SIGNIFICANCE OF THE RESULTS ON NUCLEAR WASTE MANAGEMENT

The results of this research project are very important in understanding the deformation mechanisms and expected lifetime of the copper spent nuclear fuel canister. The welds of the canister are the location of significant discontinuities (microstructural elastic and plastic discontinuity, high residual stresses and strains, strong stress concentrations, and the effect of possible welding defects). The microstructures of welds have not been characterized earlier using modern orientation microscopy methods (FEG-SEM/EBSD) nor has nanoindentation been used to locate the local elastic and plastic discontinuities of the microstructure. Using these new methods is imperative in understanding the different possible deformation mechanisms as well as their effect on the scientific estimation of the lifetime of the spent nuclear fuel canister. Understanding the microstructural discontinuities of the copper canister is particularly important in evaluating the creep and stress corrosion cracking resistance of the copper canister. It has been suggested that the hydrogen formed during general corrosion of copper is absorbed into the copper, embrittling the canister and possibly leading to premature fracture. It is very important to know the absorption, solubility, and diffusivity of hydrogen in copper in order to scientifically exclude the hydrogen effects in regards to embrittlement and creep mechanisms.

METHODS USED IN THE STUDY

- Electron beam (EB) and friction stir welding (FSW)
- Scanning electron microscopy and energy dispersive spectroscopy (SEM/EDS)
- Electron backscatter diffraction (EBSD)
- Light optical microscopy
- Mechanical testing (tensile testing, hardness measurements, nanoindentation)
- Optical strain measurement
- Mechanical spectroscopy (internal friction)
- Electrochemical hydrogen charging
- Thermal desorption spectroscopy (TDS)

8.7 Measurement of the corrosion rate of copper in gas phase at first stage of the final disposal

Jari Aromaa, Aalto University

In the project the corrosion rate of copper in the gas phase and the effect of solution variables on the corrosion were studied related to first stage of the final disposal. It has been supposed that the corrosion, which happens at the first stage, will be corrosion under immersion conditions. During this time the general corrosion of copper can cause a thinning of 1–2 mm. Based on literature the corrosion rates are in the order of 0.1 $\mu\text{m}/\text{year}$ in atmosphere, 2–3 $\mu\text{m}/\text{year}$ in bentonite and 20 m/year under immersion. After the swelling of the bentonite buffer general corrosion should not happen any more.

Before the swelling of the bentonite buffer into the copper capsule there is a thin gas gap between them. The starting point for this study was that in the beginning the corrosion could be similar to atmospheric corrosion. The evaporation and the condensation produce a solution film to the surface of the sample, which starts the corrosion. In the atmospheric corrosion the rates of reaction may be higher than in solution because the mass transport phenomena in the condensed solution film are faster than during immersion in to the solution. The evaporation and condensation of the solution in the gas phase also can lead to the enrichment of impurities to the surface of the top of the capsule. The enrichment of impurities can have an effect for the progress of general corrosion, pitting corrosion and stress corrosion. It is generally assumed that an oxygen-free reducing environment should not cause general corrosion of copper. The environment that has pH high enough passivates the surface of copper. The sulphide ions passivate copper in reducing conditions. The chloride ions, however, may cause the increasing of corrosion. The local damages, which have formed to the surface can serve as the starting points of pitting corrosion or stress corrosion. The corrosion studies were conducted by using in the gas phase of a closed reactor using a quartz crystal microbalance, which is able to measure weight change of nanogram level.

MAIN RESULTS

In the study it was clarified how the quartz crystal microbalance can be used for corrosion measurements in the gas phase. In the measurements resolution weight changes and their analysis corresponds to corrosion rate 0.1 $\mu\text{m}/\text{year}$. The measurement method and analysis method of the microbalance results were sufficient for the measurement of small corrosion rates. The reactor that was built to control the corrosive environment operated well.

In the tests the effect of the oxidising atmosphere and composition of the solution on the corrosion rate of copper surface were studied. During atmospheric testing

a reaction product layer, whose thickness is of the order 50–700 nm, forms to the surface. First there is fast increase in weight, which is in accordance with a logarithmic rate law, and it is followed by a linear change of weight as a function of the time. The weight increase of the first stage happens during some tens of minutes. After this the reaction product layer remains stable in some cases, it begins to dissolve in some cases and oxidises further in some cases.

The clean copper surface that has been just prepared oxidises fast in a dry gas phase. The weight increase is 2–4 µg. After this the sample will reach the steady state. In a moist gas phase that has been produced with distilled the forming of the oxidized layer is seen as a clearly stronger weight increase, in the order of 20–25 µg, compared to dry air. Synthetic Allard ground water was used as a base solution to produce a moist gas phase. In the tests the chloride content of Allard water, acidity and the content of the ammonium ions and sulphide ions were varied.

With the moisture and condensation of distilled water the corrosion rate of copper in air was about 10–20 m/year at temperature 40–60 °C but at 80 °C it was only 4–8 m/year. In oxygen-free conditions prepared by nitrogen copper did not always dissolve but formed reaction products, which was seen as increase of weight. In oxygen-free conditions the weight change varied from 1 m/year thinning caused by corrosion to 3–4 µm/year growth of oxide layer. Oxygen purging of the water produced highly oxidising conditions and the growth of the oxide layer continued at 30 µm/year.

The effect of the salts in the Allard water was seen as higher corrosion rates than in distilled water when the atmosphere is air. In the oxygen-containing atmosphere, the corrosion rate was 10–70 µm/year. In the oxygen-free atmosphere produced by nitrogen the composition of water did not have significance and the change in the weight varied from 1 µm/year thinning caused by corrosion to 3–4 µm/year growth of oxide layer. When the chloride content of Allard water was increased, in the gas phase weight decrease slowed down and changed to weight increase when the chloride content was more than 1000 mg/l. In the immersion tests the effect of the chloride was opposite and the corrosion rate of copper increased when the chloride content increased. At temperature 80 °C, in Allard water with chloride content 2–20 g/l the corrosion rate was in the range 1–100 of µm/year. The addition of the sulphide ions or ammonium ions or the decrease of pH did not have significant effect on weight change.

The oxidation of the surface and forming of reaction product layer has an effect on the progress of the corrosion. With clean copper surface the weight increase obeys first a logarithmic rate law and is followed by a linear change of weight. This linear part has been used in the comparison of corrosion rates. In the oxide layers formed before corrosion test copper oxide CuO was analysed. When a thin oxide layer covered the copper surface, the weight change with time was similar to that of clean copper. A thicker oxide layer caused that the weight change with time did not always follow linear rate law. When the thickness of the oxide layer was 30 nm before the corrosion test, the weight change can follow a parabolic rate law. The linear part was

not seen in these tests. When the surface has a thin oxide film the same methods as for clean copper can be used for the evaluation of corrosion rate. To a surface, which has oxidised more strongly, the rate laws used in atmospheric corrosion and high-temperature corrosion must be adapted. In the gas phase the oxidised oxide layer did not affect corrosion rate compared with clean copper surface. In the immersion the presence of a thin oxide layer, which covers the surface probably only partly, resulted in approximately same corrosion rate as with clean copper surface. Thicker pre-formed oxide layers did not corrode in the immersion tests.

SIGNIFICANCE OF THE RESULTS

In the final disposal the conditions are first oxidising and change to reducing when oxygen is consumed out of the closed space. In a moist atmosphere, when distilled water is used, the weight of the sample did not change in oxygen-free conditions, in oxidising conditions the weight decreased and in the highly oxidising conditions the weight increased. When Allard water was used as corrosive environment, the same trends were seen. The results support the assumption that in oxygen-free conditions copper does not corrode significantly.

The composition of water will affect a corrosion rate when there is oxygen in the environment. In the oxygen-free environment that was produced by nitrogen bubbling the weight change with distilled water was -0,04 to 0,1 ng/cm²/s and with Allard water -0,1 to 0,03 ng/cm²/s, so the weight changes do not have a difference. In oxidising conditions, when the solution was purged with air, the weight change with distilled water was -0,1 to -0,6 ng/cm²/s and with Allard water -0,05 to -2 ng/cm²/s. In oxidizing moist atmosphere the calculated thinning with Allard water was three-fold compared to distilled water. The increase of the chloride content slowed down corrosion in the gas phase but increased in the immersion test. Changing the condition to slightly acidic or addition of sulphide ions or ammonium ions did not affect corrosion in the gas phase. When solution condenses to the surface of copper in an atmosphere, which contains oxygen, the corrosion rates calculated from the weight change results can be tens of micrometres per year. In oxygen-free conditions the corrosion rates are some micrometres per year maximum. It is suggested to study further the effect of the corrosive environment and especially of chlorides on the corrosion rate.

The outer surface of the capsule has time to oxidise before placing it in the repository. This oxide layer on the surface can have an effect on the corrosion rates. Especially a thin oxide layer, which does not cover surface totally, can increase corrosion rate in oxidizing conditions. It is suggested to study further the effect of oxide film and other reaction product layers on the corrosion rate.

When evaluating determined corrosion rates, one must remember that they are results of short tests, which lasted for maximum 1–3 days. The obtained corrosion rates are very likely higher than seen in long-term corrosion, which will last for years. The calculated corrosion rates have been ten times higher compared to the

corrosion rates that have been measured in bentonite and the same order as the corrosion rates that have been measured in immersion. The possibility that the rate of general corrosion in the oxidising conditions can be higher than estimated cannot be excluded.

RESEARCH METHODS

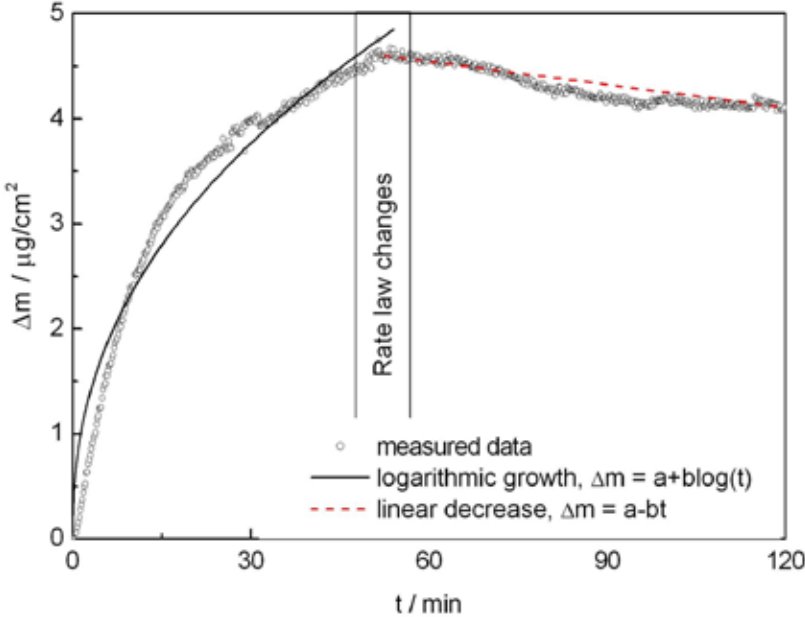
In the study a Stanford Research Systems SRS200 quartz crystal microbalance was used. The operation of the quartz crystal microbalance is based on the change of the crystal resonance frequency when the weight of the crystal sample changes. The equipment that has been used in the study resolution of order 10 ng/cm^2 is generally presented. In the measurements that have been done to clean copper a rapid weight increase following logarithmic rate law is followed by a slower linear change in the weight as a function of the time. This linear part was used in the comparison of corrosion rates. From the measurement results the slope of linear weight change with time was calculated and this was further utilised in the calculation of the corrosion rate. It was possible to determine linear weight changes at the level $10^{-2} \text{ ng/cm}^2/\text{s}$, which corresponds to a general corrosion rate $0.1 \text{ }\mu\text{m/year}$.

The tests were done by putting a copper plated quartz crystal sample in atmosphere, which was formed as the test solution in reactor vaporised. The samples were made by electrodepositing $5 \text{ }\mu\text{m}$ thick layer of copper from copper sulphate and sulphuric acid solution to the surface of the quartz crystal. The pure copper does not correspond to the phosphorus-alloyed copper of capsules but it was assumed not to be significant for the general corrosion that was examined.

The reactor made for the tests was based on closed gas-tight jacketed glass vessel. The reactor was heated with the help of the water bath. At the bottom of the reactor was a constant volume of vaporising solution and the measuring sensor of the quartz crystal microbalance was in the top of the reactor in the gas phase. The control of the atmosphere was done by bubbling air, oxygen or nitrogen to the solution at the bottom and by leading the gas through water lock out of the reactor. The reactor worked well in the test conditions.

The tests were based on weight changes measured with a quartz crystal microbalance. The measurement results were interpreted as shown in the example in Figure 12. The slope of the linear part of Figure 12 represents the weight change as a function of the time that was used for the calculation of corrosion rates.

Figure 12. The principle of the analysis of corrosion tests.



8.8 Sulphide induced stress corrosion cracking of copper

Timo Saario, VTT

INTRODUCTION

Copper canister is a central technical barrier for radioactive release from high level nuclear waste. Stress corrosion cracking (SCC) is a failure mechanism which has the potential capacity of damaging all the canisters in a relatively short time.

In 2007 a Japanese research group showed that sulphide (S^{2-}) can cause SCC in pure copper under anoxic high chloride water conditions. Sulphides may come to contact the copper canister surface through three different scenarios: 1) transport via groundwater flow, 2) production at the bentonite/rock interface via sulphate reducing bacteria (SRB) and further transport and 3) through SRB activity within bentonite (e.g. pyrite reduction).

The goals of the project were to evaluate the sulphide induced SCC risk of copper canisters under repository conditions. Technical targets were:

- 1 Evaluate the maximum sulphide concentration which can form at the bentonite/rock interface because of SRB activity.
- 2 Develop a diffusion model and make a quantitative estimate of the sulphide concentration reaching the surface of the copper canister in three different scenarios.
- 3 Develop an experimental arrangement for SCC tests in sulphide containing groundwater.
- 4 Determine experimentally the minimum concentration of sulphide in groundwater which can cause SCC in pure copper (CuOFP).

MAIN RESULTS

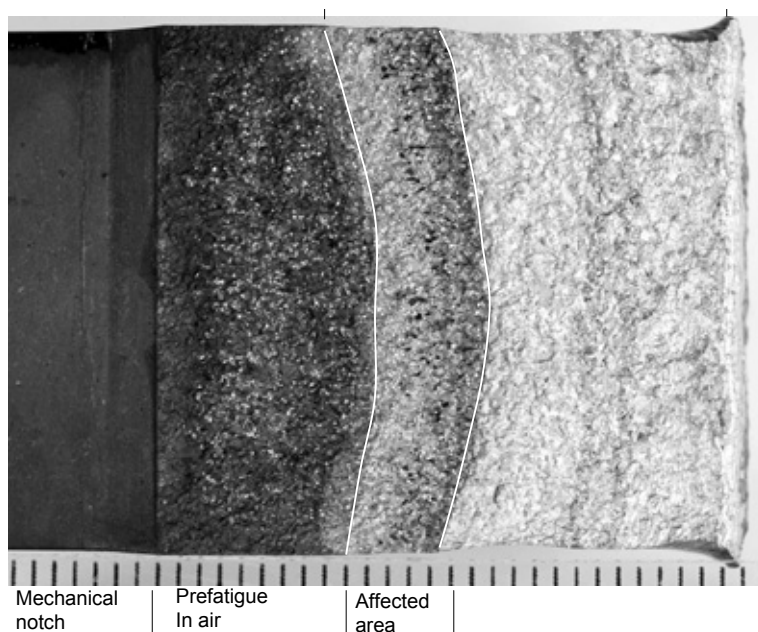
1) The literature study performed as part of this project showed that the maximum sulphide concentration that may form at the bentonite/rock interface due to bacterial activity may be 400–450 mg/l at near neutral pH. At higher pH -values the maximum concentration may be much higher. In the groundwater sulphide concentrations are typically relatively low, 1–3 mg/l. The sulphide concentration that forms because of SRB activity within bentonite itself is very low.

2) In this project the transport of hydrogen sulphide in compacted bentonite in conditions of variable dry density was modelled by molecular diffusion and advection. In scenarios where the density of bentonite locally decreases resulting in advection (e.g. piping, erosion-corrosion) the access of sulphide to the copper surface will be much easier. Maximum sulphide flux values onto the copper canister surface were observed in advection cases where the concentration at source (rock/bentonite - interface) was set to 450 mg/l. In these cases, the maximum flux was almost 200 000

$\text{mgm}^{-2}\cdot\text{a}^{-1}$. The model can be used to evaluate the flux of sulphide onto the canister surface, as generated in different scenarios.

3) A new experimental arrangement was developed in this project to simulate the conditions copper canisters are facing in the disposal vault. The arrangement was used to study at room temperature the effect of exposure of CuOFP to Olkiluoto-type saline groundwater with sulphide contamination. Precracked CT-specimens of CuOFP have been exposed to groundwater with 200 mg/l, 100 mg/l and 10 mg/l sulphide. The occurrence of SCC in CuOFP when exposed to sulphide containing groundwater at room temperature could not be unequivocally shown. However, scanning electron microscope (SEM/EDS) studies of the fracture surface showed that during the exposure sulphur/sulphide and to a lesser extent also other species present in the groundwater had entered the material ahead of the precrack tip, with a penetration rate of more than 1 mm per week (Fig. 13). Optical metallography of sliced and polished cuts of exposed samples showed that sulphide containing inclusions could be found in all three dimensions of the material and not only in the crack plane, although the area fraction of the inclusions was normally the highest close to the crack plane.

Figure 13. Fracture surface of a specimen exposed at room temperature to groundwater with 200 mg/l of sulphide for five weeks. The affected area (ahead of the crack tip, left from the prefatigue area in the picture) with black spots (inclusions) was not in touch with water during the exposure, and has only been revealed through further fatigue (performed in air) of the specimen after the exposure.



The finding that species from groundwater can enter into solid CuOFP material and penetrate (via diffusion) to such distances within a few weeks is a new one. The possible effect of such penetration on the mechanical properties of the CuOFP material will be clarified in a separate project within the KYT 2014 -programme.

8.9 Corrosion of copper by water under oxygen free conditions

Antero Pehkonen, Aalto University

ESSENTIAL RESULTS OF THE PROJECT

Some recent publications have generated some media attention and concern, given the important role of the copper canister in the Scandinavian disposal concept for spent nuclear fuel. In the concept, the 50 mm thick copper canister is placed to a depth of about 420 m in bedrock and isolated by engineered barriers. Based on the measured conditions deep in the bedrock and well known corrosion behavior of copper, the estimated time of endurance for the canister is around 100,000 years. The thickness is planned to be enough for preventing the radioactive waste from interacting with groundwater too early. The recent publications about copper corrosion in oxygen-free water state that this time could be dramatically shorter. A short review on the disposal plans and repository conditions is included in this paper.

These recently published research studies /1-3/ argue that there may be stronger evidence about corrosion of copper in pure water than previously believed. The study has generated media attention and concern, mostly because of the ongoing nuclear waste disposal plans. The first evidence about this possible phenomenon was brought up more than two decades ago /4/. According to known copper thermodynamics, corrosion of copper should not happen in such conditions.

Hultquist and Szakálos /1-4/ suggest, that copper dissolves, in contrast to the knowledge so far, also in water without any dissolved oxygen according to reaction $\text{Cu} + y\text{H}_2\text{O} \rightarrow \text{H}_x\text{CuO}_y + (2y-x)\text{H}_{\text{ads}}$. The existence of new corrosion product compounds is based on the H/O ratio in SIMS analyses.

The research project aims at providing an independent and reliable verification of the corrosion mechanism proposed by Szakálos et al. as well as estimating the possible consequences of this mechanism under realistic repository conditions. The research work will be done the Laboratory of Corrosion and Materials Chemistry at Aalto University, VTT Technical Research Centre of Finland and Studsvik Nuclear AB in Sweden. The content of this thesis is focused on theoretical evaluation related to the proposed corrosion mechanism between copper and pure water, and on the design and construction of the experimental apparatus.

The results of this project are more closely given in a master of thesis of Ikonen /5/. From the viewpoint of thermodynamics, either the results from the latest experiments are faulty or the study on the corrosion of copper in pure water must be taken further and the thermodynamics must be completed with the new knowledge. It is clear that the known corrosion reactions in pure water are not possible. The only possible conditions for copper corrosion by pure water in the traditional thermodynamic data are highly unnatural and these conditions are not present in

distilled, oxygen-free water. Highly unnatural stands for a combination of relatively high temperature (e.g. 150 °C and acidic conditions, extremely low concentration of dissolved copper ions (e.g. 10^{-8} mg/l) or low partial pressure of hydrogen. This also means that traditional corrosion thermodynamics do not support corrosion of copper in repository environment.

There are some other possibilities for hydrogen evolution in the laboratory experiments when e.g. stainless steel vessels are used.

- 1 The iron and chromium in stainless steel react with water and form hydrogen
- 2 In the natural partial dissociation of water some H^+ ions exist in water and these ions form gaseous hydrogen and the copper surface acts as a catalyzer.
- 3 Some hydrogen remains in the bulk copper in the copper production process and this hydrogen is little by little released.

Two identical test equipment has been constructed in this project. The design has done as closely as possible based on the information got from the equipment designed by Hultquist. A major problem is the inadequate information of the equipment used by Hultquist.

THE STATUS OF THE RESULTS ON THE RESEARCH OF NUCLEAR WASTE MANAGEMENT

Based on the thermodynamic calculation supported by the publications in the literature the compound ($HxCuOy$) suggested by Hultquist to form in the corrosion reaction of copper in water without any dissolved oxygen, is possible only in conditions, which are not possible in the final disposal of nuclear waste.

Even if the suggested corrosion reaction could be possible in the final disposal, the reaction will without any doubt stop based on the thermodynamic facts. This is due to the passivating oxide layer formed on the surface of the copper canister.

EXPERIMENTAL

Two identical test equipment has been fabricated and the other has been sent to Studsvik Nuclear AB at the beginning of March 2011. Figure 14 illustrates the principle of the equipment and in figure 15 the equipment used by Hultquist is shown and in figure 16 the equipment of this project respectively. All parts in contact with water or gases inside the equipment are made of stainless steel AISI 316. A glass vessel is placed in the test vessel, where water and copper foils will be inserted. The possible hydrogen formed in the corrosion reaction will drift through a Pd foil to the upper part of the equipment, where a vacuum (about 10^{-6} bar) has been pumped. The increase in the pressure due to the hydrogen is measured using a pressure gauge.

Experiments will be done at temperatures of 20 and 60 °C. The higher temperature is done by a heating element outside the test vessel. In order to prevent the condensation of water on the lower surface of the Pd foil, also the Pd flange will be heated. Experiments are done both in distilled water and in simulated ground water.

Figure 14. A schematic figure of the test system.

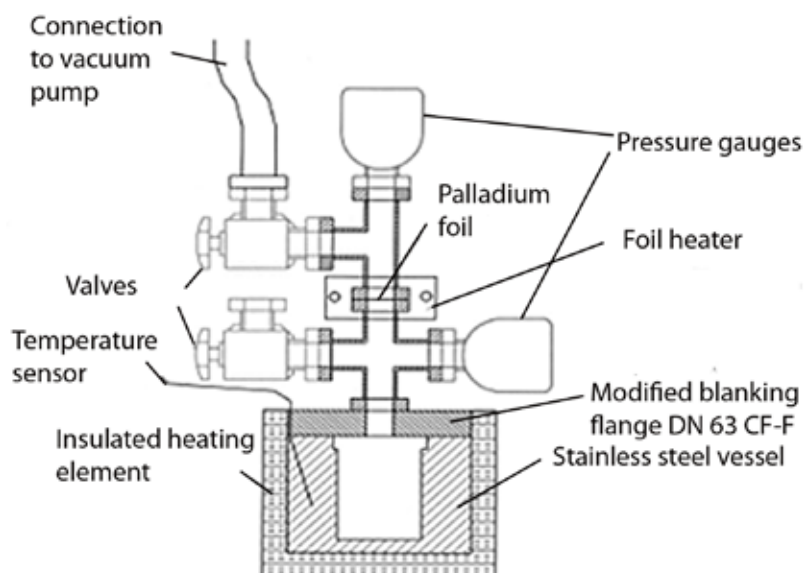


Figure 15. The test equipment of Hultquist.

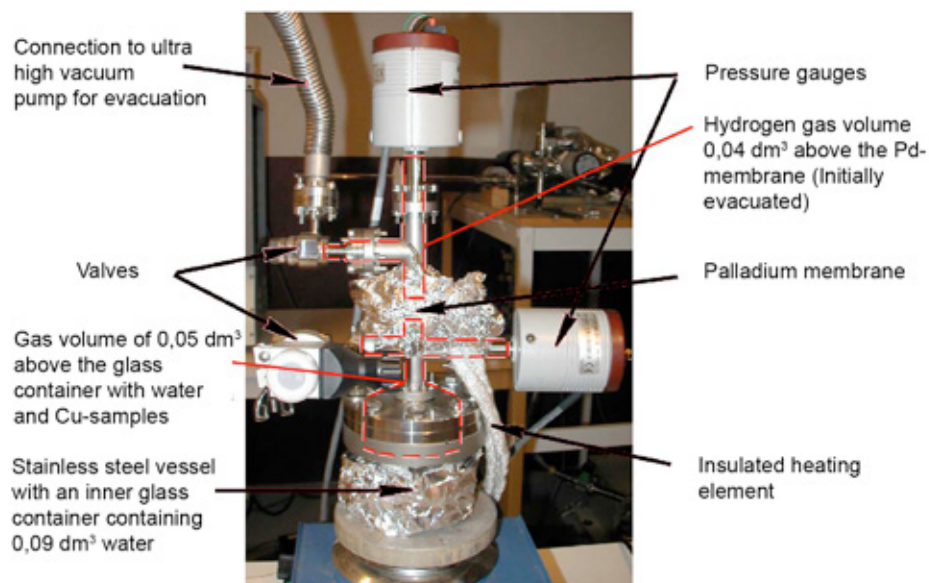
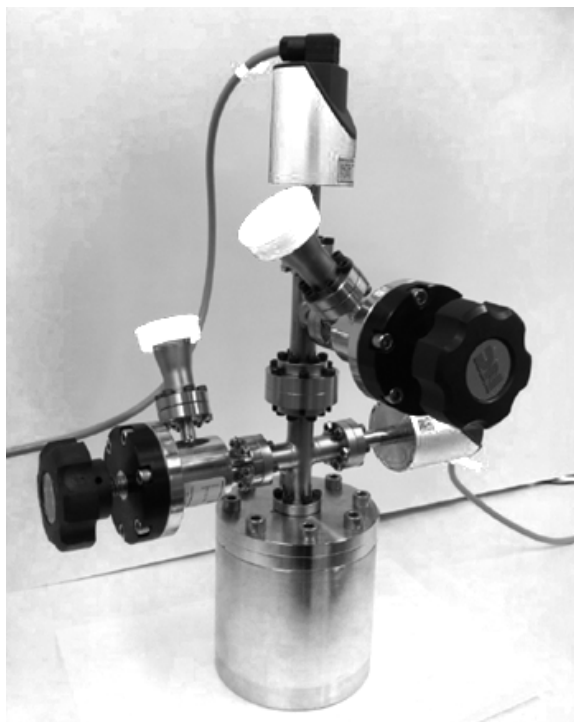


Figure 16. Test equipment of this project.



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8.10 Bentomap: Survey of bentonite and tunnel backfill knowledge

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Leena Korkiala-Tanttu, Aalto University

The knowledge of bentonite and other backfill materials for nuclear waste disposal purposes in Finland has been fragmented. The conducted research has mainly been based on the collaborative projects between Posiva Oy and SKB. Finnish experts have also participated in some international projects. Most of the research projects have, however, been single projects without the aim of creating comprehensive knowledge. The Bentomap survey was produced to meet the needs for comprehensive knowledge. The survey included the compilation of a state-of-the-art report (Rautioaho et al. 2009) and a database of bentonite literature. The state-of-the-art report and its database can be used by authorities and experts. Besides that, it can be used for the training of authorities and experts. Another import task of the survey was to show the deficiency and reliability of the bentonite knowledge. The type of comprehensive knowledge presented in the Bentomap survey is needed in the preparation of the application of the construction license for the disposal facility by the end of the year 2012 and in its evaluation. Altogether the results of the survey can be used for the orientation of bentonite research in the future.

The Bentomap survey was financed by the KYT programme, and it started in spring 2007 and was finished in April 2009. In the first year, a large portion of the material (research reports, articles, and publications) was collected and classified. During 2008 and 2009 additions were made to the database, and the database was evaluated and the state-of-the-art report compiled. VTT managed and coordinated the survey. Aside from VTT, the database was evaluated by Paula Keto from Saanio & Riekkola Oy and Antti Lempinen from Marintel. The coordinator of the survey at VTT was Leena Korkiala-Tanttu, and the other researchers involved at VTT were Elisa Rautioaho, Erko Pyy and Rainer Laaksonen.

The scope of the survey was to compile the THM knowledge of bentonite materials, but some research on other backfilling materials and coupled processes was also included. The database has been created so that its core is the buffer and backfill research done by SKB and Posiva Oy. A large key words search was also done to VTT's electronic library databases, like Compendex, Inspec, NTIS, Elsevier's Scopus and Science Direct. Besides these, also Scirus and Google Scholar databases were used. Other material included EU projects, such as the Febex project and the Marie Curie programme, journal articles and conference papers. In the beginning, the database search was limited to the latest ten years (from 1997-2008). After the evaluation of the first year it was suggested that the survey should also cover some older research, which was included accordingly. The database has been updated continuously during 2007-2009, and now includes reports all the way from 1982

to 2009. The database was created with the Microsoft Excel and Access programs. The state-of-the-art report was compiled based on the evaluation of the publications contained in the database. The report concentrated on the experimental thermo-hydro-mechanical properties of the buffer and backfill materials.

To summarize, the objectives of the two-year Bentomap survey were:

- to map the conducted research for buffer and backfilling materials,
- to evaluate the applicability of the test results in Finnish repository solutions,
- to present a database of the bentonite research including an evaluation of the research results,
- to present a separate state-of-the-art report of the conducted research results including evaluation, scope, limitations and possible deficiencies of the knowledge, and
- to give a wider perspective of the level of the national and international know-how on bentonite and backfilling research.

The Bentomap database includes over 200 different publications on bentonite and backfill materials, and their thermal (T), hydraulic (H), mechanical (M), chemical (C), gaseous (G) and, on the rare occasion, biological (B) property studies. The survey has ranged from small scale laboratory tests to the full-scale tests in real deep depository conditions together with the modelling of these experiments and other feasible scenarios. The information collected in the database for each article of study includes characterization of the material and description of methods and their applicability (or limitations). The modelling analysis describes the input data and boundary conditions together with the reliability of the results. More specifically, the table compiled for the various studies is comprised of the following categories: topic of research, writers, year of publication, publisher and publication information, key words, researched materials, laboratory or modelling study categorization, scale of the study, methods and models, studied properties, long-term properties, limitations, representativeness, results and conclusions, further research needs, characterization, reliability, evaluation of the results, applicability and comments. However, not all categories have been filled out for each study due to unavailability of information and evaluation, and resource limitations.

The state-of-the-art report concentrates on the conducted experimental thermo-hydro-mechanical research and knowledge for buffer and backfilling materials, particularly bentonite, and largely excludes the modelling work collected in the Bentomap database. The various studies and their results are listed in the report in condensed form covering the more important THM properties of a variety of bentonite materials and clays, including Friedland clay and FEBEX, Asha and, in particular, MX-80 bentonite. The thermal properties examined are thermal conductivity, specific heat, and the thermal expansion coefficient. The compiled hydraulic property results, on the other hand, pertain to hydraulic conductivity and the water retention curve, and the investigated mechanical properties include strength, deformation, rheological and swelling properties. The presentation of

results from the experimental research is followed by conclusions drawn from both the individual studies and the Bentomap database as a whole. The report presents evaluation, scope, limitations and possible deficiencies of the knowledge and provides wider perspective on the level of national and international know-how on bentonite and backfilling research.

Some general conclusions can be drawn based on the issues covered by the Bentomap database and knowledge presented in the state-of-the-art report. Buffer and backfill materials such as MX-80 and FEBEX and Friedland clay have been particularly extensively studied in the recent years. Mechanical and hydraulic parameters have generally been more widely and thoroughly investigated by experimental methods in the articles and reports than thermal parameters. However, effects of temperature and water content on several bentonite properties are fairly well represented. It may be noteworthy for material selection in nuclear waste repositories that the same suitable basic mineralogical characteristics and thereby similar sealing properties can be exhibited by bentonite materials with quite different geological origin. Effects of salinity on some of the important properties of the buffer and backfill materials are well presented mainly for concentrations less than 3.5% and for a single type of salt (mixture of sodium and calcium chlorides).

Some issues not extensively covered or requiring further study in the current knowledge as presented by the Bentomap database are also summarized in the state-of-the-art report. These issues include the long-term properties of bentonites, the effect of time on sealing and safety functions, evolution of saturation in the buffer and backfill, piping and erosion and the effects of higher salt concentrations, as well as the effect of test scale and up-scaling of small scale experiments to full scale.

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8.11 Modelling in near field (LÄHITHC)

Modelling of glacial erosion in bentonite buffer (GLASE)

Markus Olin, VTT

INTRODUCTION

In the summary two projects are briefly reviewed. The project LÄHITHC modelling in near field (Lähialueen termo-hydro-kemiallinen mallinnus, LÄHITHC) was carried out during the years 2007–2008 and Modelling of glacial erosion in bentonite buffer (Bentoniittipuskurin jääkausieroosion mallinnus, GLASE) during 2008.

In the evolution of near field in spent nuclear fuel repository, couplings between different phenomena play an essential role. Especially important these couplings are during the relatively short period after closing of the final repository, when temperature, groundwater flow and chemical gradients and mechanical phenomena in materials are maximal. Main research topic has been in understanding thermo-hydro-mechanical (THM) phenomena due to uncertainties in the duration of saturation phase. On the other hand, coupling temperature, ground water flow and chemical reactions together to thermo-hydro-chemical (THC) modelling has been quite limited in Finland. Consequently, operators in Finnish nuclear waste management have recognized reactive transport modelling as an important development area.

In addition to uncertainties in saturation of bentonite buffer, the preservation of its functionality must be shown in expected geochemical environments. During the thermal phase, possible mineralogical alteration of bentonite is enhanced due to elevated temperature. From the point of view of long term functionality of bentonite, it is essential to recognise mechanisms, which can reduce it.

A problem is connected to the enrichment of soluble silica in bentonite; this may by precipitation cause cementation of bentonite. Then bentonite may become brittle and lose its plastic and swelling properties. Weathering products of cement materials can cause problems in the long term stability of the bentonite via dissolution of the main component of the bentonite, montmorillonite, which mainly gives the beneficial properties to it.

Also the effects of the composition of groundwater in surrounding bedrock to mineralogical alteration of bentonite should be known better. An example of that is the behaviour of certain important elements for copper canister's corrosion tolerance, like sulphate's, which by microbial activity may change to form causing copper corrosion, precipitation in bentonite hot end.

The goal of LÄHITHC project was to create clearer understanding of chemical evolution (e.g. mineralogical alteration, pore water chemistry) and the functioning of bentonite buffer, after the closing of the repository in groundwater conditions prevailing during thermal phase. The target was to develop a computation model, which seamlessly couples chemical reactions to the heat production and saturation

of bentonite. Model parameters were checked and set by utilising published research material. By applying the experience created during the years 2006 and 2007, the goal was to create as realistic as possible description integrating thermal, hydraulic and chemical phenomena in near field.

The goal of GLASE project was to create better understanding about chemical evolution in bentonite buffer and the erosion following that, in post glacial groundwater conditions, especially in very dilute waters, which have a tendency to alter bentonite into colloidal form. The goal was to develop a computational model, which seamlessly combines physico-chemical changes during bentonite alteration. Model parameters were checked and set by utilising published research material.

METHODS

As modelling tool, we used THC phenomena integrating commercial PetraSim program, the graphic user interface of which guides TOUGH2- and TOUGHREACT calculation modules. Model calculation result will be the temporal and spatial distribution of temperature, saturation and chemical properties (e.g. mineralogy, soluble species) in bentonite buffer.

RESULTS

In GLASE project, an extensive literature survey about glacial erosion from the point of view of (T)HC modelling was written (Liukkonen 2008). Modelling work was started based on the report, and results were published in TOUGH meeting in San Francisco as an oral presentation in September 2009.

LÄHITHC project was started by learning to use the models at our disposal (EQ3/6, PetraSim ja COMSOL Multiphysics). Aku Itälä participated in PetraSim's (TOUGH and TOUGHREACT also) user course in the USA. In the project, modelling was prepared to be used as a part of the performance analysis of the final disposal of spent nuclear fuel. For this purpose, usability of COMSOL Multiphysics program was evaluated and a paper was written about these experience into COMSOL conference 4.-6.10.2008. Due these actions a COMSOL course was arranged at VTT about nuclear waste management 4.-5.2.2009.

In the project, Aku Itälä's Master of Science thesis about THC-modelling of bentonite was started. The main target in the work was in modelling, and therefore concrete experimental material was also needed. In the project, it was negotiated about the use of experimental material produced in LOT experiment (Long Term Test of Buffer Material) in Äspö hard rock laboratory in Sweden. In Finland, this work has been usually financed by Posiva Oy while the work is coordinated by SKB and Clay Technology. All partners had a positive attitude and the permission to use the material was obtained.

CONCLUSIONS AND UTILISATION OF THE RESULTS

By reactive transport modelling it is possible to estimate better than earlier the effects of varying thermo-hydro-(geo) chemical conditions in near-field to the functioning of bentonite, and finally how they affect the safety of the whole final disposal. Even very dilute and possible oxygen rich groundwater in post glacial conditions can be included in model calculations.

The work created new know-how and trained young researcher generation for the needs in the Finnish nuclear waste management. The readiness developed, useful both for regulators and waste producers, can be utilised in long term research projects, in which the goal is to create a THC model for the whole near field. Results of the project were readily utilisable as a part of the performance analysis of the final repository. The know-how developed in the projects enables also tighter collaboration between experimental work and modelling.

Reference

Liukkonen M. 2008. Literature study of the glacial erosion of the bentonite buffer - Theoretical background for modelling. 33 p., VTT-R-11267-08.

8.12 Coupled behaviour of bentonite buffer (PUSKURI)

Markus Olin, VTT

INTRODUCTION

The interest to study bentonite has been increasing during the last years in Finland. Several young scientists have started in the field, and most of them are now doing their PhD studies.

In KYT2010 programme there was a set of bentonite related projects, of which the largest one was VTT's, University of Jyväskylä's, Numerola Oy's, GTK's and Aalto University's collaborative project Coupled behaviour of bentonite buffer (Bentoniitipuskurin kytketty käyttäytyminen, PUSKURI). The coordinator of PUSKURI project was Kari Rasilainen (VTT) while the scientific coordinator was Markus Olin (VTT). The persons in charge in University of Jyväskylä, Numerola Oy, GTK and Aalto University were Markku Kataja, Antti Niemistö, Lasse Ahonen and Rolf Stenberg, respectively.

In addition to pure scientific and technical goals, PUSKURI project's goal was to compile a research plan for bentonite in the coming years. To compiling of that plan, in addition to PUSKURI scientist, also Emmi Myllykylä and Merja Tanhua-Tyrkkö (VTT, montmorillonite solubility), Pirkko Hölttä (University of Helsinki, colloids) and Merja Itävaara (VTT, microbes) were participating. The compiled plan will be published as a separate report in English.

METHODS

PUSKURI project involved both experimental work and modelling studies. In the experimental work at the University of Jyväskylä, the goal was to determine certain mechanic properties and especially to apply different tomographic methods.

In the modelling, both development of theory and numerical modelling by different tools was done. Numerical modelling was carried on by TOUGHREACT, Numerrin and COMSOL Multiphysics.

RESULTS

Results of PUSKURI project were so numerous that in this summary only some examples are given. A more detailed description about scientific results is available elsewhere (Olin et al. 2011).

In THC modelling, Aku Itälä made and published his Master of Science Thesis, and continued the work in some scientific seminars and articles (to be published later). Itälä was able to successfully model the LOT-experiment. Additionally, he also listed the problems and development proposals in THC-modelling of bentonite buffer.

Jarmo Lehtikoinen participated as an invited lecturer in the fall meeting of Atomic Energy Society of Japan Sapporo by the presentation "Research on Bentonite

Chemistry for Spent-Fuel Disposal in Finland” and also as an invited lecturer in the bentonite workshop in Hokkaido University 18.9.2010 by the presentation ”Diffusion model for ions from the viewpoint of the EDL theory”.

VTT and Numerola created in collaboration a model coupling saturation, diffusion and cation exchange; the model was implemented and tested in Numerrin, COMSOL and TOUGHREACT. An oral presentation was given of the results in the COMSOL meeting 17.-19.11.2010 in Paris, including an article in conference proceedings.

Petri Jussila’s PhD THM-model was implemented into COMSOL and an oral presentation was given in COMSOL meeting 17.-19.11. 2010 Paris, including an article in conference proceedings.

During the project discussions about possible collaboration in studies of microbial activity in bentonite was started by Merja Itävaara (VTT).

At GTK, the mineralogical characterisation of bentonite was planned.

In Aalto University’s Mathematical department, the numerical analysis of equations in earth mechanics was studied, especially Brinkman’s equations and mass conserving FE methods.

THM -MODELLING OF BENTONITE: FINITE DEFORMATIONS AND PLASTICITY

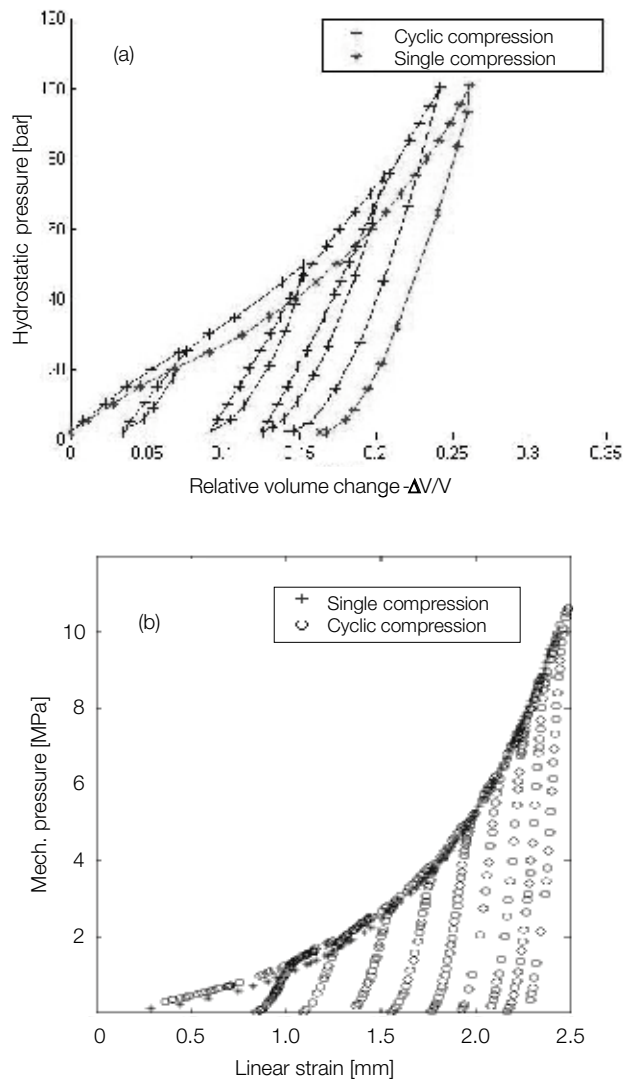
The original goal of this subproject was to generalize the previous model to include finite deformations. During the course of the projects, the objectives were extended to include plastic deformations and development of tailored experimental methods for measuring the relevant model parameters and transport mechanisms of moisture in compacted bentonite.

The previous THM model including only small deformations was successfully generalized to finite deformations. The generalization is based on a covariant spatial approach, which at least in a formal level preserves the original free energy formalism. In particular, the finite strain measure is constructed in terms of the metric tensor expressed in the coordinate system comoving with the deforming material. It appears, that the theory allows combining the hypoelastic and hyperelastic formulations of finite strains and thereby seems to also give a possibility to include finite plastic deformations in the theory.

In order to measure the relevant mechanical properties of compacted bentonite, two different experiments, namely hydrostatic compression experiment and one-dimensional compression experiment were designed. In the hydrostatic compression experiment, a cylindrical sample of compacted bentonite covered with liquid rubber coating is placed in the sample chamber equipped with a piston. The device allows measuring the volume change of the sample as a function of hydrostatic pressure. The same device was also used in one-dimensional compression experiment. For that purpose it was modified so that the piston head and sample chamber base was equipped with permeable rigid compression surfaces.

Examples of results of the two compression experiments are shown in Figure 17. In both cases, the (hydrostatic or mechanical) pressure is increased both with a single stroke and with repeated compression/release cycles to a maximum value of approximately 10 MPa. The results show plastic deformation with well identifiable yield points and hardening behaviour typical to many soils.

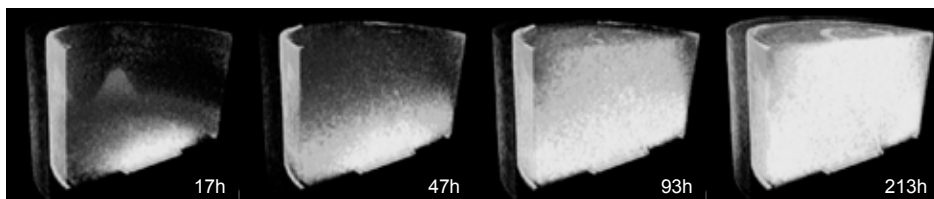
Figure 17. Typical results of hydrostatic compression (a) and one-dimensional compression (b) experiments.



TRANSPORT OF WATER IN COMPACTED BENTONITE. PRELIMINARY RESULTS

X-ray microtomographic techniques was used in order to study the basic mechanisms of water transport in bentonite. To this end, a carbon fibre sample chamber that allows water inflow to a bentonite sample at a constant volume was constructed. The dry sample was first imaged with x-ray scanner (SkyScan 1172). Wetting of the sample was then initiated by letting water through a channel and a sintered block into contact with the lower surface of the sample. Experiments were carried out for macroscopically identical samples varying the gage pressure of the wetting water in the range from 0 to 2 bar. The wetting procedure was continued for about two weeks during which it was interrupted approximately once a day and the sample chamber was weighed and scanned with x-ray tomograph. Figure 18 shows the series of images obtained as a difference of tomographic images taken at various times during the wetting period, and the image of the initial dry sample. Based on the first results it seems that intrusion of water in bentonite does not proceed as a distinct wetting front that would be characteristic to pressure-driven imbibition to porous material. No marked qualitative or quantitative dependence on pressure of water transport was observed. Furthermore, comparison with theoretical results obtained assuming either pure pressure-driven flow (Darcy's law) or pure diffusion also show that the qualitative behaviour of wetting process is better explained by diffusion than by Darcian flow. These preliminary results indicate that in the present experimental set-up, water transport is dominated by a dispersive mechanism such as diffusion of vapour in gas phase or diffusion of water in solid phase.

Figure 18. Transport of water in compacted bentonite. Shown are cross sections of a series of x-ray tomographic images of a cylindrical bentonite sample sealed in a closed volume and wetted through a porous block in contact with the lower surface. Light colour indicates higher mean density of material due to presence of water. The diameter and height of the sample is 30 mm.



CONCLUSIONS AND UTILISATION OF RESULTS

A very long term research has been carried out about bentonite, but there still exist many problems mainly due to complicated structure of bentonite, which makes it difficult to understand in the conditions and scales needed.

Advancement is though all the time happening, and new efficient experimental methods, like X-ray tomography and different XRD methods appear promising. New modelling approaches, where the goal is to combine THM- and TMC-modelling were implemented on new type computing platforms like COMSOL and Numerrin. Collaboration between different scientists was extended already in PUSKURI project and is even more extensive in the new BOA project starting during 2011.

Both the experimental and modelling methods developed support directly the nuclear waste management carried out in Finland during the coming years. Also many young scientists have started their PhD work in the PUSKURI project.

Reference

Olin, M., Rasilainen, K., Itälä, A., Pulkkanen, V.-M., Matusewicz, M., Tanhua-Tyrkkö, M., Muurinen, A., Ahonen, L., Kataja, M., Kekäläinen, P., Niemistö, A., Laitinen, M., 2011. The results of PUSKURI project (Coupled behaviour of bentonite buffer) will be published in the VTT Research Notes -series.

8.13 Dissolution of montmorillonite in groundwater simulants

Emmi Myllykylä, VTT

The behaviour of bentonite in alternating groundwater conditions is an important part of safety assessment of nuclear waste repository. The main mineral component of bentonite is montmorillonite $(\text{Ca}_{0.06}\text{Na}_{0.16}\text{K}_{0.025})(\text{Al}_{1.51}\text{Fe}_{0.205}\text{Mn}_{0.005}\text{Mg}_{0.28}\text{Ti}_{0.01})(\text{Si}_{3.99}\text{Al}_{0.01})\text{O}_{10}(\text{OH})_2$. The favourable properties of bentonite for nuclear waste disposal, like high swelling capacity and high cation exchange capacity are greatly based on the properties of montmorillonite. However, most of the process studies done so far, have concentrated on bentonite, which contains also accessory minerals. The presence of accessory minerals complicates substantially the interpretation of analysed data.

For modellers, it would be important to gain data, which has as little as possible error marginal and interpretation options. The solution background in the waters analysed from process studies, as leach- and elution experiments, has been chemically too complex. In addition, the concentrations of Al and Fe have mostly been so low, that their concentrations could not be analysed reliably.

The main aim in this project is to gain better understanding on the interactions between montmorillonite and groundwaters, forming of deformation products and secondary minerals. The results of the experiments are compared to the modelled results and the models are then hopefully advanced further. The purpose was also to develop analyses for low concentrations of Al, Fe, Si, and Mg in simplified groundwater simulants. The solution samples are analysed with high resolution inter coupled plasma mass spectrometer (HR-ICP-MS). The whole project was planned to last three years and it begun 2010. The first part of the project concentrated on dissolution of Na-montmorillonite. Within following years similar experiments are planned to be conducted with Ca-montmorillonite.

The project was started with a literature review. The purpose was to gain some background of dissolution, factors affecting to dissolution, and similar experiments of montmorillonite dissolution. The experiments were planned based on the review and pre-models of montmorillonite dissolution.

Before the actual experiment, the equilibria of groundwater simulants and the dissolution of montmorillonite as a function of time and pH were calculated. The thermodynamic equilibrium of groundwater simulants were calculated by using geochemical equilibrium model EQ3/6. By calculating the equilibrium of groundwater simulants, it was ensured (at least theoretically) that nothing precipitates from water itself. The compositions of calculated groundwater simulants are presented in table 5.

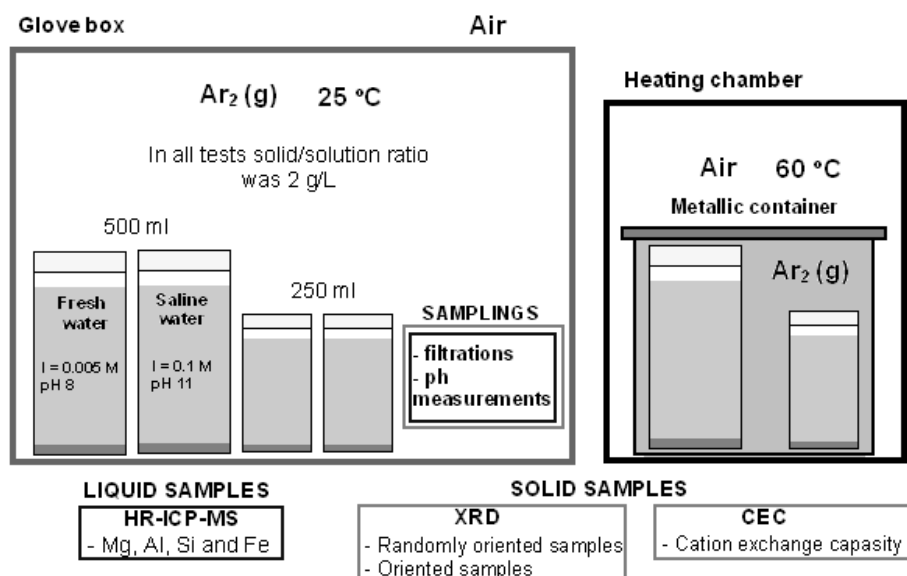
Table 5. The groundwater simulants used in the experiments, were calculated with EG3/6.

	Fresh water mmol/L	Saline water mmol/L
Na ⁺	3.23	40.38
Ca ²⁺	0.36	20.41
K ⁺	0.28	0.17
Cl ⁻	3.66	79.70
SO ₄ ²⁻	0.28	-
Br ⁻	-	0.24
pH	8	11
Ion strength	0.005	0.1

The experiments were conducted with purified Swy-2 montmorillonite in two types of water at 25 °C and 60 °C in anaerobic conditions (either in glove box, 25 °C or in pressure containers in the heating chamber, 60 °C). Fresh water had pH of 8 and ion concentration of 0.005 M. Moderately saline water had pH of 11 and ion concentration of 0.1 M. The solid/solution ratio was 2 g/L in all experiments.

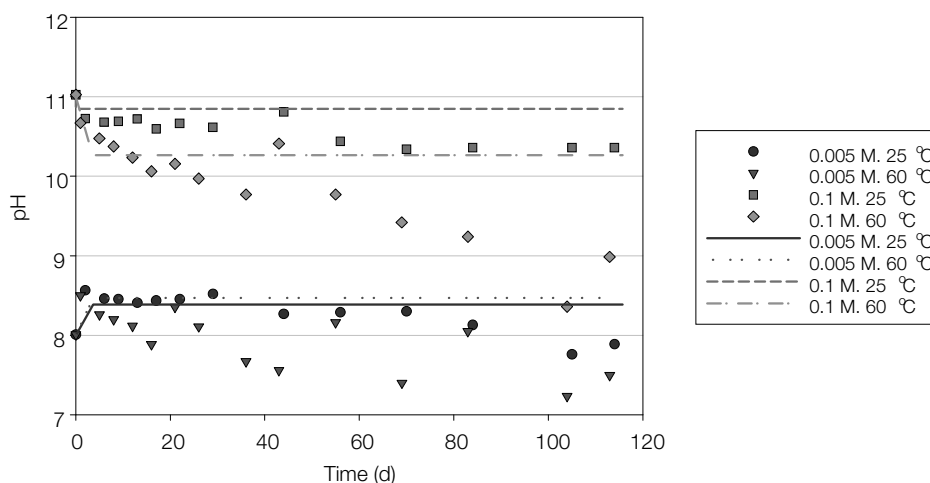
The experiments included two types of batch experiment. Ones with sequential sampling from the initial solid solution mixture and the others with exchange of the whole solution phase in few weeks intervals. The experiments and the analyses of the different samples are presented in figure 19.

Figure 19. Schematic view of the experiments. Experiments done in glove box (on left) and heat chamber (on right) and analyses made.



The sampling intervals for experiment were planned by using data from literature, from which an estimate for dissolution rate (as function of pH and temperature) was calculated. In addition, the dissolution of montmorillonite was modelled with TOUGHREACT, so that the salinity of waters is taken into account. The results gained from this “blind” model were compared with the experimental results (see figure 20). From the figure it can be seen that within the first 30 days the experimental and modelling results correspond quite nicely to each other but as time goes by, the model does not correspond to experimental results. This reveals the need for development of the models. Models are evaluated and interpreted in more detail according to research plan during next years.

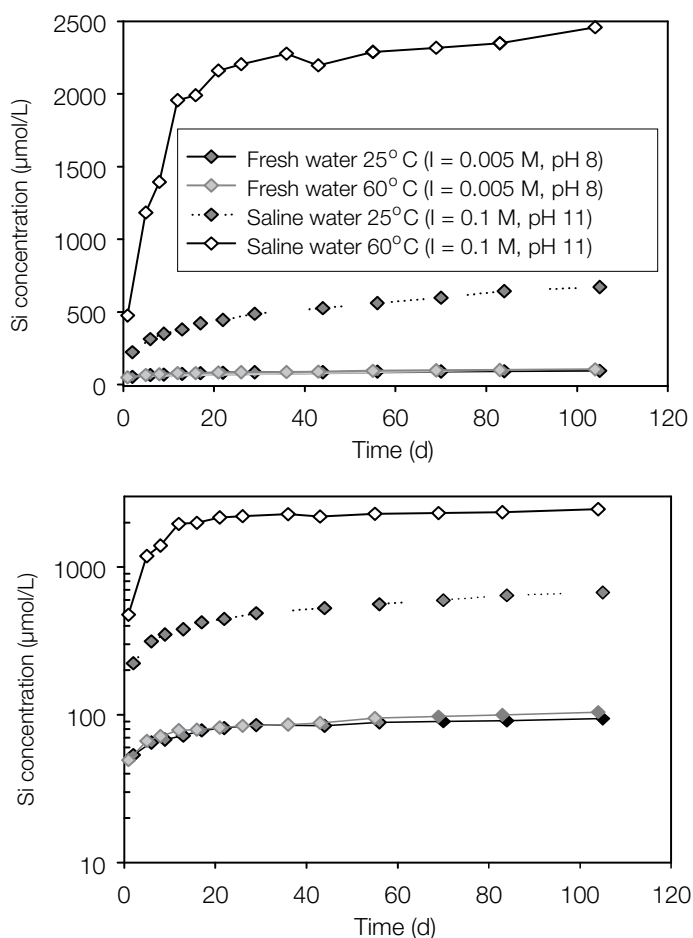
Figure 20. Experimental and “blind” modelling results for tests without solution exchange. The experimental results are marked as points and modelling results as lines.



In the experimental part, the concentrations of Al, Mg, Si, and Fe were analysed in ultrafiltrated (10 kD \approx 1 nm) samples. During the experiments, concentrations of Al, Mg and Fe showed only minor changes compared to Si concentration. The analysed concentrations of Al, Mg and Fe were between 0.01 and 2 $\mu\text{mol/L}$ and no clear increase in these concentrations was detected. Al and Fe concentrations fluctuated in most cases. Mg concentrations were usually higher at the beginning of the experiment than at the end, except in the case of moderately saline solution in 60 °C, in which concentration first decreased and then started to increase. These results agree with the observation from the literature, which indicate that especially Al and Fe take part of precipitation or sorb on developing secondary minerals. The concentrations of Si were much higher than concentrations for other measured elements. The results for Si are presented in Figure 21 and it should be noted that

these are preliminary results. In the multielemental analysis, high Si concentrations did not fall between used calibration points. To gain more exact results for Si, the samples are going to be analysed again from more diluted samples. From the figure it can be seen that the experimental conditions have a remarkable (even decade) effect on dissolved Si concentrations. In fresh water, near neutral conditions (pH=8), the temperature does not seem to have a significant effect on dissolution of tetrahedral layers, where as in experiments with high salinity and pH the temperature seems to have more remarkable effect on dissolution.

Figure 21. Preliminary results for Si concentration as a function of time in tests without solution exchange. The results are presented in linear (above) and in logarithmic (below) scale.



The purified montmorillonite and the solid material gained from experiments were analysed with XRD (X-ray diffraction) at ERM (Etudes Recherches Matériaux) in Poitiers, France. Actual change in nature of the smectite minerals was not observed (aluminous smectite was always present). Instead, the experimental conditions more or less modify the structure of montmorillonite (e.g. layer stacking). In mineral analysis halite (NaCl) and tridymite (SiO₂) were observed as potential secondary minerals. Halite might develop in the saline waters as the solution equilibrium changes or crystallize during sample preparation. Potentially formed trimyditite was observed only in warmer and saline conditions.

The peak positions varied in accordance to the cations present in the interlayer space of smectites and to the number of water layers (0, 1 or 2) present in the interlayer space. The main cation of purified montmorillonite, Na, seemed to be substituted by Ca in some extent in all experimental conditions used in these experiments. The substitution was seen to take place in saline waters, but also partly in fresh waters, in which the concentration of calcium was substantially lower. In addition to typical smectite peaks, the XRD spectra of samples from fresh waters showed peaks typical for mixed layer minerals. These peaks can refer to the presence of either randomly ordered illite/smectite or randomly ordered collapsed smectite/smectite layers. The shrinkage of interlayer spaces could also refer to the sticking of the layers e.g. by cementation. To gain identification and proper conclusions an additional work is needed.

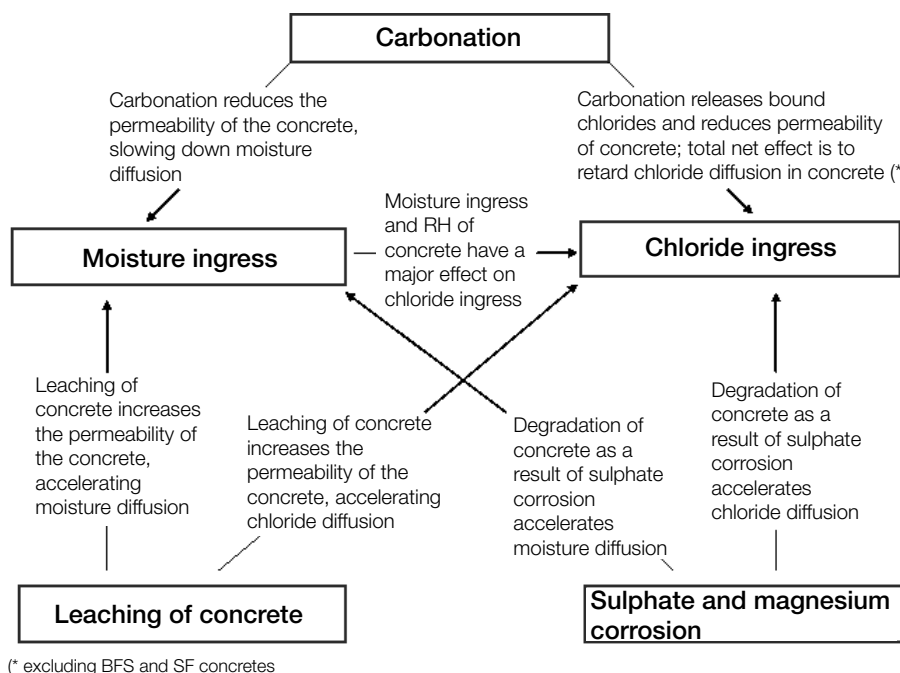
8.14 Modelling the durability of concrete for nuclear waste disposal facilities

Olli-Pekka Kari, Aalto University

MAIN RESULTS OF THE STUDY

The principal result of the study was a theoretical model that combines different deterioration mechanism of concrete. The mathematical formulation of the model is based on a group of differential equations that will be solved stepwise in time domain. The numerical implementation of the model was made by the finite element method applying a commercially available computer code. Thus, the model is exploitable publicly for simulating the interaction of different ageing mechanisms of reinforced concrete. The numerical implementation of the model was made keeping in mind particularly conditions of underground disposal facilities. This affected deterioration parameters included in to the model. The ageing mechanisms considered were (Figure 22): the carbonation of concrete by air; moisture ingress; chloride penetration; concrete corrosion caused by both sulphate and magnesium intrusion, and the leaching of cement paste compounds into groundwater.

Figure 22. Main ageing mechanisms and interaction.



The model can also be used to describe deterioration caused by a single ageing mechanism and for a time period typical for ordinary reinforced concrete structures. In addition, the effects of concrete admixtures, in this case silica fume and blast furnace slag, were included in the model. Therefore, as a by-product, the latest knowledge about the ageing mechanisms discussed were collected and analysed in the study. The finite element implementation is basically three dimensional, although the analyses made were two dimensional as the primary research objective was to explain the importance of interaction of various deterioration mechanism of reinforced concrete structures in final disposal conditions.

The results received clearly indicated that interaction between deterioration mechanisms changes the nature of intrusion of chlorides at the surface layer of concrete, where the primary concern is the time required of chloride from an external source such as saline groundwater to reach the location of main reinforcement bars. The intrusion does not obey a normal diffusion equation but the changes originated, for example, from the leaching of cement paste can be observed in calculated intrusion profiles. Concerning the leaching the explanation is that it affects effective diffusion coefficient of concrete matrix by changing local porosity of cement paste. The results highlighted also the importance to improve understanding concerning the role of sulphates for chloride intrusion. The literature review suggested that sulphate originated deterioration would have a pronounced accelerating influence on the intrusion of chlorides in to concrete. This was tried to clarify experimentally in this project, but the results did not support that kind of accelerating effect of sulphates. However, the role of sulphates for the chloride intrusion requires further experiments with concrete types intended to use in final disposal facilities to be sure of the sulphates influence on their lifetime. The results of the study suggested that the use of silica fume or blast furnace slag as a cement replacement with a low water-to-binder ratio is optional for the engineered barriers in final disposal facilities. The numerical simulations indicate that concrete layer at the surface of the structure should be 50 mm or more to protect the reinforcement against corrosion for 500 years in the conditions typical in bedrock caverns in Finland. The important finding was also to realize once again the complexity of concrete matrix. The model developed is general covering different mechanism. In a way, the model changes the intrinsic or aleatory uncertainty to epistemic uncertainty. The model itself does not increase knowledge. This was also observed in the model validation generating research needs that could not have been forecast at the beginning of KYT2010. The deterministic nature of the numerical implementation is to be changed to include uncertainties associated to the parameters of the model. The dominating uncertainties are application dependent. The results received indicate that the main parameter is the effective porosity for the studied disposal facilities. In the future it is important to improve the understanding of its variations on the surface layer of concrete, but statistical methods are necessary to quantify uncertainty in the model systematically. The required amount of chlorides in the pore water to initiate

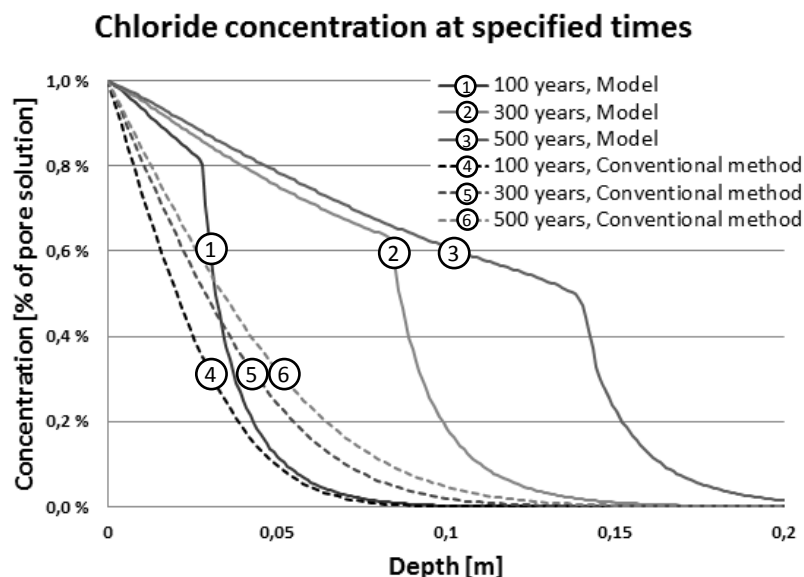
reinforcement corrosion under the particular disposal environment would also be a beneficial piece of information in improving the accuracy of lifetime estimations. Singling out these dominating parameters paves the way to define focus of further research.

SIGNIFICANCE OF THE RESULTS CONSIDERING THE RESEARCH OF NUCLEAR WASTE MANAGEMENT

The low- and intermediate-level wastes that accumulate during the operation of nuclear plants will be disposed of in an underground repository in the bedrock. The safety of the repository is ensured by multiple engineered barriers which also consist of concrete structures. It is required by YVL 8.1 2003 that the engineered barriers must be serviceable at least 500 years after the repository has been sealed. However, there is a lack of direct experience in reinforced concrete structures with a service life even close to that demanded, as reinforced concrete has only been used as a material for the construction of buildings for a little less than 150 years. It should also be noted that during the first decades the disposal facility is subjected to concrete carbonation and after that the structures are surrounded by saline groundwater. It is important that the change of the environment can be simulated by numerical models in advance.

Traditionally, the mathematical examination of the durability of reinforced concrete has been performed by studying each individual ageing mechanism one at a time. The results received in this project clearly indicate the necessity to consider the interaction between different aging mechanisms, which may support to use thicker concrete layers at the surface of the structure to protect the main reinforcement in disposal facilities. The effect of the interaction on the chloride ingress can be seen from the Figure 23, where are presented the chloride concentrations at specified times for the example case calculated by using the model and conventional single mechanism methods. The chloride penetration after 500 years according to the interaction model is much more harmful than the conventional method predicted. The project also reveals difficulties in applying short term test results or practices developed for ordinary concrete structures to the time period of 500 years. The model developed will help to recognise the phenomena which most probably restrict the lifetime of concrete structures of disposal facilities, which makes it possible to find proper design solutions.

Figure 23. Chloride concentration at specified times; Model vs. conventional method.



The use of the model developed is not restricted to reinforced concrete structures under disposal conditions, but it can be used also in the design and lifetime management of new or existing nuclear power plants. The model can also be adapted for design and lifetime management of conventional structures. The results support the research projects relating to service life management of reinforced concrete structures that are carried out in SAFIR -program. The nuclear companies as well as the companies working in the consulting business of safety analyses can utilise the results. The results of the study are expected to be useful also for controlling authority the Radiation and Nuclear Safety Authority Finland (STUK).

The research was performed in the Aalto University. The results received were immediately incorporated into teaching, where the value of the model is that it demonstrates the complexity of phenomena affecting the life time evaluation of load bearing structures and introduces to students present possibilities to simulate these phenomena numerically.

RESEARCH METHODS

The model was constructed by applying the various ageing theories that were available in the literature leading to a group of partial differential equations. The solution of the model was based on the use of the finite element method, which is an application of weighted residuals. In simulation were used commonly available software and PCs. The validation of the model was performed by using the results of experiments that were partly made in this research project.

The methods used in the experimental part of the research were a titration of chlorides, electron microanalyser, a gravimetric method for sulphate analyses, mercury intrusion porosimetry, Environmental Scanning Electron Microscope (ESEM) combined with an Energy Dispersive Spectrometer (EDS) and ordinary measurements concerning concrete carbonation. Three different kinds of concretes exposed to two different kinds of exposure solution were used for validation. Concrete mixes consisted of three different binder combinations with constant water-to-binder and aggregate-to-binder ratios. Silica fume (Micropoz) and/or blast furnace slag (KJ400) were used partly as a cement replacement in two mixes while one mix consisted of pure sulphate-resisting cement as a binder. The solutions contained either only chlorides or chlorides combined with sulphates and magnesium.

The estimation of the durability of disposal facilities was performed by assuming conservative boundary values and conditions. Carbonation was presumed to last 50 years during the operating phase of the repository. The length of the post-closure period, when the structure is submerged and exposed to various mechanisms of ageing, was assumed to be 500 years. The modelling was performed for the same test concretes that were used in the validation.

8.15 Rockmass classification, determination and visualization of rock quality and associated uncertainties using statistical interference and geological 3D and GIS software

Mira Markovaara-Koivisto, Aalto University

FOCAL RESULTS OF THE RESEARCH

The project aims to visualize and model rock quality parameters in 1-3-dimensions and to assess and visualize the reliability of the models produced. Initial data for modelling the rock quality parameters is gathered from geological observations, geophysical survey results and statistical methods. Assessing the reliability of the models will take into account among others distance from the direct observation to a modelled point and observation's quality.

Work began in 2006 with a grant from K.H. Renlund foundation. First it was clarified which rock quality systems are in use in Finland and abroad. Then nine representatives of the areas using rock quality systems were chosen for an interview on their practice. Results of the interviews were published in a domestic magazine, *Materia* issue 3/2010: Markovaara-Koivisto M: "Kalliomassan laadun karakterisointi käytännössä - haastattelu". In addition, for four rock fills in southern Finland, Mäntsälä, Lohja, Talma and Karkkila, were selected for subsequent rock quality research and development of methodology.

In 2007, with a funding from K.H. Renlund foundation, structural geological mapping and ground penetrating radar (GPR) survey was carried out in Mäntsälä. In Mäntsälä and Lohja also stereo photographs were taken of the rock walls. Since 2009 the project has been part of the Finnish Research Programme on Nuclear Waste Management. Rock fracturing in the Mäntsälä quarry was modelled in 3D with Gocad program. The model was based on the above-mentioned research methods. First article to be included in the doctoral dissertation of Markovaara-Koivisto was published in 2009 in EUREGEO Conference Proceedings. In ground penetrating radar survey based fracture network modelling the modeller has to take into account similarity between the reflections on adjacent lines and may use results of structural geological mapping in coupling reflections into surfaces.

The following rock quality parameters were calculated in the space containing the 3D fracture model: number of discontinuities in a cubic meter of rock and Rock Quality Designation (RQD value), which could be derived from the previous by an empirical formula. The topic was presented in a seminar given by the Finnish Geotechnical Society (SGY), Finnish Tunneling Association (FTA-MTR), the Finnish National Group of ISRM and Suomen Betoniyhdistys (BY). The second article to be included in the dissertation was published in the conference proceedings in 2009.

In 2007–2009 detailed discontinuity mapping in the Mäntsälä quarry was conducted with scanline method. In 2008 structural geological mapping was conducted along two ca. 10 km long intersecting survey lines in the Mäntsälä batholite area, where the quarry is located.

Here scanline method was used to gather location-related observations, but tools to visualize them were lacking. A simple MATLAB (Mathworks) script was written to produce 3D surfaces from the observations. These were imported into Gocad program to refine the 3D fracture network model based on GPR survey. Third article to be included in the dissertation was written about making the refined model and rock quality parameters derived from it. The article was published in IAEG conference proceedings. The article discussed how the difference in the dimensions of the observation and computational spaces affects the reliability of the calculated parameters.

Later this simple MATLAB script was developed into a visualization and analysis tool for observations made specifically with scanline method. In 2009, the tool was presented in the National Geological Colloquium. In addition a short summary was published. Dissertation will include an article that was submitted in 2011 to Computers & Geosciences journal. The script will be published as an annex of the article. In addition, the article demonstrates new ways to present the direction of data in the industry of engineering geology.

Predicting prevailing rock structures and fracturing in individual site from the geology or the surrounding area is being researched with a case study from Lohja. Structural geological observations, made from Lohja quarry in 2006, are compared with the geological 3D model of the surrounding areas. Building the geological 3D model started with gathering input data, information of schistosity, structures, fracturing and rock type, from the archives and databases of Geological Survey of Finland. In addition, the geological maps made from the area are used to determine rock type boundaries.

Discontinuity's effect on a GPR signal's behavior was studied under laboratory conditions. Palin Granit Oy gave to research rock blocks sawn to different sizes. Changes in the GPR signal were studied with varying distance between two blocks. An article will be written about the laboratory studies to the EAGE's Near Surface 2011 conference.

So far, dissertation has dealt only with direct or indirect (ground penetrating radar) observations. The next aim is to extrapolate structures, seen on the surface, deeper into the bedrock. Finally, fracturing and rock quality are studied theoretically through their parameters, and their values are predicted in spaces without any observations.

SIGNIFICANCE AND EXPLOITABILITY OF THE RESULTS

The results can be exploited in rock engineering projects in Finland and in areas with similar bedrock conditions. The algorithm to visualize rock quality parameters in 3D

is applicable especially in nuclear waste management, because such sites are well studied and geological and geophysical information is abundant.

The articles published during the project present practical examples of methods to model rock quality in 3D and discuss their pros and cons. The results can be directly utilized in the security-oriented rock investigations and data processing of final disposal, which was mentioned in the KYT2014 research program.

RESEARCH METHODS

Parameters of structural geological mapping were studied according to a form developed at Geological Survey of Finland, in a project for construction suitability of urban areas. The results were saved into an Access database (Microsoft) developed for the mapping form. A new logging form partially developed in this study was used for discontinuity mapping with scanline method. The new form took into account also the parameters for later use in 2D and 3D visualizations.

Stereo photography was used at the quarries to produce 3D images of the quarry's walls with SiroVision program (CSIRO). The images mimic the walls and their shapes. Orientations of the discontinuity surfaces seen on the 3D-images were studied with SiroJoint program (CSIRO).

GPR survey was used to study persistency of the discontinuities, seen on the rock walls, deeper into the rock mass. In the study RAMAC-GPR unit with shielded 250 MHz, 500 MHz and 800 MHz antennas were used. GPR results were interpreted Reflexw 4.5 software (Sandmeier Scientific Software). This program was chosen because with it, the reflections may be digitized and saved in ASCII format.

Digital GPR reflections from parallel survey lines were joined to form surfaces using Surpac software (Gemcom). Later the same work was conducted with Gocad program (ParadigmTM). Gocad program was used for making the 3D-discontinuity models, as well as the rock quality parameter calculations within the modelled space.

Visualization and analysis of scanline mapping results began with simple Excel (Microsoft) spreadsheet calculations. Soon this work was carried out with a tool developed in MATLAB language (MathWorks). In support of the programming work Mira Markovaara-Koivisto participated a MathWorks's MATLAB course organized in Helsinki 13-15th April 2010.

Already existing bedrock observations from Mäntsälä and Lohja were searched from the archives and databases of Geological Survey. Observations on the database can be browsed with ArcView software. However, Excel spreadsheets were used in data processing.

8.16 KABIO – Gases and biogeochemical processes in the crystalline bedrock

Ilmo Kukkonen, Lasse Ahonen, Arto Pullinen, Geological Survey of Finland

In studies related to nuclear waste disposal in crystalline bedrock it is important to know the chemical composition, gases and possible microbes of deep groundwater as well processes controlling these phenomena. Abiotic and biotic reactions between dissolved components in groundwater and minerals of the host rock may potentially affect the near-field behavior of the planned waste disposal system, its functionality and durability (especially the bentonite buffer and the waste canister). The project KABIO (2008-2010) focused on the composition of gases in crystalline rock and biogeochemical processes influencing the gases. Unknown factors are still related to biogeochemical processes which require compilation of basic information, hydrogeological sampling, modeling of processes as well as microbiological studies. The project KABIO was carried out in close co-operation with the GEOMOL project of VTT.

The project KABIO utilized the deepest drill hole in Finland located in Outokumpu. The 2516 m deep Outokumpu Deep Drill Hole provided a unique possibility to study fracture zones and deep fluids in a depth range which extend from surface to planned “repository depth” and up to more than 2.5 km below surface. Therefore it is possible to identify and study phenomena in a depth range much larger than the using conventionally much shallower drill holes. Although the lithologies in Outokumpu differ from those in Olkiluoto and the metamorphic degree is lower, the bedrock in Outokumpu can be considered quite typical of crystalline rock in general and fairly similar to that in Olkiluoto (excluding the altered ultramafic rocks in Outokumpu). Therefore, biogeochemical processes identified in Outokumpu can be expected to be present also elsewhere in crystalline rock where corresponding saline fluids and gases exist.

The project KABIO carried out sampling deep fluids in the Outokumpu Deep Drill Hole during all field seasons in 2008-2010. Fluid sampling was done with the traditional tube sampling method of GTK for defining the overall vertical characteristics of the fluid column in the hole. In addition, we developed methods and readiness for pumping fluids from fracture zones isolated by hydraulic packers. Sampling with pumping was developed in co-operation with Posiva Oy and Lapela Oy that constructed special inflatable packers for the Outokumpu hole (diameter 22 cm). Instrumentation for automatic monitoring of the pumping, fluid electrical conductivity, pH, Eh and redox potential was installed in the research hut next to the deep hole. Pumping periods ranged from 2 to 4 months.

According to recovered samples and earlier geophysical downhole logs (hole temperature and fluid electrical conductivity) it is known that the fluid becomes gradually more saline with time in the deep hole. It had been observed already

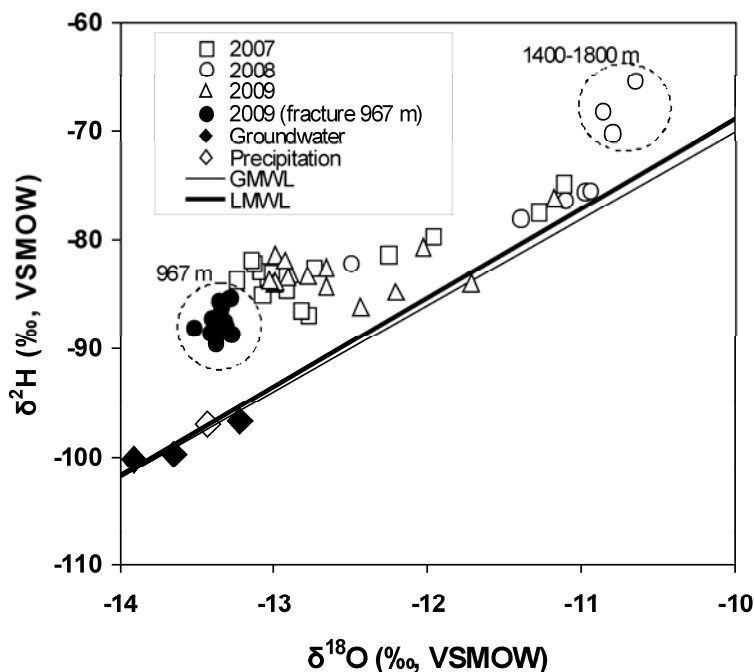
during drilling as well as after drilling. There are a few hydraulically fracture zones which are responsible for flow in and out of the hole. The flow rate is very slow, and surficial fluids participating in meteoric circulation are not present, not even in the uppermost part of the drill hole. It makes the Outokumpu Deep Drill Hole suitable for hydrogeological and microbiological investigations.

Table 6. Composition of gas in the Outokumpu Deep Drill Hole. Concentrations are given in vol-% at NTP conditions.

Depth (m)	N ₂	O ₂	Ar	He	CH ₄	C ₂ H ₆	C ₃ H ₈	Sum
500	22	0,51	1	2,2	72	0,56	0,014	98,3
600	15	1,2	0,9	0,35	79	0,8	0,018	97,3
700	17	0,42	0,69	0,53	78	0,68	0,017	97,3
800	18	0,37	0,6	0,42	77	0,66	0,018	97,1
900	26	0,53	0,52	2,5	67	0,46	0,012	97
1000	16	0,64	0,71	0,34	79	0,75	0,018	97,5
1100	28	0,45	0,66	2,1	66	0,45	0,012	97,7
1200	18	0,48	0,96	0,63	77	0,65	0,017	97,7
1300	21	1,4	0,71	0,7	73	0,62	0,017	97,4
1400	21	0,6	1	1,6	73	0,57	0,017	97,8
1500	22	0,64	0,72	5,6	68	0,5	0,019	97,5
Average	20,4	0,66	0,77	1,54	73,5	0,61	0,016	

The fluids in the deep hole are Ca-Na-Cl fluids with a high content of gas. Samples pumped to the surface release plenty of gas (fluid:gas volume ratio is about 1:1). The gas composition is mostly methane and nitrogen. Stable isotopes of water indicate that the deep hole fluids do not participate in meteoric circulation, but are results of long-term interaction between fluid and rock. Drill hole water seems to originate from separate fracture zones located at distances of several hundred meters of each other in the hole, and that are not hydraulically connected. Fluids from different fracture zones differ by their chemical, isotopic and microbiological characteristics.

Figure 24. $\delta^2\text{H}$ and $\delta^{18}\text{O}$ analyses of drill hole water in Outokumpu Deep Drill Hole from tube sampling in 2007–2009 and in pumped water from the fracture zone at 967 m. The fluids do not participate in meteoric circulation which is represented by global meteoric water line (GMWL, thin), the local meteoric water line (thick) for eastern Finland, the averages of groundwater in eastern Finland (solid diamonds), and the average of precipitation in Kuopio (open diamond). The circled data points represent different water compositions of water discharging from fracture zones at 967 and 1715 m and mixing with borehole water.



Microbiological studies were carried out in co-operation with the project GEOMOL of VTT since 2007. Microbes have been analyzed from both tube fluid samples and pumped samples from fracture zones isolated by inflatable packers. An important observation was that there is a ‘deep biosphere’ characterized by considerably diverse species. Molecular biological studies have shown that about 15–20 % of the observed bacteria and archaea carry genotypes resembling methanogenes and sulphate reducers reported elsewhere. A large fraction of the species detected in Outokumpu cannot be functionally classified yet.

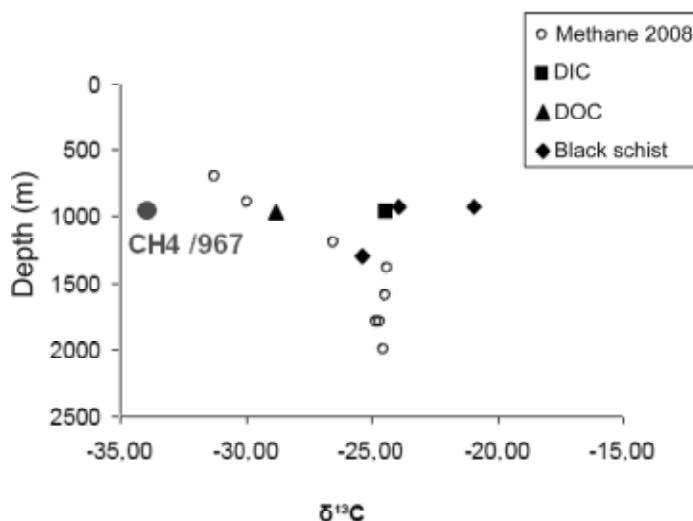
Special attention was paid in the KABIO project to the potential connections between gases, fluid chemistry and microbes in the Outokumpu Deep Drill Hole. The stable isotope composition of methane was analyzed in several samplings, and the results may indicate a thermogenic origin of methane. Thus, the methane in

Outokumpu would be very old and result from dissociation of longer hydrocarbons at high temperatures, or alternatively, methane has migrated from greater depths in the crust. On the other hand, biogenic origin of methane cannot be excluded, because biological cycling of carbon in a closed system would not produce any significant variations in isotope proportions in contrast to the starting materials. Observations of methanogenic archaea also support this hypothesis. On the other hand, water samples do not contain sulphate above detection limit which could also point to microbiological reduction of sulphate.

Fluid profiles obtained with tube sampling show that the C-13 composition of methane varies in the vertical dimension. Fluids pumped from fracture systems differ somewhat from this trend as well as graphite in metasediments. On the other hand, dissolved organic carbon, seems to fit with the methane trend in borehole water.

In addition to experimental studies, KABIO included also theoretical studies on potential energy sources of deep biosphere. The subject is highly relevant for understanding the biogeochemical and microbiological processes. They will support the modeling of macroscopically observed processes that may be relevant for disposal of spent nuclear fuel. Two basically different chains of energy transfer were considered: (1) Supply of energy from surface to deep levels with a heterotrophic electrons transfer chain (oxygenic respiratory system -> fermentative reactions), and (2) autotrophic chains of energy transfer independent of surface biosphere. One interesting example of such processes is oxidation of methane by reduction of sulphate. Methanogenic autotrophic processes require most probably hydrogen to act as an electron donor. In addition, we studied reactions involving sulphur as potential source of energy. The result of these thermodynamic considerations was that available flow of energy from the studied chemical components is small, even if compared with the geothermal heat flux. Therefore, possible diffusive flow of gas from deeper levels in the crust and mantle should be investigated as one option, but also hydrogen production by radiolysis from natural gamma radiation of the rock itself.

Figure 25. Carbon isotope compositions of dissolved organic (DOC) and inorganic carbon (DIC) and methane of drill hole fluid in the Outokumpu Deep Drill Hole compared with graphite of black schist interlayers. Isotope compositions ($\delta^{13}\text{C}$) are given with reference to PDB standard.



Educational aspects were an important constituent of the KABIO project. A master student project was included in the project, and geology student Taru Toppi (Department of Geosciences, University of Helsinki) prepared her Master's thesis in the project with the title "Composition and isotopes of deep fluids in the Outokumpu Deep Drill Hole".

An important spin-off effect was reached when the research project DEEP LIFE (2010–2013) was initiated as co-operative action of GTK, VTT and University of Helsinki. The project aims for investigating deep biosphere processes in the Outokumpu Deep Drill Hole. The project is partly funded from a research grant by the Academy of Finland and two doctoral students are working in the project.

8.17 Deep subsurface geomicrobial processes – Molecular monitoring methods – GEOMOL

Merja Itävaara, Malin Bomberg, VTT

Final disposal of high radioactive wastes requires consideration of safety aspects. Deeper knowledge of microbial processes is required for safety analysis about microbial risks connected to biocorrosion of copper, biodeterioration and biodegradation of materials, gas formation and microbial role in bentonite clay. The role of microorganisms has been considered positive phenomena as related to consumption of oxygen, however, on the other hand in anaerobic conditions reduction of sulphates due to the activity of sulphate reducing bacteria is an adverse effect and may cause corrosion of copper canisters. The stable long-term conditions expected to prevail in the repository are affected by geochemistry and microbial metabolites, the formation of gases and change in redox processes due to microbial activity.

Until now there is still little information about the diversity and function of deep subsurface geomicrobial processes. Majority of deep subsurface microorganisms do not grow on any growth media. Therefore this project aimed to generate information based on molecular methods about earth crust aquifer microbial communities in Fennoscandinavian shield.

The major aim of this project is to develop deep microbiological sampling techniques and molecular tools for deep geological environments. Sulphate reducing bacteria have been detected to be the major species involved in biocorrosion of copper and therefore we have been focusing on the developments of detection methods for sulphate reducers but also to methanogens which are known to be connected in the related redox processes. However, because microbial species in the communities interact and they have synergistic effects we found it very important to study the whole communities.

In the Geomol project, we have developed to know how about the deep sampling techniques performed in the field. In addition we have developed knowledge about preservation of the samples in the field and transportation to the laboratory for different analysis. In addition a transportable anaerobic chamber was purchased for direct pumping and anaerobic filtration of waters in the hood (Figure 26).

Figure 26. Field laboratory in Outokumpu.



Deep microbiological samples were taken by tube sampling technique and with PAVE pressurized sampling device as packer sampling during the years 2006–2009. The boreholes which were studied were located in Nummi-Pusula R-387, Outokumpu deep borehole (2,516 m), and Olkiluoto boreholes ONK-PVA3, OL-KR8, OL-KR42, OL-KR43 and OL-KR40. The tube sampling technique provided opportunity for parallel studies of microbiology, geochemistry and gases as a function of depth.

Microbial diversity did not correlate with microbial density, however, microbial density decreased as a function of depth from the surface from around 10^4 – 10^5 cells/ml to 10^3 cells/ml.

The diversities of microbial communities differed considerably in all the deep boreholes in Nummi-Pusula, Outokumpu and Olkiluoto in Finland. The density of microbial communities was analyzed from filtrated samples where microbes were stained with fluorescent dye and analyzed microscopically. Deep subsurface microbes were also studied by scanning electron microscopy (SEM). Microorganisms were detected by molecular methods based on 16S rRNA PCR-DGGE for bacteria and archaea. More specific detection of sulphate reducers was determined by functional genes, such as dissimilatory sulphite reductase (*dsrB*) gene and methanogens based on *mcrA* gene. In addition clone libraries provided wider information of the species involved. Water samples were filtrated to concentrate microbial biomass for molecular analysis.

Groundwater samples from Nummi-Pusula, natural uranium was studied by 16S rRNA-DGGE and uranium reducing microbial species were detected based on the sequence databank comparison. Nummi-Pusula was the first field experimental site, where the development of field sampling techniques and methods occurred in 2006.

The final disposal site of high radioactive wastes will be located in Olkiluoto. There are several boreholes at the area to study hydrogeological conditions in the bedrock

aquifers. The samples were received from Posiva Ltd. and had been taken by a pressurized sampling device PAVE after four weeks pumping between the packers. The sterilization efficiency of PAVE sampling device was studied after cleaning up with chlorine, because molecular techniques are more sensitive for contamination than culture based techniques. The sampling device was demonstrated to clean up well, instead the tubings and packers may form a source for contamination when transferred between the boreholes. The samples received from Olkiluoto all originated from different depths. In addition purification of the borehole section separated by the packers was detected as the decrease in microbial density and change in diversity which indicated cleaning up the borehole section and being replaced by endemic fracture water in the bedrock.

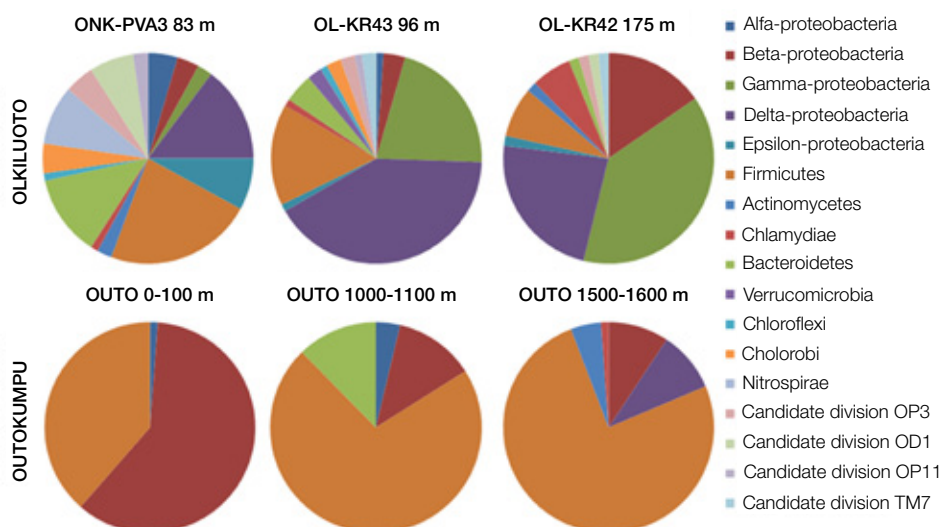
Environmental conditions affect the presence and activity of microbial communities. The role of depth and geology were found to be important factors affecting the presence of microorganisms. The salinity of groundwaters increased downwards which probably affected the presence of microbial communities in addition to pressure and geochemistry. Microbial communities changed downwards and different species were present as major communities at different sampling depths. Several environmental conditions such as changes in gases and geochemistry seemed to affect the results. Statistical analysis could not be performed because the samples were each representing different boreholes and different depth. However, some trend can be detected such as the effect of the increase in salinity due to depth and the tentative effect of black shale on microbial diversity. In addition the diversity seemed to be higher at depths with higher magnesium concentration.

The Outokumpu deep borehole (2.516 km) research is coordinated by Professor Ilmo Kukkonen, Geological Survey of Finland. The deep borehole has been drilled during 2004–2005 and has since been subjected to a major international geophysical and geochemical research. Samples for microbiological research within the framework of this project were retrieved during the years 2007, 2008 and 2009.

The microbiological studies of the Outokumpu deep drill hole showed clear differences in the composition of microbial communities at different depths (Figure 27). The correlation between geochemistry and the occurrence of microorganisms was clear from the studies of the borehole, which was studied throughout its depth profile. Sulphate reducing microorganisms appeared in all the examined samples, although it was found that in the deeper samples of the Outokumpu borehole the sulphate reducers belonged to different species, mostly autotrophic carbon dioxide fixers, than those in the surface samples.

The sulphate reducers usually live close to the zones of methane-producing microbes, if mixing of the water column does not occur. At 1000 m depth, methane is bubbling into the borehole from a fracture in the bedrock. Saline water also flows into the borehole from the other fracture zones, which produces currents and causes some degree of mixing of the water in the borehole. However, the mixing is not strong enough to prevent the stratification of the microbial communities.

Figure 27. The composition of the microbial communities in Olkiluoto samples (upper row) and Outokumpu deep borehole samples (lower row) based on clone libraries of the bacterial ribosomal 16S genes.



Some bacterial species have been shown to occur throughout the borehole depth profile, for example *Hydrogenophaga* sp. However, the relative amount of this group decreases downward in the borehole and species belonging to the Firmicutes group increase. Of these, a significant proportion are sulphate reducers. At 1000–1500 m depth of the Outokumpu deep borehole the microbial communities have been found to be richer than at other depths.

Both autotrophic CO₂-fixing and heterotrophic organic matter decomposing sulphate reducers were found. Methane-producing microbes, or methanogens, were found at specific depths, but not in all samples. Anaerobic methane-oxidizing microbes occurred in some samples. The relative number of sulphate reducers in the groundwater of Olkiluoto was generally low, at the most 0.5% of the total microbial community. In Outokumpu deep borehole water, the proportion of sulphate reducers was much higher, up to 7–20%. The proportion of methanogens of the entire microbial community in both places was low, only 0–2%. By linking the geochemistry and the gas composition of the water with the microbiology indicated that the rock types have an important role for the occurrence of different microorganisms in the bedrock groundwater.

The presence of sulfate in groundwater is not a prerequisite for the presence of sulfate reducing microorganisms, since several species have the ability to break down organic matter. The presence of organic matter and the cooperation of sulfur oxidizing microbes may erode the bedrock and release sulfur compounds, which are immediately transformed to corroding sulphides by microbial redox processes.

Cooperation between different groups of microbes, such as anaerobic methane oxidizers and sulphate reducers, can create conditions where corrosive sulphides are produced. Microbial metabolites are another potential risk factor. Microbes are able to bind nitrogen and form ammonium ions, as well as amines, both of which are corrosive to copper.

In the Geomol research project, methods for sampling and analysis of the microbial communities were developed and have created a strong foundation for studying the deep geological environments and geomicrobiology in Finland. During the project, two PhD students, Lotta Purkamo and Maija Nupponen, have started their studies with funding received from other sources. Pauliina Rajala is also performing her master's thesis work within this project.

Researchers have annually participated in international educational activities in Austria, Germany and the USA, as well as in a number of international conferences where the research findings of the project have been presented. The lack of geomicrobiological training opportunities has also lead VTT to participate in the design of the Nordic geobiology training programme, which will start in the summer 2011 in Iceland.

Research scientists involved in the Geomol project at VTT are PhD Mari Nyssönen, MSc Aura Nousiainen, MSc Anu Kapanen, PhD Malin Bomberg. The project is led by Principal Scientist, Docent Merja Itävaara, VTT. The Geomol project was a joint project with Docent Lasse Ahonen (GTK) during 2006–2008, after which the sampling for geobiochemistry was separated to Kabio project. Collaborative research with GTK received funding from the Academy of Finland for the project Deep Life (2010–2013). The investigation relates to the Outokumpu deep borehole and stable isotopes, the exploitation of deep microbial processes in energy research.

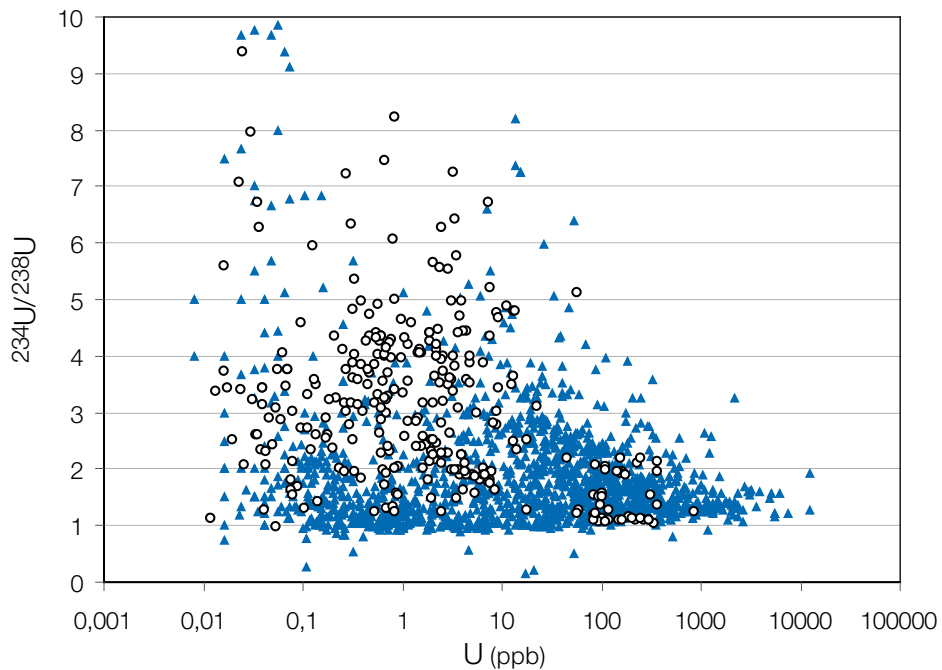
8.18 Behaviour of uranium as an indicator of groundwater conditions

Juhani Suksi, University of Helsinki Laboratory of Radiochemistry

INTRODUCTION

Understanding of changes in groundwater conditions is central part of the safety analysis of spent nuclear fuel repository. The performance of technical barriers as well as release and transport of radioactive substances depend on the chemical conditions of the groundwater. Input of different recharge waters (precipitation, sea water, glacial melt water) is known to introduce temporal changes in groundwater conditions but their importance to safety has not been clarified conclusively. The purpose of the present project was to examine the use dissolved uranium in studying chemical impact of infiltrations. In the interaction between infiltration and flow channels' wall U isotopes are transferred into water modifying $^{234}\text{U}/^{238}\text{U}$ ratio in groundwater. Because the $^{234}\text{U}/^{238}\text{U}$ ratio does not change but remains the same in water after the original conditions have restored, it can in principle be used in assessing chemical conditions where isotopes were transferred into water. U concentrations and isotope ratios presently seen in groundwater (Fig. 28) can be considered mainly as a result of infiltrations occurred after the last glaciation. Due to the long half-lives of the isotopes the effect of radioactive decay on the formation of the $^{234}\text{U}/^{238}\text{U}$ ratio and its change in water can be considered insignificant. Why the $^{234}\text{U}/^{238}\text{U}$ ratio varies in groundwater as seen in Figure 28? Can one use this variation in clarifying the impact of infiltrations? These questions were presented at the start of the project. The final report of the project in Finnish can be found in Suksi (2009).

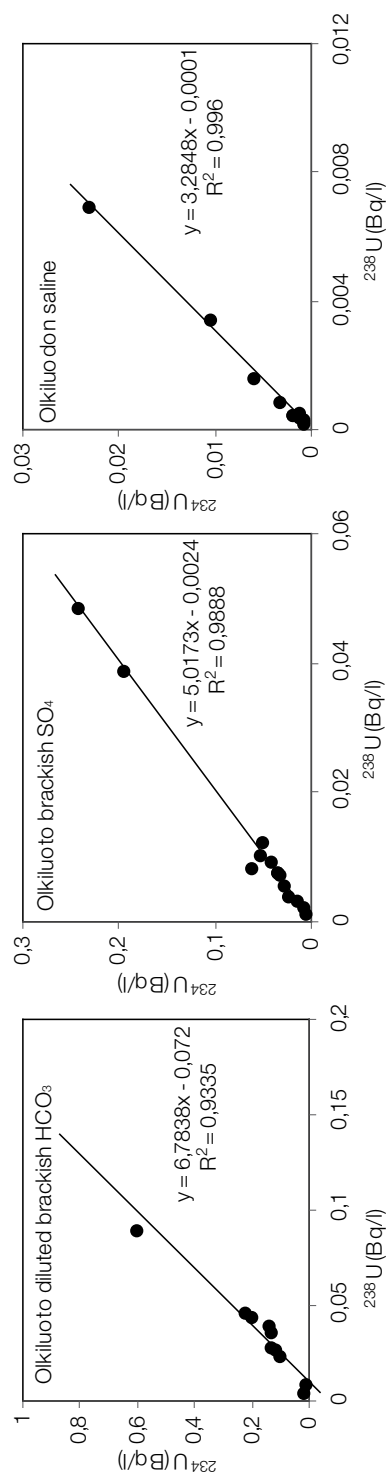
Figure 28. U concentrations and isotope ratios in groundwaters in different study site. Triangles represent drill well samples. Largest U concentrations occur generally in the oxidising upper bedrock. Large variation in the U concentration reflects variation in rock types and labile U available in flow channels. Labile U consists of secondary U compounds associated in weathering products which are distributed in spaces between rock forming minerals. Generally larger U concentrations in drill wells are probably due to drilling induced change in chemical conditions that favours U dissolution. Variation in the $^{234}\text{U}/^{238}\text{U}$ ratio is due to the concentration variation of the more mobile ^{234}U isotope. The reason for variation in the ^{234}U concentration is believed to be infiltration of waters into the bedrock.



MAIN RESULTS OF THE STUDY

The main result of the study was to show the dependence between the U isotope ratio $^{234}\text{U}/^{238}\text{U}$ and groundwater conditions. The dependence could be shown by comparing U isotope release ratios in different groundwater types. The release ratio was determined from the measured isotope concentrations using linear regression. Figure 29 shows the results of the regression analysis of three groundwater types in Olkiluoto. Excellent correlation can be explained by successful sampling.

Figure 29. Determination of U isotope $^{234}\text{U}/^{238}\text{U}$ release ratio (slope).



In the shore line (Olkiluoto) $^{234}\text{U}/^{238}\text{U}$ release ratio varied according to groundwater types (Table 7). At inland sites (Kivetty ja Romuvaara) such variation could not be seen. The probable reason for the variation in the shore line is its more versatile infiltration history. In Olkiluoto and Hästholmen the influence of infiltrations during the development of the Baltic Sea is seen. In Olkiluoto the largest release ratio value (~6.8) was obtained in dilute brackish HCO_3^- -water in the upper bedrock and the smallest value for saline water (~3.3). The groundwaters at Hästholmen show the same trend: brackish SO_4^{2-} -water has larger release ratio than saline water, 3.6 and 2.6, respectively. That the largest isotope release ratio is obtained in dilute brackish water is an important observation because the water represents input of normal recharge, i.e. infiltration of chemically the most reactive water type. Based on the observations in this study one can say that U isotope release ratio depends on the salinity of the water and probably also on redox-conditions.

Table 7. The dependence between U isotope release ratio and groundwater conditions.

Study site	Hydrochemistry	$^{234}\text{U}/^{238}\text{U}$ - release ratio	Remarks
Olkiluoto	Dilute, brackish HCO_3^-	6,8	Dilute brackish HCO_3^- water down to 150 m, hydrogeologically dynamic zone; infiltration during the Litorina Sea stage down to 300 m, melt water remnants; groundwater becomes saline at 400–500 m depth.
	Brackish SO_4^{2-}	5,0	
	Brackish Cl^-	3,3	
	Saline	3,3	
Hästholmen	Brackish SO_4^{2-}	3,6	Same feature as in Olkiluoto; groundwater becomes saline at 500–600 m depth
	Saline	2,6	
Romuvaara	Flow path I	3,8	Dilute brackish water down to several hundred meters; evolution from Ca-Na-Mg- HCO_3^- -type to Na-Ca- HCO_3^- -type. Marks of melt water are missing.
	Flow path II	3,0	
	Flow path III	3,4	
Kivetty	Flow path I	4,3	Dilute brackish water down to several hundred meters; same type of evolution as in Romuvaara; marks of melt water infiltration

The project investigated also uranium data from over 1200 drill wells (see Fig. 28). Compared to the other groundwater data the $^{234}\text{U}/^{238}\text{U}$ -activity ratio in part of the samples was below equilibrium value unity. Similar values were not observed in normal groundwater samples. The unusually low ratios can be explained by drill wells which were made with much larger diameter drill ($\phi 15\text{--}25$ cm) than usual drill holes (4–5 cm). Drilled wells have probably changed groundwater flow system so that oxidising water has flowed into anoxic flow channel dissolving there old ^{234}U -poor uranium. The $^{234}\text{U}/^{238}\text{U}$ activity ratio below unity can thus be considered as an evidence of changed redox-conditions.

THE IMPORTANCE OF THE STUDY IN THE NUCLEAR WASTE MANAGEMENT

One of the central questions in the nuclear waste management is whether groundwater conditions remain sufficiently stable at the nuclear waste repository depth during the forthcoming infiltrations. To answer the question conclusively necessitates understanding of the hydrogeochemical evolution of the site and detailed knowledge of the influence of different recharge waters. The technique developed in the project offers a new way to study infiltrations. The use of the knowledge of infiltration effects in safety argumentation can make sense to non-experts more easily than does computational reasoning. One can also derive justifiable information from the observations to be used in computational sensitivity considerations. Infiltration observations have also an important role in forming alternative conceptual models and interpretations, which can be used in constructing and modifying scenarios for safety analysis.

THE METHODS

The study was carried out mainly by analysing published material of U isotope concentrations in groundwater. Material was collected from Finland and Sweden. Linear regression was in the analysis of U data. Groundwater chemistry from the study sites, knowledge of the hydrogeochemical evolution at the sites and U isotope nuclear geochemistry were used to interpret the results. The preliminary analysis has been published in Suksi et al. (2006). The U isotope results from a migration experiment performed in natural water conducting fracture were also utilised in the study. The experiment has been published in Kienzler et al. (2006).

The aim of the second phase of the project was to study $^{234}\text{U}/^{238}\text{U}$ release ratio in controlled conditions to better connect the formation of $^{234}\text{U}/^{238}\text{U}$ ratio to U dissolution and chemical conditions. For that purpose a water-rock-interaction simulator was developed in the Laboratory of Radiochemistry. The apparatus was tested but the planned investigations had to be closed down due to insufficient funding.

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8.19 Fractured bedrock and hydrogeology

Lasse Ahonen, Geological Survey of Finland

In the changing conditions of the earth's surface, crystalline bedrock provides a stable environment for the final disposal of waste generated by the nuclear power industry. In case of a possible failure of technical barriers, bedrock acts as a barrier for the released radioactive emission, ensuring the safety by means of various hydrogeological and hydrogeochemical retardation processes. Safety case of the final disposal requires that features and processes of the bedrock are well understood, can be quantified and classified as well as simulated by models of different levels.

The occurrence of fractures in different scales is a characteristic feature of crystalline bedrock. With respect to southern Finland, fracture formation is evidently associated with the about 1800 million years geological history of the rock. Features and processes of the fractured crystalline rock differ in many respects from those of the much younger clay, salt and volcanic formations. Project "Fractured bedrock and hydrogeology" started within the previous KYT program and was completed by the end of 2006. The aim of the project was to carry out a combined methodological study, which integrates structural geology, hydrogeological and geophysical measurements, drill core logging and video recording of the borehole wall.

Initially research was concentrated on the Kopparnäs site in Inkoo, and later in 2006 also on the Palmottu natural analogue site, after which emphasis of the research was addressed to the Outokumpu deep drill hole. Work proceeded from the interpretation of maps and aerial photos to drilling through one of the interpreted fracture zones, and to the investigations carried out in the drill hole. Mode of occurrence of fractured zones was interpreted using the drill core material and compared with the results of the borehole video recording. Both Kopparnäs and Palmottu research drill holes were widely studied by means of various geophysical measurements, and observations obtained were compared with fracture zones verified by other methods, especially by video recording.

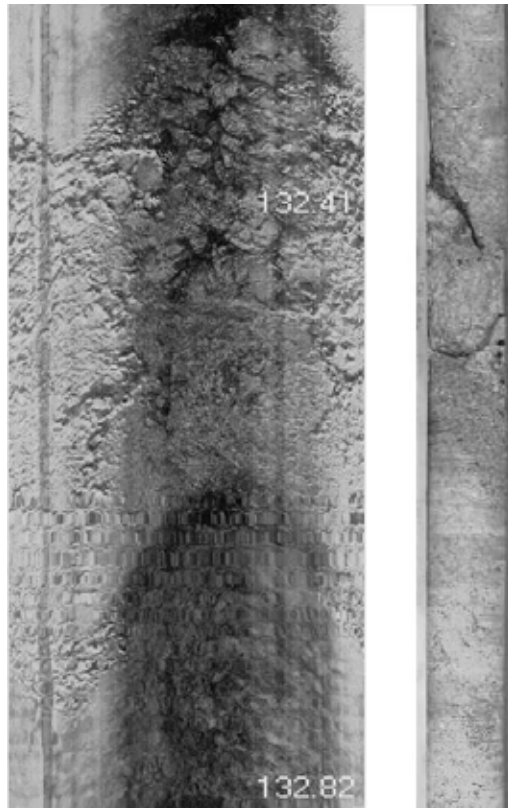
In the Kopparnäs study, a good quality video record of the drill hole wall was obtained, including detailed picture of the fracture apertures, which then was compared with fractures interpreted from the drill core (Figure 30). Furthermore, comparison of geophysical drill hole log with the observed fractures produced supporting information to be used in interpreting existence of fractures, if direct observations are not possible.

At the Palmottu site (DH387) additional work was carried out to develop and test drill hole geophysical methods to be applied in hydrogeology. This borehole was selected, because Posiva difference flow log was performed earlier in the borehole. It was observed that additional information can be obtained in drill hole logging, if the hole is pumped contemporaneously, because flowing fractures can be more easily detected. It was also observed that self potential log along the drill hole may give

hydrogeologically significant information, because self potential anomalies were detected at the depth levels, where earlier difference flow logging had indicated fracture flow.

By the end of the project, research was increasingly directed to the new Outokumpu deep drill hole, where a master's thesis (technology) was done using the drill hole geophysical data. In total, two master's theses were completed in the framework of the project, and young scientists had a significant work contribution as a whole.

Figure 30. The drill hole wall and fracture apertures.



8.20 Grimsel Test Site – Phase VI and Long Term Diffusion –project (LTD)

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BACKGROUND

In Finland, as in many other countries, high-level radioactive waste is planned to be disposed of in deep-lying crystalline rock. There has been a tendency to play down the potential role of the geosphere as a safety barrier in repository performance assessment. Among other reasons current uncertainties in transport pathway definition and pore space characterisation of crystalline rock can be mentioned. Repository safety evaluation today requires going from laboratory and surface-based field work to the underground repository level. Little is known about the changes to rock transport properties during sampling and decompression. Recent investigations using resin impregnation of the rock matrix at the Grimsel Test Site imply that non-conservative errors in calculated transport properties derived from laboratory data may reach factors of 2/3. This may lead to over-conservative estimates of limited matrix diffusion depths.

The Long Term Diffusion (LTD) Project aims at obtaining quantitative information on matrix diffusion in crystalline rocks. *In situ*, lab-based experiments and pre- and post modeling are planned. In Phase I, an *in situ* monopole diffusion experiment aims to quantify the extent of matrix diffusion in an undisturbed rock matrix. In addition, *in situ* porosity and pore connectivity will be studied separately by an impregnation experiment. Laboratory-based studies include analysis of natural tracer profiles, analysis of diffusion processes in archived samples as well as support of the *in situ* tests.

A multi-method approach was performed, consisting of a series of three main *in situ* test and two lab-based studies of existing core material from the Grimsel Test Site. The work consisted of four work packages: WP 1 Monopole experiment, WP 2 *In situ* porosity determination, WP 3 Natural Tracer Studies (NTS) and WP 4 Diffusion Processes Study (DPS).

RESULTS

Clear demonstration of *in situ* radionuclide matrix diffusion was defined in the rock matrix, especially for the suite of weakly and non sorbing elements. The verification of the conceptualisation in existing transport codes (via predictive and post-mortem testing) of the diffusion into the rock matrix has been achieved by comparing the field and laboratory data. The characterisation of the spatial distribution of porosity in the matrix, and its link with the mineralogy, in order to better identify microstructure-derived residence time distributions in the matrix has been demonstrated.

The diffusion behavior of the three radioactive elements; Na-22, Cs-134 and HTO and the stable isotope of I-127 were successfully studied in *in situ* conditions at the Grimsel Test site within the LTD project Phase I. Na-22 penetrated into a depth of 6 cm from the injection bore hole wall and Cs-134 penetrated into a depth of 2.5 cm during 2.5 years time. HTO and iodine which are classified as non sorbing elements penetrated to several tens of centimeters into the Grimsel granodiorite matrix. Figures 31 and 32 show the diffusion profiles of Na-22 and Cs-134.

Figure 31. The diffusion profile of Na-22 into the Grimsel granodiorite rock matrix during 2.5 years *in situ* experiment.

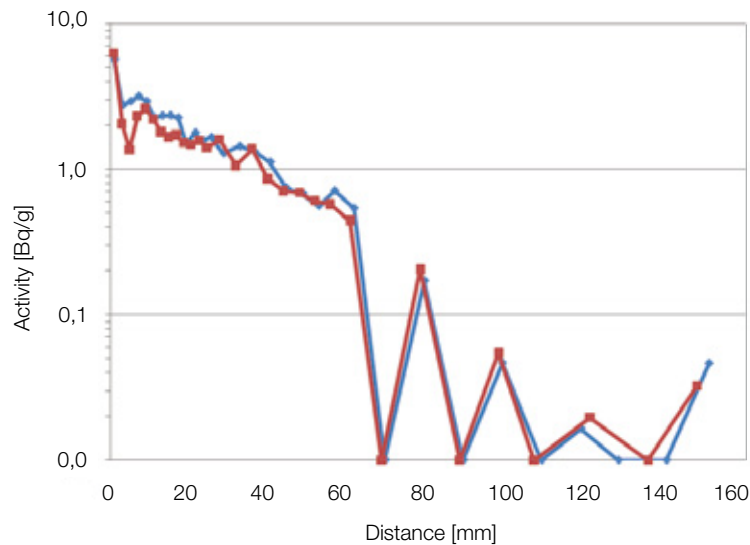
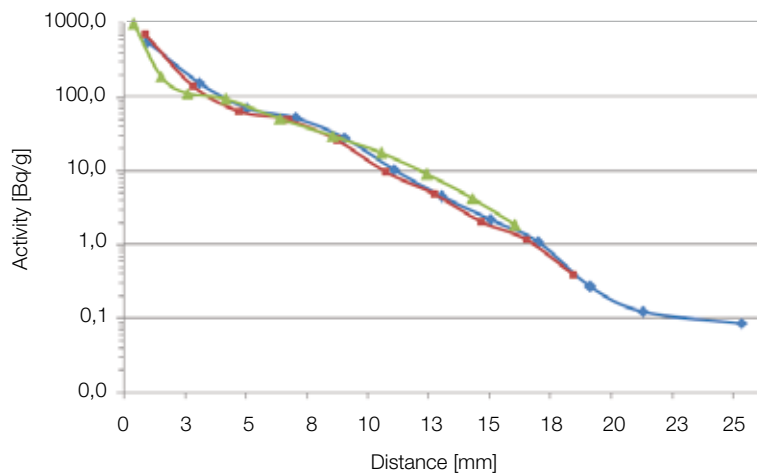
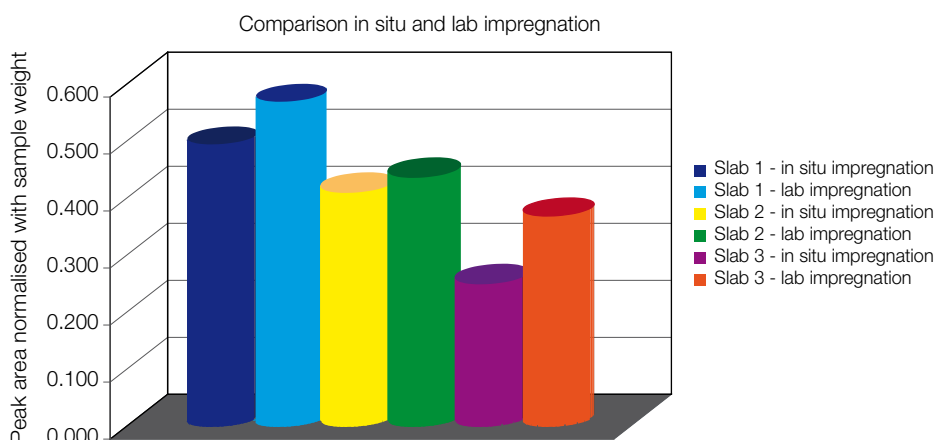


Figure 32. The diffusion profile of Cs-134 into the Grimsel granodiorite rock matrix during 2.5 years *in situ* experiment.



The *in situ* porosity values were found to be 10–30 % lower than the laboratory based porosity values of the Grimsel granodiorite rock cores. The results obtained from *in situ* study suggest that stress relaxation found in the micro fractures of the rock samples can affect to this difference between *in situ* and laboratory based porosity values. Figure 33 shows the comparison between the porosities from *in situ* experiments and laboratory experiments.

Figure 33. In situ porosity values are compared to the laboratory based porosity values for Grimsel granodiorite.



USABILITY OF THE RESULTS

C-14-PMMA impregnation based technique for use *in situ* characterization of rock porosity has been developed within this project. The know-how gained is transferrable and can be utilized in the context of different nuclear waste repository situations. Safety-relevant information has been obtained regarding *in situ* radionuclide migration, radionuclide-rock matrix interactions, rock structure adjacent to the water conducting flow paths. Groundwater chemistry has been investigated in terms of long-term migration behaviour and cesium sorption in *in situ* conditions. The understanding regarding these processes has increased as a result of the aforementioned investigation. These results have been utilized in modelling and evaluating the radionuclide retention of rock matrix in bedrock migration studies with rock heterogeneity taken in to account. During this project several radiochemists have been trained and familiarised within national and international scientific community to face the challenges of the nuclear waste reposition.

METHODS AND MATERIALS USED IN THIS STUDY

The autoradiography method was used to measure porosity of the rock samples. The samples were impregnated with ^{14}C -labelled methylmetacrylate, which was

then polymerized by irradiation to the pores of the samples. Sawed and polished subsamples are then placed on the X-ray film. Radioactive decay of the ^{14}C in the rock pores forms an image of the rock matrix to the film. Subsequent image analyses tools were applied to determine the porosities of the rock samples. Quantitative measurement of total or mineral-specific local porosities has been also obtained using image analysis tools. Electron microscopy examinations and mercury porosimetry measurements have provided detailed information of pore and fissure apertures.

Laser Ablation Inductively Coupled Plasma Mass Spectrometry (LA-ICP-MS) was used to determine the migration pathways of the radionuclides (Np-237, U-235, U-234) adjacent to the water flowing fractures. Normal Inductively Coupled Plasma Mass Spectrometry was used to measure elemental concentrations in groundwaters as well as I-127 from *in situ* diffusion experiment. Liquid scintillation counting and gamma spectrometry were used for measuring radioactive elements.

PHREEQC, hydrogeochemical modeling software was used to model selenium behavior in the rock matrix. This software is useful in speciation, solubility and precipitation calculations.

8.21 Fracture flow and radionuclide transport in granitic rock

Pirkko Hölttä, University of Helsinki Laboratory of Radiochemistry
Antti Poteri, VTT

Retention of radionuclides in geosphere can be an important component of the safety case for deep geological disposal of radioactive wastes. Performance assessment is directly concerned with the contribution of mobile radionuclides e.g. ^{129}I , ^{36}Cl ^{99}Tc , in their long-term exposure risks. The most important retention process of the solute transport in fractured rock is matrix diffusion. However, due to required very low flow rates and long time scales it is not possible to study directly the matrix diffusion under the flow conditions that will prevail around the closed underground repository. Estimation of the radionuclide transport and retention need to be based on the identification and understanding of the processes. Block fracture and core column migration experiments were performed to evaluate the simplified radionuclide transport concept used in assessing the safety of the underground waste repositories.

In earlier KYT programs, rock block migration experiments were introduced to examine fracture flow and radionuclide transport in a horizontal natural fracture. Rock core column experiments were introduced to estimate the diffusion and sorption properties of Kuru Grey granite used in block scale experiments. Results from column experiments were used to estimate radionuclide transport times and retardation parameters in the fracture before block scale experiments. The objective of this study was to examine the processes that cause retention in solute transport through rock fractures. Especially, the focus of the work was on the matrix diffusion. The results can be used to estimate the importance of the retention processes during transport in different scales and flow conditions. In 2006–2007, previous results were aggregated and additional experiments were performed in order to explain uncertainties and demonstrate the effects of matrix diffusion.

Block scale migration experiments were performed using Kuru Grey granite block. An experimental set-up for a block is illustrated in Figure 34. The block contains a natural hydraulically conducting fracture (0.9 m x 0.9 m) intersected by nine vertical boreholes, which were equipped with injection or sealing packers. Water pools were installed on the vertical sides and top of the block in order to ensure the saturation of the block and to stabilize the hydraulic head on the vertical faces. The block was instrumented also at the outer vertical boundary of the block where the horizontal fracture intersects the faces of the block for the collection of the tracer. Flow path tests with uranine dye tracer showed that migration took place through distinct channels. In parallel with the block scale experiments core column experiments were performed to estimate the diffusion properties of Kuru Grey granite. Core columns illustrated in Figure 35 were constructed from cores drilled to the fracture

and were placed inside a tube to form a flow channel representing an artificial fracture formed by the 0.5 mm gap between the core and the tube. Several tracer tests were performed both in a natural fracture and core columns using uranine, HTO, ^{36}Cl , ^{131}I as non-sorbing and ^{22}Na as a slightly sorbing tracer with injection flow rates of 0.7 to 20 $\mu\text{L min}^{-1}$.

Transport of tracers through the flow channels in a core column or in a natural fracture was modelled using an advection–dispersion model based on the generalized Taylor dispersion. It was assumed that a linear velocity profile existed across the flow channel, from zero velocity to some maximum flow velocity, and that the flow field and molecular diffusion perpendicular to the flow dominate the transport of the tracer particles in the mobile pore space of the fracture. Measured and modelled breakthrough curves through the core column are presented for uranine and ^{22}Na in Figure 36. Effects of the matrix diffusion in the breakthrough curves can be observed when results for non-sorbing and sorbing tracer are compared side by side. The model explains the transport of the uranine for the lower flow rates well. Applying exactly the same model to sodium breakthrough curves, only taking the sorption of the sodium into account, show very good agreement with the measured breakthrough curves.

Figure 37 shows experimental and modelled results for a block fracture experiment. Two transport channels were active in the analyzed experiment that makes it possible to examine dynamic behaviour under changes of flow rate. Different sorption properties of two tracers make it possible look at matrix diffusion properties along the same flow path. The same model that was used to examine core column experiments was applied to the fracture experiment. There are strong indications that effects of the matrix diffusion were observed in the fracture flow experiment because the dynamic behaviour of the breakthrough curves is well explained by the matrix diffusion model for both channels, i.e. for different flow rates. The same sorption properties for sodium as in the core column experiment explain the difference in breakthrough curves between uranine and sodium and the tailings of the sodium breakthrough curves is typical for matrix diffusion. Results give strong evidence that sodium breakthrough curves are affected by matrix diffusion. The same model is able to reproduce advection–dispersion dominated the breakthrough of the uranine and even predicts that matrix diffusion effects are beyond the range of measured data. The same model is able to reproduce the breakthrough curves of the sodium. Comparing modelled results for sodium with and without matrix diffusion supports the conclusion that tailings of the sodium breakthrough curves are affected by matrix diffusion.

Tracer tests were performed for a mobile tracer ^{131}I using flow rates 0.7–10 $\mu\text{L min}^{-1}$ in a core column and 3–10 $\mu\text{L min}^{-1}$ in a block fracture. Examples of the measured and modelled breakthrough curves are presented in Figure 38. The model used earlier for fracture column experiments and the same transport parameterization that were used to examine uranine and sodium experiments were applied to independently

interpret iodide results. Comparing modelled results for iodide with and without matrix diffusion supports the conclusion that tailings of the iodide breakthrough curves are affected by matrix diffusion.

Two different experimental configurations could be modelled applying consistent transport processes and parameters. The processes, advection–dispersion and matrix diffusion, were conceptualized with sufficient accuracy to reproduce the experimental results. The results provided show that it is possible to investigate matrix diffusion in low porosity crystalline rock at the laboratory scale. The effects of matrix diffusion were demonstrated on the slightly sorbing sodium and mobile iodine breakthrough curves. The modelled experiment builds confidence on the model predictions of the solute retention in groundwater flow. This knowledge and understanding of the transport and retention processes is transferable from the laboratory scale to in-situ conditions though specific parameters cannot be transferred directly to the spatial and temporal repository scale. 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 the radionuclide transport in the expected repository conditions.

FIGURES

Figure 34. Kuru Grey granite block having a natural hydraulically conducting fracture (0.9 m x 0.9 m). The vertical boreholes are equipped with injection or sealing packers. Water pools equipped with adjacent tracer collection channels ensure saturation and stabilize the hydraulic head around the vertical faces.

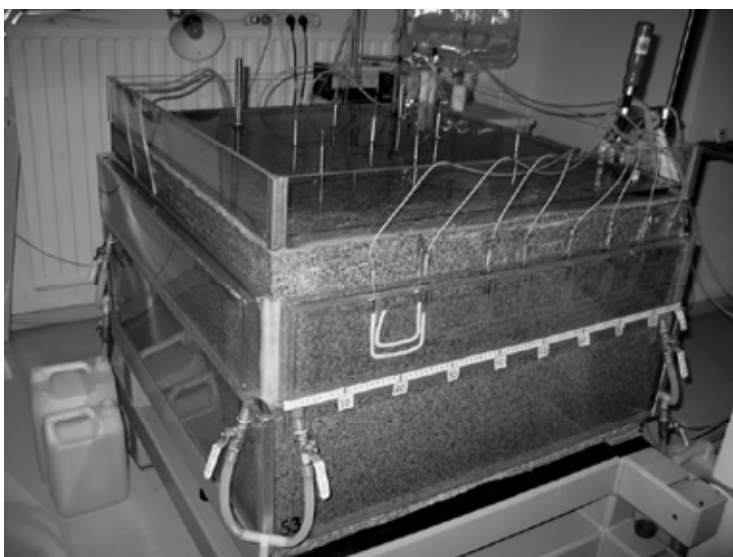
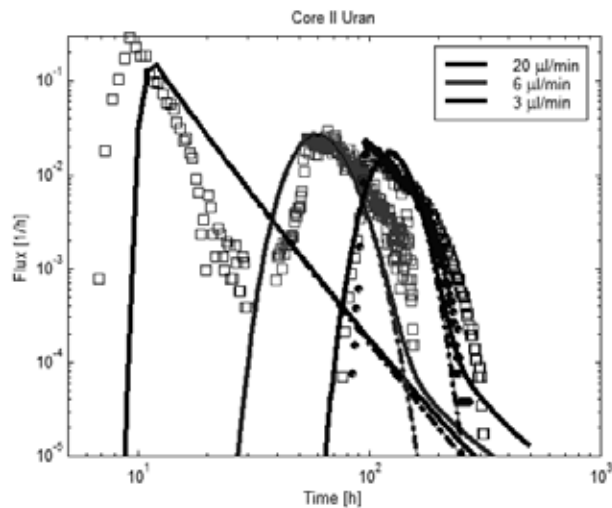


Figure 35. Experimental design used in core column experiments. Cores drilled to the fracture of the Kuru Grey granite, are placed inside a tube to form an artificial flow channel in a gap between the core and the tube.



Figure 36. Modelled and measured breakthrough curves of uranine (above) and ^{22}Na (below) through a core column with flow rates of $20 \mu\text{L min}^{-1}$, $6 \mu\text{L min}^{-1}$ and two experiments with flow rate of $3 \mu\text{L min}^{-1}$ (squares and full circles). Solid lines are model results for advection–dispersion and matrix diffusion. Dotted lines are for the advection–dispersion only.



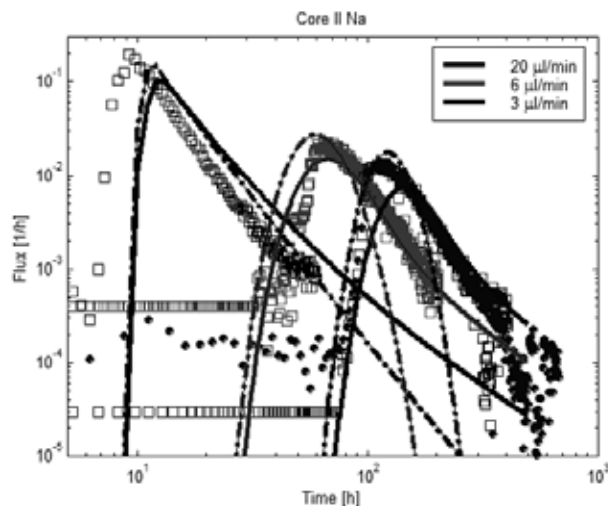


Figure 37. Modelled and measured breakthrough curves of uranine (full circles) and ^{22}Na (squares) through a natural fracture with injection flow rate of $10 \mu\text{L min}^{-1}$. All breakthrough curves are normalized to give a unit mass in the experimental time scale.

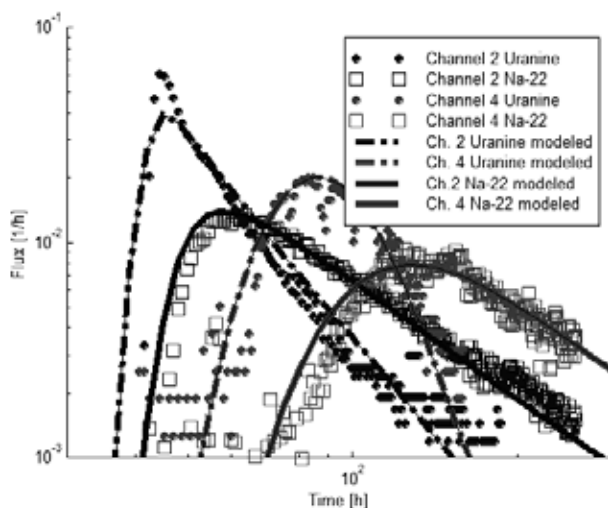
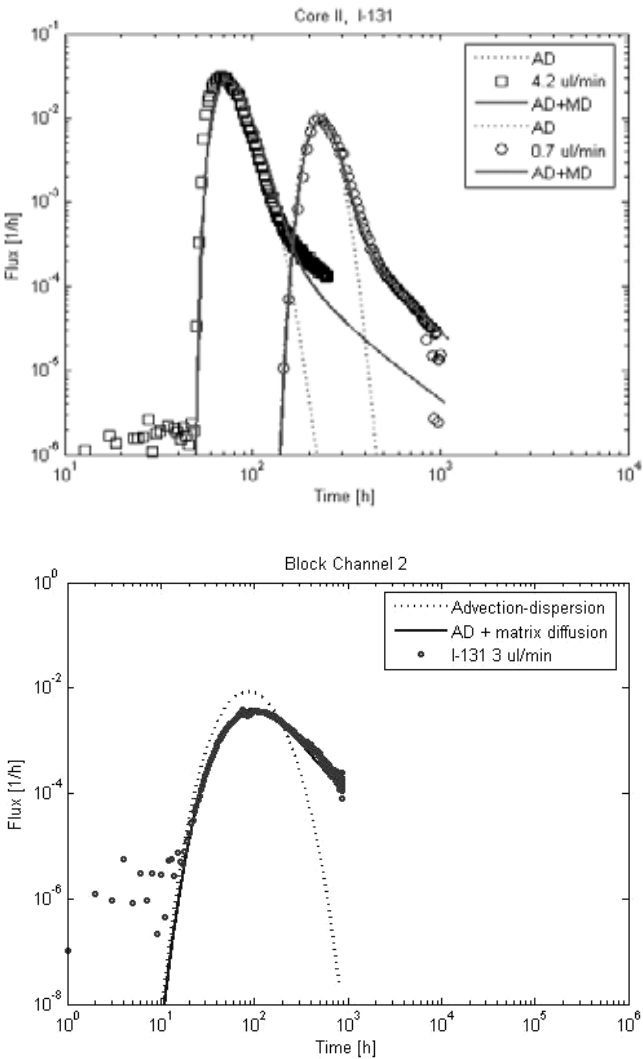


Figure 38. Modelled and measured breakthrough curves of ^{131}I through a core column (above) and through a natural fracture channel II (below). Solid lines are model results for advection–dispersion and matrix diffusion. Dotted lines are for the advection–dispersion only.



8.22 Colloid formation and their relevance in promoting the transport of radionuclides

Pirkko Hölttä, Mari Lahtinen, University of Helsinki Laboratory of Radiochemistry

Colloid-facilitated transport of radionuclides may be significant to the long-term performance of a spent nuclear fuel repository. In a colloidal system, solid particles dispersed in liquid form suspension where particle size in one dimension ranges from 1 nm to 1 µm in diameter and surface-to-volume ratio is very high. For colloid-facilitated transport it is essential that colloids are generated, radionuclides associated with colloids are transported with the groundwater flow. The potential relevance of colloids for radionuclide transport is highly dependent on the stability of colloids in different chemical environments and their interaction with radionuclides. Colloids can be produced from degraded Engineered Barrier System (EBS) materials such as the bentonite clay buffer, bentonite-crushed rock back-fill, the copper-iron container and grouting materials as well as from uranium fuel itself. The objective of this project was to determine the release and stability of inorganic colloids, study bentonite erosion in colloidal form and test and apply characterization methods.

In the batch dispersion experiments, powdered MX-80 type bentonite, crushed rock, low-pH cement or colloidal silica were added to deionized water, saline OLSO or low salinity Allard reference water. In the additional colloid stability experiments, MX-80 bentonite powder was added to diluted OLSO reference groundwater, sodium chloride and calcium chloride electrolyte solutions whose ionic strengths were adjusted between 0.001–0.1 M. The colloidal particle fraction was separated by filtration and the pH, particle size distribution, zeta potential, colloidal particle concentration and morphology were determined. Colloidal particle size distribution was determined applying the dynamic light scattering method (DLS/PCS) and zeta potential applying the dynamic electrophoretic mobility. Colloidal particle size distributions were partly also determined applying asymmetrical flow field-flow fractionation (AsFFF). Colloid concentration was estimated applying the DLS measurement count rate which is roughly proportional to the concentration of particles in the suspension. The morphology of colloids was determined by field emission scanning electron microscopy (FESEM). The elemental composition of colloids was analyzed using inductively coupled plasma-mass spectrometry (ICP-MS).

The particle size distributions determined applying DLS and AsFFF are shown in Figure 39. In general the particle size distributions are quite similar and the differences can be explained by the basis of the methods. DLS is based on the analysis of the temporal fluctuation of the scattered laser light intensity and the particle size strongly depends on the shape of the particles. It is very insensitive to small particles in the presence of larger sized particles, which produce the major part of the scattering light intensity. AsFFF is based on a physical fractionation of particles according to their size resulting in a complete size distribution of particles

in the sample. In low salinity Allard, mean particle size varied from 60 nm to 350 nm. In saline OLSO, particle size range was wide, from 1 nm to 900 nm. The morphology of strongly altered tonalite colloids in Allard water is illustrated in Figure 40. Rock and bentonite colloids seemed to be misshapen discs like particles and the particle diameters were in agreement with the DLS and AsFlFFF determinations. In low salinity Allard, zeta potential values were -10 mV – -45 mV. The lowest negative values were determined for bentonite colloids in Allard water. In saline OLSO, zeta potential values were near zero (+10 mV – -10 mV) indicating particle aggregation and sedimentation and instable colloidal dispersion.

The stability of bentonite colloids in diluted OLSO and electrolyte solutions did not change substantially during one year. The stability of colloids strongly depended on the ionic strength of the solution and the valence of the cation. Zeta potential for bentonite colloids in diluted OLSO reference groundwater is presented as a function of ionic strength in Figure 41. The zeta potential was approximately -40 mV at 0.005 M ionic strength and increased to -10 mV at ionic strength 0.07 M. In monovalent sodium chloride solution colloids were stable (zeta potential under -30 mV) when the ionic strength was 0.04 M or lower. In divalent calcium chloride solution colloids were only stable in the ionic strengths of 0.01 M or lower. The particle sizes grew as the ionic strength of the solution got higher. In both electrolyte solutions, the particle size was approximately 400 nm at ionic strength 0.001 M. In sodium chloride, the particle size exceeds 1000 nm at ionic strength 0.025 M. In calcium chloride, the particle size exceeds 1000 nm at ionic strength 0.015 M and after that point particles are much larger than in NaCl.

In saline OLSO, zeta potential values near zero, wide particle size range and low colloid concentrations indicated particle aggregation and sedimentation resulting in instable colloidal dispersion. In low salinity Allard and in deionized water, high or moderate negative zeta potential values, smaller particle sizes and higher colloid concentrations than in OLSO indicated the existence of stable colloids. At current conditions in Olkiluoto, the ionic strength of the groundwater is around 0.5 M and the colloids released from the bentonite barrier are aggregated and unstable ones do not have an influence on the migration of radionuclides. However, this knowledge and understanding of bentonite erosion in colloidal form can be utilized in the estimation performance of the bentonite barrier. The possibility of a future glacial period and subsequent post-glacial phase when the infiltration of fresh, glacial melt water dilutes groundwater, implies that dilute groundwater conditions cannot be excluded and the influence of bentonite and other colloids has to take into consideration.

Figure 39. Particle size distributions from rock and bentonite samples determined by AsFIFFF (Left) and DLS (Right). A = strongly altered tonalite, B = mica gneiss, D = moderately altered tonalite and E = MX-80 bentonite after four months exposure.

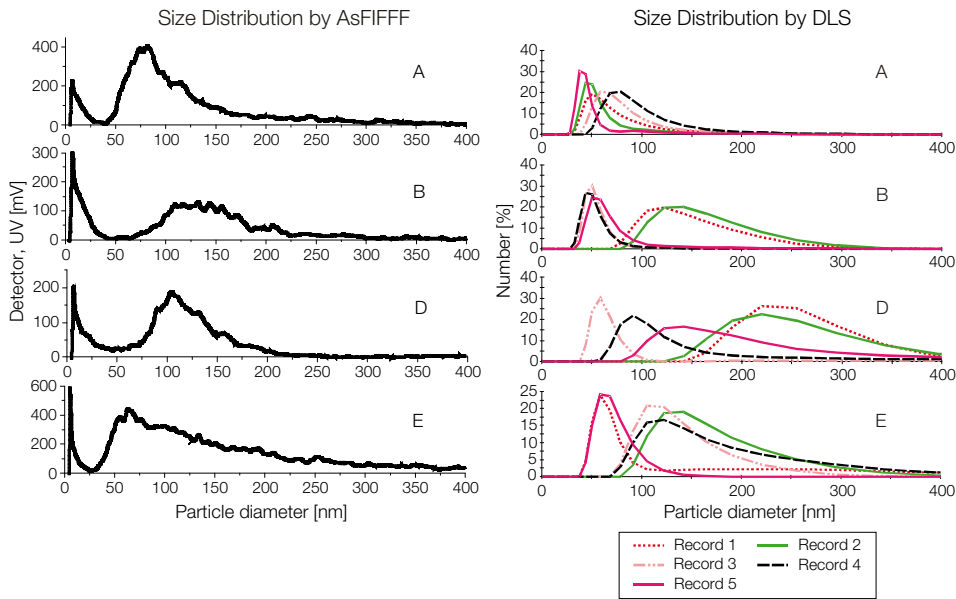


Figure 40. Field emission scanning electron microscopy (FESEM) images of colloids released from strongly altered tonalite in Allard reference groundwater.

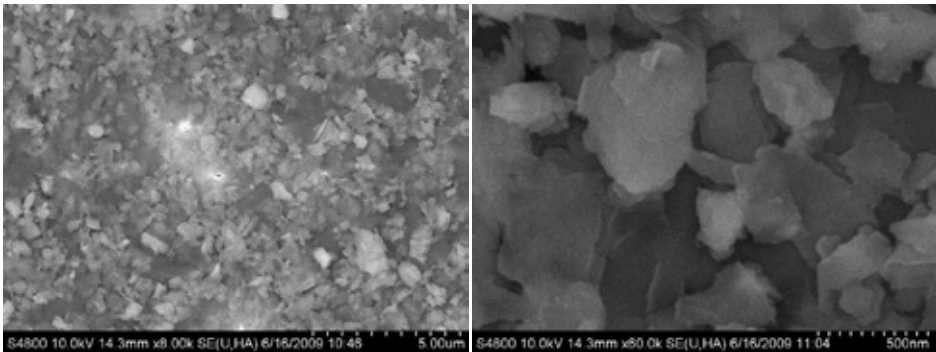
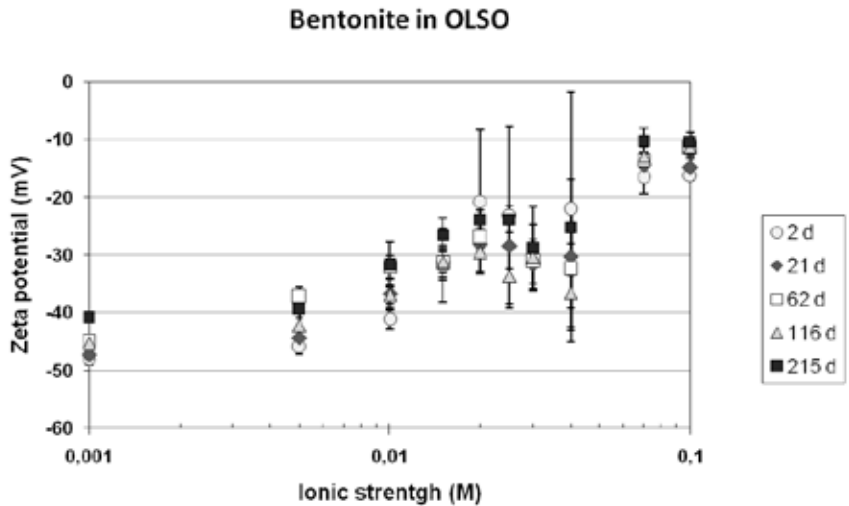


Figure 41. The zeta potential of bentonite colloids in diluted OLSO reference groundwater as a function of ionic strength.



8.23 Imaging of rock porosity by tomographic methods and combining it with matrix diffusion modelling

Jussi Timonen, University of Jyväskylä

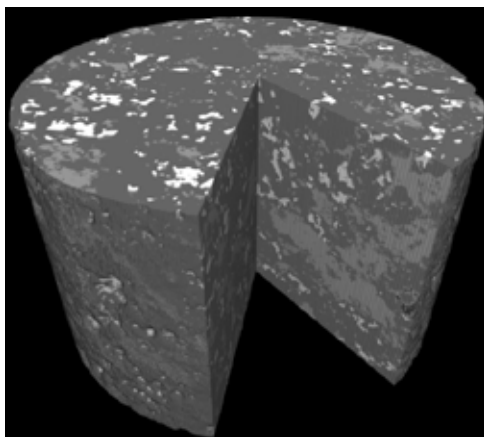
The first phase of the project, Imaging of rock porosity with tomographic methods, was realized in 2006-2008, and the second phase, Combining the three dimensional distribution of porosity with matrix diffusion modeling, in 2009-2010. Both phases of the project were part of the research effort dedicated to advancing the long term safety of the final disposal of spent nuclear fuel, and were closely connected to the Long Term Diffusion (LTD) project in the Grimsel Test Phase VI programme.

During the project period, in 2009, two new x-ray scanners were acquired and put in use: Wradia micro-XCT-400 with a best resolution of 0.7-1.0 μm , and Xradia nanoXCT with a best resolution of 50 nm. In addition to these scanners equipment for sample preparation were also acquired, and imaging methods for rock samples in particular were significantly developed.

Use of x-ray tomography in three dimensional materials research demanded a considerable effort on the development of image analysis methods. The widely used two dimensional methods are typically too slow for the large data sets related to three dimensional images. First of all it is essential to remove noise from the images before they are analyzed. To this end, several different methods were tested and their performances compared, and finally a new method based on monitoring the local variation in the noise variance was developed. This method proved to be both fast and efficient. Segmentation of the images is necessary for their quantitative characterization. Several new methods were developed for segmentation. Different structural features often demand different means of analysis, and thus methods were developed for identifying pores, micro-cracks, distribution of mineral components and surfaces/interfaces. In addition, methods were developed for the determination of pore size (and pore throat) distributions and specific surface areas from tomographic images.

Close collaboration with the laboratory of Radiochemistry of the University of Helsinki (HYRL) was involved in combining the porosity analysis by the radiocarbon-labelled PMMA method with the rock structure analysis by x-ray tomography. With the PMMA method (conducting) porosity below the resolution of x-ray tomography can be made visible although the spatial resolution of the method is not very high otherwise. Using in addition electron microscopy, mineralogical analysis of the samples studied could be done such that mineral specific porosities could be determined. Combining such analysis with the information about three dimensional distribution of minerals gained by x-ray tomography, three dimensional distributions of porosity, such as the one shown in Fig. 42, could be determined.

Figure 42. Three dimensional distribution of porosity in altered Sievi tonalite. Dark shades of gray indicate low and light shades of gray high porosity.

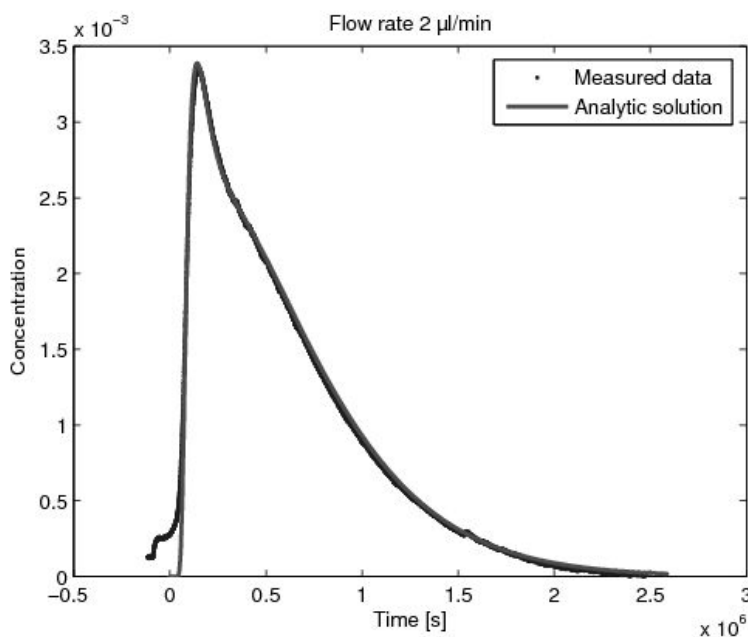


During the project several types of rock were analyzed: Kuru granite, Grimsel granodiorite and granite, samples from excavation disturbed zones, concrete (aged for 40 years under specific conditions, collaboration with the University of Berkeley), bentonite (transferred later to another project) and Sievi tonalite in particular. The last type of rock received special emphasis since alteration has made it very porous (about 10%), which made analysis and thus method development much easier. Apart from porosity, micro-cracks and three dimensional mineral distributions, also pore size distributions and distribution of porosity of small sample volumes, e.g., were analyzed.

In the second phase of the project, the above methods were utilized for improved and more accurate analysis of matrix diffusion of radionuclides in the bedrock. To begin with the model widely used for modeling matrix diffusion was validated. For several different cases analytical solutions to the relevant advection-diffusion problem were derived, most of which were new. A laboratory system was constructed in which potassium chloride was used as the tracer and water as the solute, and the spatial and temporal evolution of tracer distribution was measured via local conductivity of water. For two special cases, the measured breakthrough curve was compared with that of the corresponding analytical solution with excellent agreement. New specific features related to the finite depth of the porous matrix could successfully be detected. There after a fully numerical model for matrix diffusion was developed in collaboration with the University of Poitiers in France. This model was designed to utilize the three dimensional porosity distributions resulting from combining x-ray tomography and PMMA analyses. In this way we could analyze the effect of heterogeneity on the in-diffusion of tracer from a water conducting fracture. In addition, a similar modeling could be applied to analysis

of breakthrough curves from through diffusion experiments at HYRL with specific sample geometries not amenable to analytical solutions.

Figure 43. A measure breakthrough curve together with a fit by analytical solution for a matrix diffusion experiment with a volume flow rate of $2 \mu\text{l}/\text{min}$. The shoulder after the main peak caused by the finite depth of the matrix is clearly visible.



In conclusion, the imaging methods developed can and have already been used for analyzing various problems related to final deposition of spent nuclear fuel. The work started in the second phase of the project on various aspects of matrix diffusion targets in particular proper interpretation of the in situ experiments done or to be done at Grimsel and Olkiluoto. Now that the modeling principles have been validated and new features can be included, new and more useful information can be expected to be found. Several publications, one PhD thesis (2011) and parts of two other theses have been produced.

8.24 Migration and retention of uranium in granitic rock (GeoChem)

Lalli Jokelainen, Marja Siitari-Kauppi, University of Helsinki Laboratory of Radiochemistry

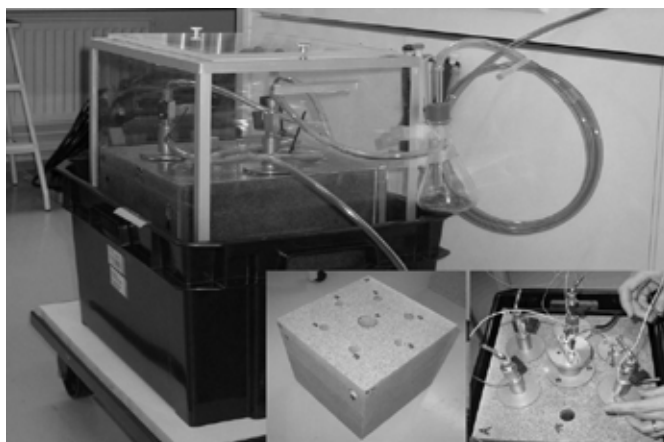
Mira Markovaara-Koivisto, Aalto University

BACKGROUND

Between years 2006 and 2008, the behavior of uranium in granitic rock was studied in Laboratory of Radiochemistry in collaboration with Aalto University School of Science and Technology and Geological Survey of Finland. The project was initiated by Karl-Heinz Hellmuth (Radiation and Safety Authority, STUK) and was a continuation of the previous IAEA project. The project was carried out in co-operation with Professor David Read (Enterpris Ltd + Univ. of Reading). The project was linked to studies concerning the safety case analyses of spent nuclear fuel reposition to the deep crystalline granitic rock repository. In the first part of the study, drill core samples from Palmottu and Askola natural uranium deposits were studied. Uranium minerals in the samples were analyzed with electron microscopy (SEM) coupled with energy dispersive spectrometer (EDS) or wave length dispersive spectrometer (WDS).

In the second part of the project the diffusion of uranium through granite was studied. The diffusion studies were carried out on block-scale using Kuru grey granite rock block (KGG) (Fig. 44) under oxidic conditions. A metallic depleted uranium disc was chosen as the uranium source. The diffusion of dissolved uranium to the rock matrix was observed by taking water samples from the observation holes in the rock block. Uranium concentrations of these water samples were analyzed with ICP-MS. Parallel to uranium diffusion, effective diffusion coefficients for tritium and chloride were determined.

Figure 44. Uranium diffusion experiment is carried out on Kuru Grey granite rock block.



Based on the experimental data from Askola and Palmottu [1,2,3] hydrogeochemical modeling was performed to verify the uranium minerals on the sites [4]. The aim of the modeling studies was to determine the effects of the prevailing conditions to uranium speciation, solubility, oxidation state and precipitation.

RESULTS

In the samples obtained from Palmottu, the richest uranium contents were detected in a boulder sample, in which uranophane was abundant. Secondary uranium phase was uranophane which was precipitated into the fractures of the rock. In Askola uranium occurred often in connection with reducing minerals such as pyrite and chalcopyrite, diffused or enriched into the lamellar structure of spherical goethite nodules (FeO (OH)) and in micro fractures as (hydr) oxide (Fig. 45 & 46)[1-3]. The age of the secondary uranium phases in the rock samples was determined with LA-ICP-MS -method using Agilent 7500 LA-ICP-MS. $^{234}\text{U}/^{238}\text{U}$ activity ratio in the samples examined was over 1, which indicates that the phase is relatively young in the geological point of view. This implies that the secondary phase is precipitated from the groundwater. [5]

Figure 45. Photographs and corresponding C-14 autoradiographs of the Askola samples. In autoradiographs dark areas correspond to conductive porosity and radioactive mineral phases.

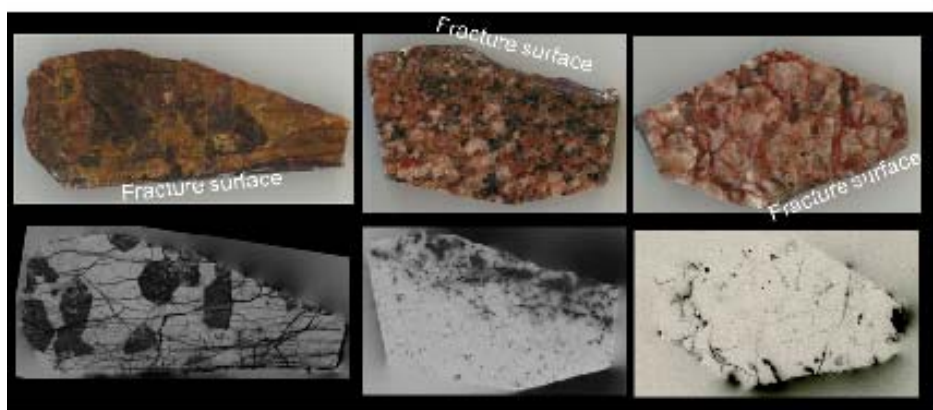
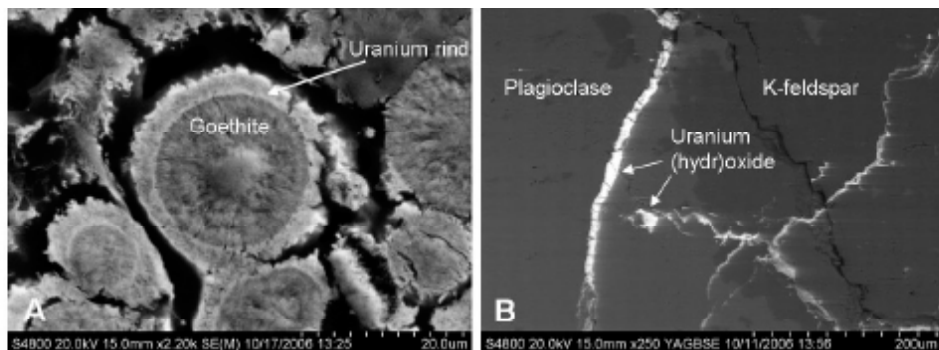


Figure 46. Uranium is enriched (ca. 55 wt.%) at the edge of the nodules shown as a white rim in figure A. Uranium (hydr)oxide filled micro-fractures in figure B.



In the block-scale experiment, the diffusion coefficients for tritium were $8,2 \times 10^{-13} \text{ m}^2/\text{s}$ and for chloride $1,9 \times 10^{-13} \text{ m}^2/\text{s}$ [6]. The diffusion of uranium through granite was slower, due to the uranium's tendency to form carbonate and hydroxide complexes which move as anionic form in the pore water. The effective diffusion coefficient of uranium will be calculated after the through diffusion curve reaches equilibrium. The modeling studies were in agreement with laboratory scale experiments showing uranium precipitation when reducing minerals are present, and soluble hexavalent uranium when reducing elements are absent. In the first case, the main secondary uranium phase found was uranophane.

USABILITY OF THE RESULTS

The results obtained from this study suggest that in oxic conditions uranium is precipitated adjacent to reducing minerals. Secondary uranium phases were found also in the micro fractures of the rock samples. Uranium can be considered as an indicator of a redox front in the water conducting fractures. No sorption of the uranium to the minerals in the samples was detected. As a result of this study the understanding of the geochemical processes governing the mobility and the retention of uranium was increased. The final goal of the study was to combine the geochemical models with the transport models. However, this goal was not achieved due the cancellation of the funding. [7,8]

METHODS AND MATERIALS USED IN THIS STUDY

The autoradiography method was used to measure the porosity of the rock samples. The samples were impregnated with ^{14}C -labelled methylmetacrylate, which was then polymerized by irradiation to the pores of the samples. Sawed and polished subsamples are then placed on the X-ray film. Radioactive decay of the ^{14}C in the rock pores forms an image of the rock matrix to the film. Subsequent image analyses tools were applied to determine the porosities of the rock samples.

Uranium minerals and secondary phases in the samples were analyzed with electron microscopy (SEM) coupled with energy dispersive spectrometer (EDS) or wave length dispersive spectrometer (WDS).

Laser Ablation Inductively Coupled Plasma Mass Spectrometry (LA-ICP-MS) was used to determine the activity ratio of $^{234}\text{U}/^{238}\text{U}$ in the uranium minerals. In LA-ICP-MS laser is utilized in order to vaporize small portion of the solid sample. This vapor is ionized and analyzed using mass spectrometry. Normal Inductively Coupled Plasma Mass Spectrometry was used to measure uranium concentration of the observation holes in the block scale experiment.

PHREEQC, hydrogeochemical modeling software was used to model uranium behavior in rock matrix. This software is useful in speciation, solubility and precipitation calculations. Uranium diffusion in to the rock matrix was also modeled with Comsol Multiphysics software.

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8.25 Sorption of trivalent actinides on clay and (hydr)oxide minerals

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RESULTS

The aim of the project was to understand sorption processes and mechanisms between trivalent metals found in nuclear waste and clay and (hydr)oxide minerals. The primary goal of the work has been to create new atomic scale data for mechanistic modeling of radionuclide transport in geological environments and to introduce new methods with which to study the sorption mechanisms. Since the research methods used are sensitive to impurities, care was taken to synthesize as pure minerals as possible for the experiments. For molecular level understanding of sorption mechanisms simple model minerals such as the pure aluminum hydroxide, gibbsite, and the simple 1:1 layered clay mineral, kaolinite, were chosen for the project. Solid-state NMR measurements proved to be difficult with a mineral like gibbsite that contains protons in the mineral bulk. Therefore, the oxide minerals α -alumina and γ -alumina were used instead of gibbsite for the NMR studies. The project was done in collaboration with the following national and international corporations/institutes:

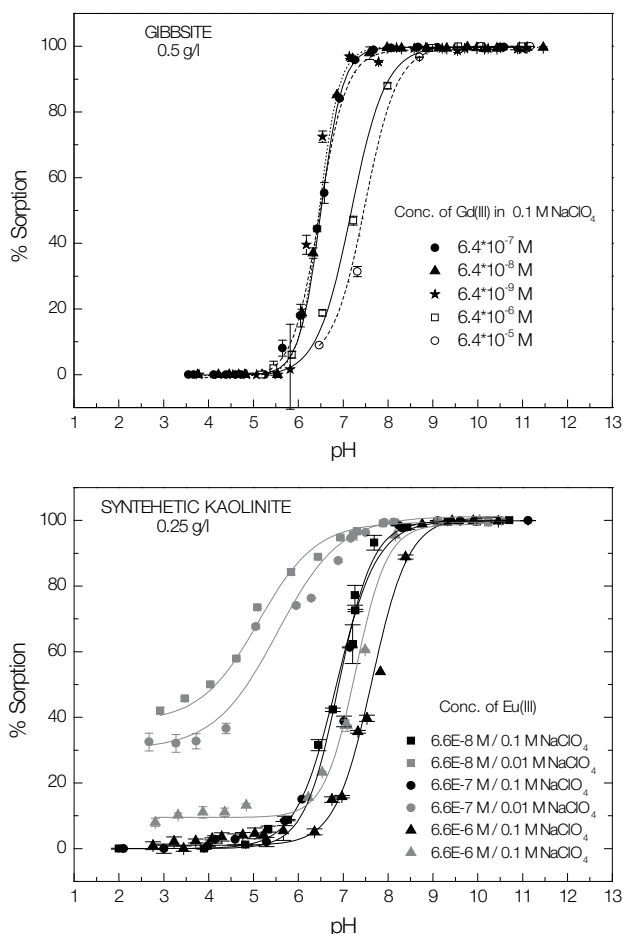
- Infra-Red (IR) studies: Neste Oil Oy, Porvoo, Finland and ASM Helsinki, Finland
- Nuclear Magnetic Resonance (NMR) studies: National Institute of Chemical Physics and Biophysics, Tallinn, Estonia
- Time-Resolved Laser Fluorescence (TRLFS) studies: Institut für Nukleare Entsorgung (INE), Karlsruhe Institute of Technology, Karlsruhe, Germany
- Kaolinite synthesis: Université de Poitiers, Poitiers, France

The most essential results of the project are presented below according to the methods used for the research.

BATCH SORPTION STUDIES

Before the spectroscopic investigations, the macroscopic sorption behaviour of trivalent metals on gibbsite and kaolinite was investigated. The sorption investigations on gibbsite were done with gadolinium, while europium was used in the kaolinite sorption studies. All experiments were performed under argon atmosphere to exclude the influence of carbon dioxide. Figure 47 presents the pH-edges for lanthanide sorption onto gibbsite (above) and kaolinite (below) at different metal-ion concentrations and ionic strengths.

Figure 47. Above- gadolinium sorption onto gibbsite as a function of pH. Below – europium sorption onto synthetic kaolinite as a function of pH in two different ionic strengths.



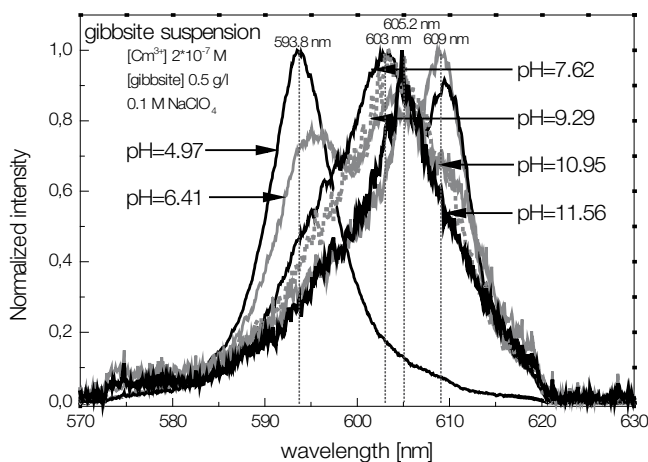
In the ideal sorption range the position of the pH-edge for a given solid concentration is independent of the metal ion concentration used. When the metal-ion concentration increases, sorption sites become saturated and repulsion between the positively charged surface complexes and the metal ions in solution occur. Changes to the non-ideal sorption range can be seen as a shift of the pH edge to higher pH values. This can be witnessed in the gadolinium pH edges when the metal ion concentration exceeds 6.4×10^{-7} M and in the europium pH-edges above 6.6×10^{-7} M metal ion concentrations in 0.1 M NaClO₄. From the kaolinite sorption curves a difference of europium uptake in the two ionic strengths can be seen. This can be attributed to outer-sphere complex formation. The more competing cations in solution, i.e. the higher the ionic strength, the less europium is adsorbed on the surface as outer-sphere complexes in acidic conditions.

TIME-RESOLVED LASER FLUORESCENCE SPECTROSCOPY (TRLFS)

The complexation of trivalent metals on the mineral surfaces was studied with TRLFS in Karlsruhe, Germany. The experiments were done, both for gibbsite and kaolinite, using the well fluorescing actinide, curium. Curium was excited at 396.6 nm with a Nd:YAG pumped dye laser set-up. The recorded emission spectrum is dependent on the energy levels of Cm(III) that further depend on the chemical environment of the metal. When curium complexation occurs, a bathochromic shift of the emission spectrum can be observed and thus, the number of complexes as well as their relative amount can be determined.

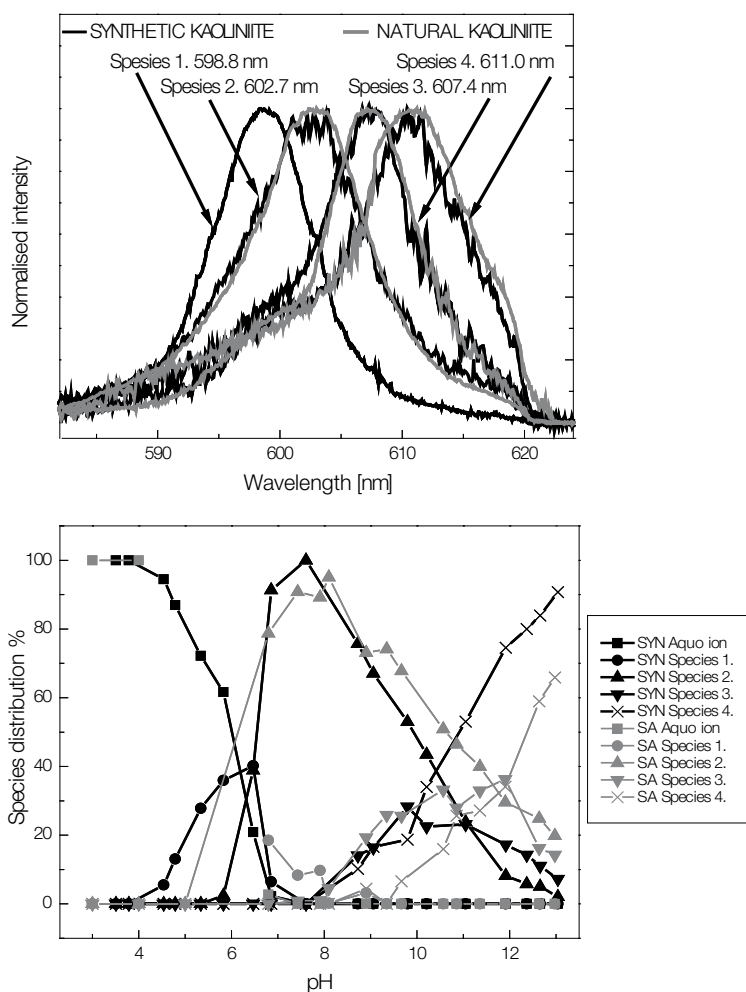
From the batch experiments gadolinium adsorption onto gibbsite was found to begin at pH values above 5. At this pH the emission spectrum of curium changes slightly from that of the pure aquo ion, indicating a change in the curium ligand field as a result of inner-sphere complex formation. The speciation of curium in gibbsite suspensions is highly dependent on suspension pH conditions as well as on the amount of dissolved aluminum. The solubility of aluminum is low in neutral pH conditions and increases rapidly both when going towards more acidic and more alkaline pH. When we move from higher Al solubility domains towards lower ones, we generate oversaturation conditions for dissolved Al species with respect to solid $\text{Al}(\text{OH})_3$ phases that consequently precipitate from solution. This precipitation incorporates surface bound curium. When the solubility again increases, the incorporation dissolves and the curium surface complex is released. Figure 48 presents curium emission spectra at different pH-values. The incorporated curium has a peak maximum at approximately 609 nm. In addition to it, two surface complexes can be distinguished with peak maxima at 603 nm and 605.2 nm.

Figure 48. Curium emission spectra in gibbsite suspensions at various pH-values. Two surface complexes with peak maxima at 603 nm and 605.2 nm can be observed along with incorporated curium at 609 nm.



Natural kaolinite from St. Austell (UK) has previously been used in TRLFS studies with curium. This study was done in the pH range 3.5–8.5. We did a similar TRLFS study on our synthetic kaolinite product in the pH range 4–13 and extended the investigations on the natural kaolinite from pH 7 to 13 for comparison. Three curium surface complexes could be identified in both kaolinite systems along with a fourth Cm(III) species in alkaline pH conditions (>9). This species was found to be a curium-silicate surface complex forming between curium adsorbed on the kaolinite surface and dissolved silicon in solution. The peak positions of all four complexes are at 598.8 nm, 602.7 nm, 607.4 nm and 611 nm (Figure 49, above) and the distribution of species as a function of pH is presented.

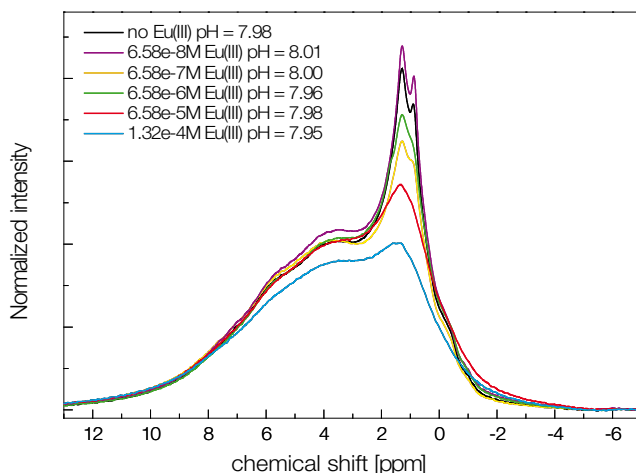
Figure 49. Four curium complexes were identified in the kaolinite suspensions according to recorded emission spectra (left). On the right hand side the distribution of these species as a function of pH is presented. (SYN = synthetic kaolinite, NAT = natural kaolinite, St. Austell)

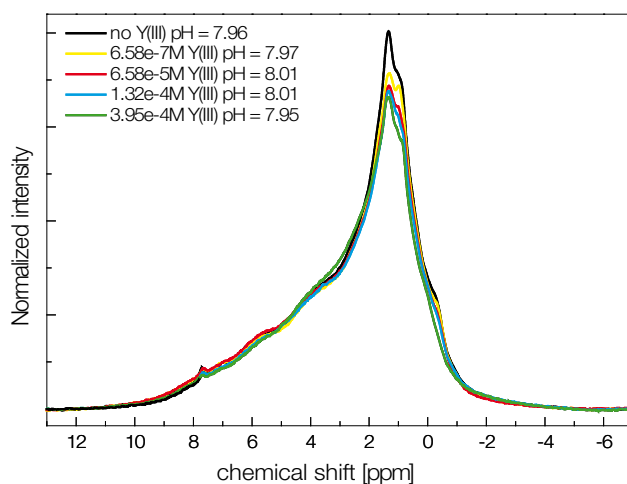


¹H NMR INVESTIGATIONS

The specific sorption of trivalent metals Eu(III) and Y(III) onto the aluminum oxides, α -alumina and γ -alumina, was investigated with solid-state ^1H and ^{27}Al NMR. The aim of the NMR investigations was to determine the specific sorption and coordination of the chosen metal ions on the hydroxyl groups present on the mineral surfaces. The protonation and dissociation reactions in solution were found to influence the recorded proton spectrum of γ -alumina. Therefore, metal-ion containing NMR samples were prepared in identical pH conditions (8.00 ± 0.05). Figure 50 presents the proton spectra of Eu(III) containing (above) and Y(III) containing (below) γ -alumina samples at different metal-ion concentrations. The europium attachment on the mineral surface influences the proton spectra to a greater extent than yttrium does. This can be attributed to the paramagnetism of the trivalent europium cation. Yttrium only influences the two most intense proton peaks with approximately 1–2 ppm chemical shifts. We recorded these two peaks also in the α -alumina spectrum and in a fumed silica sample. Therefore these peaks were attributed to protons stemming from physisorbed water still attached on the mineral surfaces. The more yttrium that is adsorbed on the γ -alumina surface, the less water can attach through formation of hydrogen bonds on the mineral.

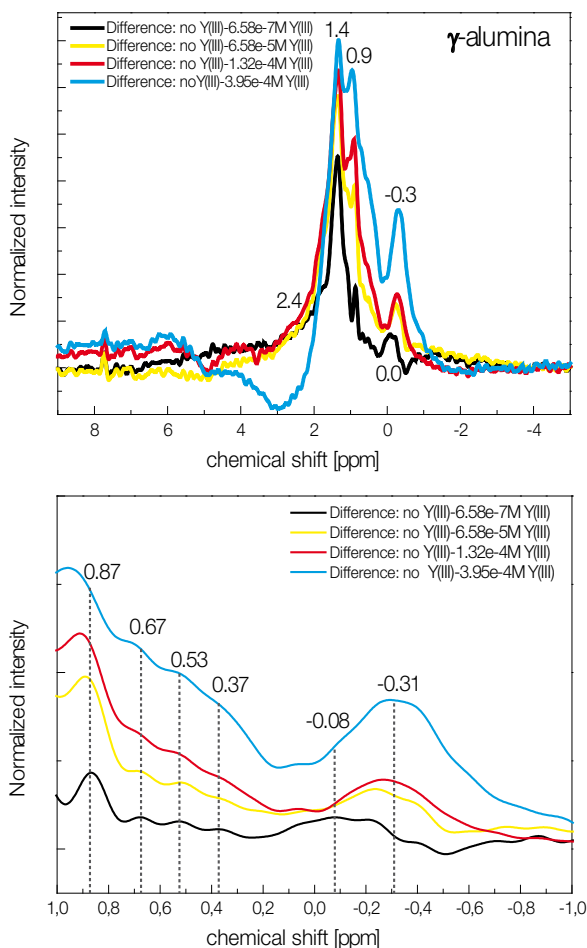
Figure 50. Proton spectra of γ -alumina with various amounts of Eu(III) (above) and Y(III) (below). Europium addition influences the intensities of recorded spectra to a greater extent than yttrium. This is attributed to the paramagnetism of the trivalent europium cation.





In order to see the spectral changes induced by the sorption reaction we must regard the differences relative to an unreacted γ -alumina sample. Therefore, to extract this information difference spectra were produced by subtracting the proton spectra of the samples containing yttrium from that of the unreacted γ -alumina mineral at 8.00 ± 0.05 . The difference spectra are presented in Figure 51. In the difference spectra clear features at 1.4, 0.9, -0.1 and -0.3 ppm can be distinguished along with a negative broad feature at approximately 3 ppm. Due to the fact that this negative peak only occurs in the spectrum for the highest yttrium concentration of 3.95×10^{-4} M, we assign the peak to protons from $Y(OH)_3$, precipitating from oversaturated solutions. Physisorbed water with the previously assigned peak positions at 1.3 and 0.9 ppm also appear to be influenced by the addition of the trivalent metal. This could be attributed to the decreasing amount of available surface sites for hydrogen bond formation between water molecules and surface hydroxyl groups upon yttrium adsorption as explained above. Acidic protons with chemical shifts above 2 ppm are not present in the difference spectra and thus, they do not participate in the sorption reaction of yttrium. On the other hand a great amount of basic protons with chemical shifts below 0.9 ppm are influenced by yttrium attachment to the mineral surface. This can be seen especially in the magnification of the difference spectra, figure 51. Such a broad range of exchanged protons with very similar chemical shifts can be explained by e.g. different O-H bond lengths of otherwise similar surface groups or by the coordination number of aluminum. The electron density at the proton is influenced by the coordination of the aluminum atom that the oxygen or oxygens are bonded to. A higher coordination of the Al^{3+} cation implies that the oxygens attached to it have higher relative negative charge as the positive charge of the aluminum is distributed among a higher number of oxygen atoms. This higher negative charge influences the electron density at the hydroxyl proton that we consequently witness at a lower chemical shift.

Figure 51. Difference spectra of yttrium containing γ -alumina samples (above). Magnification of the 1-(-1) ppm region of the difference spectra (below).



SUMMARY

In this project new molecular level insight on trivalent actinide sorption on minerals in the bedrock has been obtained. We have also introduced methods that have not been used before in Finnish repository research. From the TRLFS results important information on trivalent actinide speciation in gibbsite and kaolinite suspensions was obtained. In both systems the metal ions dissolved from the mineral itself played a large role on the curium speciation. Especially incorporation is a mechanism that retains curium in a different manner than the formation of a surface complex. While the surface complex might desorb from the mineral surface when pH conditions change, incorporated curium is mobilized only when the mineral re-dissolves.

Such mechanisms cannot be distinguished in simple batch experiments and they have to be considered in future surface complexation models as well as in radionuclide migration models. The NMR investigations are first of their kind even on an international scale, as specific trivalent metal ion sorption investigations onto aluminum oxides has not been attempted with the method before. The sorption of yttrium was found to occur on a large number of different hydroxyl groups. Also bridging OH-groups (e.g. $(\text{Al}_{\text{VI}})_2\text{-OH}$) that have not been considered to participate in sorption reactions in many studies, were shown to participate in the yttrium adsorption on the γ -alumina surface. This has to be considered in sorption and migration models, even though it does not influence the amount of sorption directly. Two articles have been published from the attained results and the manuscript of a third publication is submitted. Furthermore, results have been presented in a number of radiochemistry conferences, both orally and with poster presentations.

8.26 Disposal of spent nuclear fuel: ecological risk assessment in forest ecosystem

Jukka Juutilainen, Päivi Roivainen, Tiina Boman, Sari Makkonen, Toini Holopainen, Mikko Kolehmainen, Jonne Naarala, University of Eastern Finland/Kuopio campus

BACKGROUND

The soil-to-plant transfer of radionuclides is a key process when the exposure of organisms is assessed. In radioecological modelling this transfer is usually described by a concentration ratio (CR) between soil and plant concentrations. The CR values of most radionuclides present in radioactive waste are not well studied, especially in forest ecosystem. The lack of data is even more evident in Finnish environmental conditions, which can be remarkably different from, for example, those in Southern Europe. The aim of this project (carried out during the years 2006–2010) was to provide data for radioecological modelling adapted to the needs of spent nuclear fuel disposal and Finnish environmental conditions.

The main goal of this project was to investigate the soil-to-plant transfer of some important elements (Co, Mo, Ni, Pb, U) affecting the safety of spent nuclear fuel disposal. Soil-to-plant CR values were calculated and CR values of different plant parts were compared. The effects of soil properties and other elements on CR values of the elements studied were also investigated. The validity of the linearity assumption (the assumption of radioecological models that the relationship between soil and plant concentrations is linear) was also studied. The transfer of elements from soil to earthworms and carabid beetles was also investigated to a limited degree.

METHODS

The research was based on data collected from two uranium occurrences in Northern Savo in summer 2007. Samples of soil ($n=29$), litter ($n=29$), rowan ($n=28$), Narrow buckler fern ($n=27$), Norway spruce ($n=26$) and May lily ($n=19$) were collected from the Murtolahti sampling site in Nilsä. In addition to soil samples ($n=23$) Blueberry ($n=23$) and Norway spruce ($n=16$) were collected from Puutosmäki sampling site in Kuopio. Each of the understory samples was further divided into sections of leaves, stems and roots. The tree samples were divided into leaves/needles, fine roots (diameter < 2 mm) and coarse roots (diameter > 2 mm).

Attempts to collect earthworms and carabid beetles were made at both sampling sites in 2007 but no animals were caught at the Puutosmäki site. In 2008, sampling was done only in Murtolahti. Animals from adjacent sampling points were pooled in order to get large enough samples for the analyses. As there are different species of both earthworms and carabid beetles, the pooled samples consisted of individuals of the same species whenever possible. In total, 24 samples of carabid beetles were analysed in 2007 and 5 samples in 2008. There were 4 earthworm samples in 2007 and 7 samples in 2008. Data of both years were pooled when CR values were calculated.

The analyses were done under the assumption that all the isotopes of an element behave similarly in the environment. Instead of activity concentrations of specific radionuclides, total concentrations of elements were analysed by the ICP (inductively coupled plasma) method, which gave the concentrations of 34 elements in one analysis. In addition, the mobile fraction of elements (after ammonium acetate leach), pH, organic matter content and particle size distribution were analysed from soil samples.

The CR values were calculated for Co, Mo, Ni, Pb and U as follows: $CR = C_{\text{organism}} / C_{\text{soil}}$, where C_{organism} is the concentration of an element in an organism (mg kg^{-1} DW) and C_{soil} is the concentration of an element in soil (mg kg^{-1} DW). Regression analysis was used to investigate the effects of soil properties and other elements on CR values.

The CR values calculated as described above include the assumption that the concentrations in the tissues of an organism and in soil are linearly related. The validity of this assumption was tested by comparing the fits of the plant data with a linear model (constant CR values) and a non-linear relationship ($CR = C_{\text{plant}} / C_{\text{soil}} = a / (1 + b C_{\text{soil}}) + c$) based on the Langmuir equation.

THE MAIN RESULTS

Soil-to-root CR values were higher than soil-to-stem and soil-to-leaf values for all the elements and species studied. Thus it can be recommended that the accumulation of elements to the plant roots is taken into account. In general, the variation of CR values was high. The variation between species was not significantly higher than within species. This indicates that same CR values can be used for different plant species, which simplifies modelling. Table 8 summarises the CR values from soil to understory leaves and roots and to tree leaves/needles and fine roots. Soil P concentration was the most important nutrient affecting the CR values of the elements studied.

The CR values from soil to earthworms were higher than to carabid beetles (Table 8). The CR values for earthworms were of similar order of magnitude as those for plant roots, while CR values for carabid beetles were similar to those for plant leaves.

Table 8. Geometric means and geometric standard deviations of the concentration ratios for cobalt, lead, molybdenum, nickel and uranium from soil to understory (n=69), trees (n=70), earthworms (n=11) and carabid beetles (n=29).

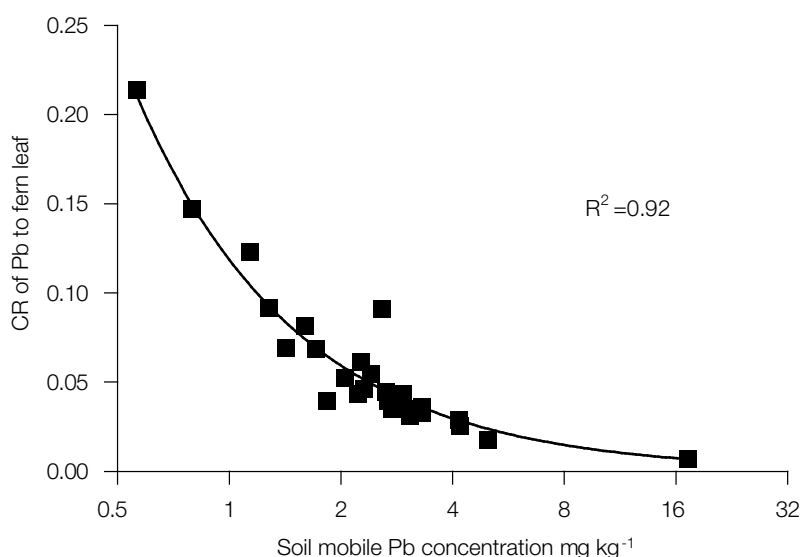
Organism	Co	Mo	Ni	Pb	U ¹
leaf, understory	0.04 (2.48)	0.17 (3.66)	0.30 (1.68)	0.01 (2.43)	0.005 (3.18)
root, understory	0.19 (3.04)	0.33 (3.23)	0.63 (1.82)	0.14 (2.18)	0.06 (3.49)
leaf/needle, trees	0.05 (2.11)	0.07 (4.49)	0.28 (2.44)	0.01 (2.09)	0.004 (3.77)
fine root, trees	0.64 (2.94)	0.43 (2.88)	0.77 (1.87)	0.54 (2.35)	0.27 (4.27)
earthworm	0.85 (1.83)	0.49 (2.59)	0.26 (2.11)	0.24 (3.74)	0.52 (4.59)
carabid beetle	0.03 (1.65)	0.69 (3.55)	0.05 (1.49)	0.008(1.66) ²	0.005 (1.93)

¹ The results are based only on data from Murtolahti since the plant concentrations at Puutosmäki were below detection limit (0.01) and no animals were caught there.

² Calculated based on the results of the year 2008 since all the concentrations in carabid beetles were below the detection limit (0.05) in the year 2007.

The linearity assumption was studied using the plant data. It was found for all of the elements and species studied that the CR values were lower at higher soil concentrations (Figure 52). The non-linear model described the relationship between CR values and soil concentrations significantly better than the linear fit. This was consistent both for the CR values based on soil total concentration and those based on soil mobile concentration. A part of the large variation of empirical CR values may be systematic variation with soil concentration. The use of non-linear methods could improve the accuracy of modelling the soil-to-plant transfer of elements.

Fig 52. Concentration ratio (CR) of Pb for fern leaf as a function of soil mobile concentration. The line represents the fitted non-linear equation ($Y = a / (1 + b C_s) + c$) and R^2 the goodness of fit.



THE SIGNIFICANCE AND EXPLOITATION OF THE RESULTS

Biosphere modelling is an important part of the risk assessment of spent nuclear fuel disposal. The empirical data produced in this project can be utilised in validating radioecological models and in evaluating the basic assumptions used in those models. The suitability of those models to Finnish conditions can thus be improved. The results of this study can also be used for assessing environmental effects of uranium prospecting and possible uranium mines.

8.27 Fate of radionuclide ^{14}C in soil-plant-atmosphere continuum

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INTRODUCTION

Radiocarbon (^{14}C) has been identified as being potentially significant in terms of releases from deep geological disposal of radioactive waste. It is particularly important with respect to low and intermediate-level waste material and spent fuel. However, detailed knowledge about the long-term environmental behavior and migration of ^{14}C in biosphere/atmosphere is still lacking.

OBJECTIVES OF THE STUDY

The overall objectives of the study have been to determine the magnitude of C-14 losses from the soil to the atmosphere, to quantify uptake of C-14 from soil sources into plants, and to study uptake of C-14 from soil into the soil microbes and fauna. More specifically, the intentions of this study were to determine 1) The impact of climatic conditions on C-14 emissions; 2) The impact of different plant types (grass/trees) on the release of C-14 from soil (so-called priming effect); 3) C-14 uptake via roots; 4) Foliar uptake of C-14 released from the soil; 5) Translocation of C-14 from roots to above-ground plant components; 6) Differences between plant types (grass/trees) in the ability to absorb C-14 from soil; 7) Environmental controls on plant uptake of C-14 from the soil; and 8) Incorporation of C-14 into soil microbial and faunal biomass.

MATERIALS AND METHODS

Experimental Approach: In order to trace the fate of C-14 from soil to plants and atmosphere, the whole soil has to carry a distinct isotopic signature. Therefore, we took advantage of large differences in age (^{14}C content) between soil and plants at a re-cultivated cutaway peatland (Figure 53). This innovation has never been used before in soil carbon and ^{14}C studies. The older, left-over peat material is more depleted in C-14 content than 'younger' plant material. Further, the C-14 signature in these older soils is reasonably spatially homogeneous. These are ideal prerequisites for source partitioning with isotopes.

Laboratory studies: Two plants (a perennial crop (reed canary grass; RCG) and pine seedlings) have been studied in the laboratory. These plants were chosen as being representative of typical plants grown on such cut-away peats. Plant material was studied during the growing season to determine the plant carbon balance, looking at the effects of different temperature treatments. CO_2 exchange measurements were also made. Samples were collected for C-14 analyses, from a range of plant tissues and CO_2 . The data stemming from the laboratory studies were content of a master thesis (Promise 2010).

Field studies: In 2009 and 2010, experiments were carried out both in the laboratory and in the field; the location of the field site was Linnansuo in Eastern Finland (soil from this site was also used in the laboratory experiments). Complementary to the laboratory studies, we chose sites with RCG cultivation and pine cultivations which are both available in Linnansuo field site. In the field study, an eddy covariance tower enables the continuous measurement of the net ecosystem exchange (Shurpali et al. 2009). In the field, a range of hydrological conditions (drained, wet, and water-logged) were maintained over the growing season; these are consistent with natural conditions – water-logged – to typical practices – drainage of peatlands – in Finland. Plant material from a range of tissues (leaves, roots, stems), soil material, CO₂ (respired from soil and accumulated within the canopy), soil microorganisms, dissolved organic carbon and soil fauna was collected, quantified and analyzed for ¹⁴C.

Analysis and Calculations: The low C-14 activity in the cut-away peat means that analyses have to be carried out using the cost-intensive accelerator mass spectrometry (AMS) technique. Samples were sent over from the Radiocarbon Dating Laboratory in Helsinki to Sweden for analysis, and also directly sent to the Poznan Radiocarbon Dating Laboratory in Poland. The two-pool isotope mixing model was applied to reveal the fraction of soil-derived carbon in ecosystem components and fluxes.

RESULTS AND DISCUSSION

Respiration data indicated that ¹⁴CO₂ from the soil, stemming from organic matter decomposition of old peat, comprised a relatively smaller proportion compared to plant-derived CO₂ in the overall measured ecosystem respiration. Whilst an increase in soil moisture content was found to lead to a decrease in net ecosystem exchange, it did not affect the C-14 content of CO₂ being respired; soil ¹⁴CO₂- and plant-derived CO₂ were thus similarly affected. Effects of temperature on the relative proportion of ¹⁴C release were of minor importance, even though the magnitude of the fluxes increased with temperature.

Microbial biomass was heavily enriched in ¹⁴C (on average by 63%), leading to the ¹⁴CO₂ emissions mentioned above (microbial respiration). Along with microbial biomass, soil carbon amounted to about one third of total dissolved organic carbon, carrying thus risks for ¹⁴C leaching and groundwater contamination. Soil fauna had generally negligible amount of ¹⁴C inside its biomass, with the exception of earthworms (23%) and mosquitos (2.4%); Earthworms feed partially on soil carbon, while the larva of mosquitos may have access to dissolved organic material leaching into the drainage ditches. Most of the mesofauna, however, prefers fresh, plant-derived carbon over old soil carbon.

In most cases, no or only traces of soil-derived carbon were found in the plant tissues of both RCG and pine (Figure 54). The high ¹⁴C value in the first root sample (22%) was thus not confirmed by our study, and is currently considered as an outlier; however, this issue is presently under investigation. The percentage of soil carbon in roots was higher than in leaves, and was on average between 0.9 and 2% in RCG

and pine, respectively. These values are well within the range of reported literature values, indicating that root uptake of carbon may contribute up to only a few percent of plant carbon. The relatively higher value found in pine roots could be due to the high mycorrhizal infection levels of pine plants (along with organic nitrogen also organic carbon in the amino-acids are taken up by the roots). The lack of C-14 signal of the leaves indicated that essentially all the carbon in leaves derives from photosynthetic uptake from the atmosphere (Figure 54). ^{14}C content in leaves were in both plant species with 0.4% and 0.5 % not significantly different from zero. This was surprising, as up to 15% of ^{14}C was measured inside of the canopy air. Perhaps this ^{14}C was not “seen” by the plant leaves as the highest level of photosynthesis occurs towards the top of the plant canopy, a region with the highest degree of mixing with free air, but more studies are needed to verify this. Current models predict up to 30% of re-assimilation of soil-derived C into plant biomass, and there are different concepts and assumptions with regard to plant canopy atmosphere in the models (e.g. Rimers, SKB-Posiva; Bioprota 2010). The measured C-14 concentrations in the respired CO_2 were, however, similar to those calculated by the models (Bioprota 2010).

In conclusion, our data have proven to be valuable for increasing the understanding of environmental behavior and movement of the radionuclide ^{14}C from soils. Models may be calibrated with data stemming from case studies as ours, and great interest to do so was shown in the international Bioprota meeting where the project leader presented the Finnish research (Bioprota 2010). Our first results do not confirm the high incorporation of soil-derived carbon into plants as used by many models, but more studies are needed to generalize this and to understand the underlying mechanisms.

Figure 53. Principle of the ^{14}C tracer used in this study; as a result of peat harvesting a natural difference in “label” (^{14}C isotopic singature or age) exists between plants and soil in cultivated cut-away peatlands.

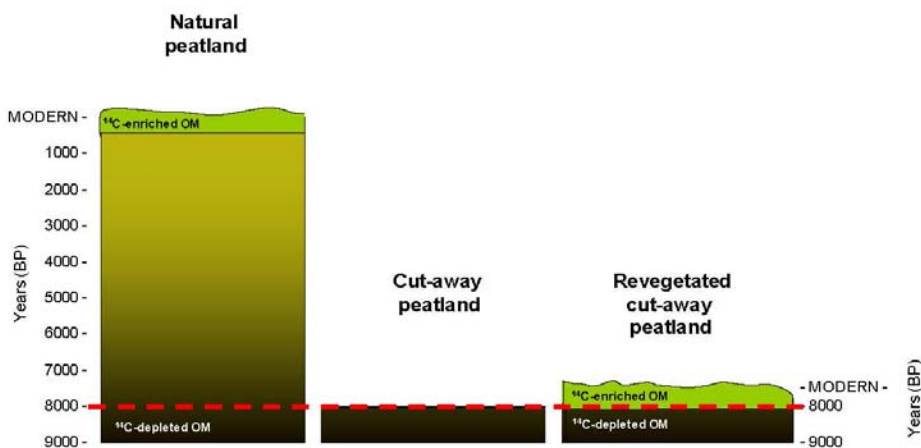
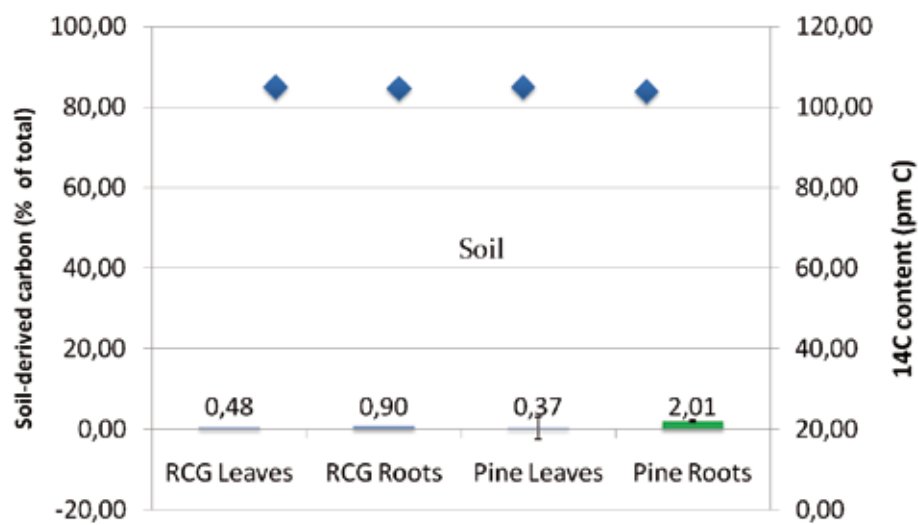


Figure 54. Average ^{14}C content in plant tissues of reed canary grass (RCG) and pine (diamonds) as percentage of soil-derived carbon (numbers on top of bars). The ^{14}C content of the two contributing sources (atmosphere and soil) are presented as lines.



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8.28 Joint projects of University of Jyväskylä and University of Tampere, 2008-2010

Tapio Litmanen, University of Jyväskylä

The SEURA research project was launched in 2008 as a joint initiative of the University of Jyväskylä (Department of Social Sciences and Philosophy) and the University of Tampere (Department of Political Science and International Relations). The SEURA project's full title, "Seurantahanke käytetyn ydinpolttoaineen loppusijoituslaitoksen sosioekonomisista vaikutuksista ja tiedonvälityksestä Eurajoen ja sen naapurikuntien asukkaiden näkökulmasta", translates into English as "Follow-up research regarding the socio-economic effects and communication of final disposal facility of spent nuclear fuel in Eurajoki and its neighbouring municipalities". The project was funded by the Finnish Research Programme on Nuclear Waste Management (KYT2010, www.ydinjatutkimus.fi) in 2008-2009. A second funded project, TEKY ("Teollisuustietoisuus ja käytetyn ydinpolttoaineen loppusijoituksen hyväksyttävyyys"), carried out in 2010, translates into English as "Industry awareness and acceptance of final disposal of spent nuclear fuel".

The main objectives of the SEURA research projects were to study residents' opinions in the municipality of Eurajoki and its neighbouring municipalities regarding 1) socio-economic and socio-political impacts of the final disposal facility and 2) information needs and ways of obtaining information regarding the final disposal plan, and 3) to determine nuclear power's status and cultural acceptability in a nuclear community. In addition to the two key projects it was considered important to further map current issues in the field of nuclear power and waste policy by conducting three minor public hearings observation projects. The final project report, "Community Divided: Adaptation and Aversion towards the Spent Nuclear Fuel Repository in Eurajoki and its Neighbouring Municipalities" (Kari et al. 2010), documents the key research findings. The report is based on a resident survey conducted in June 2008 (sample size 3000, response rate 20%, N=606), with the target population being native Finnish speakers aged between 16 and 75, residing in Eurajoki and neighbouring municipalities. In addition to the report, further results of the survey are reported in three international scientific journals (Kojo et al. 2010; Litmanen et al. 2010; Kojo et al. 2011) and in several international conferences (Kojo et al. 2008; 2009a; 2009b ; Kari 2009, 2010a, 2010b; Kojo & Kari 2010). Findings of other minor projects are reported in three working reports (Nurmi et al. 2009; Nurmi 2010; Pylkkönen et al. 2008).

KEY RESEARCH FINDINGS

"Community Divided" (Kari et al. 2010) introduces the results of a survey conducted in Eurajoki, the first municipality in the world to approve of the final disposal of spent nuclear fuel (SNF) within its own boundaries, and its neighbouring municipalities

regarding issues connected to the SNF repository project. The report draws upon two conceptual approaches in interpreting the rationality of a nuclear community. The 'nuclear oasis' approach suggests that local acceptance is based on the heavy dependency of a small, peripheral municipality on a powerful nuclear industry. The challenging 'industry awareness' approach interprets the community's readiness to accept the siting of a SNF disposal repository from the perspective of cultural adaptation. In this scenario, a community and its residents have close relations to the nuclear industry, which produces cultural adaptation, integration and acceptance of nuclear activities.

The findings indicate that those residents of Eurajoki who perceived the repository to have positive impacts on the general socio-cultural development of the municipality were more willing to accept an SNF repository in Olkiluoto. The importance of economic and employment factors behind the acceptance were identified, but the value of these factors was weaker than more general socio-cultural satisfaction indicators. Such findings suggest the presence of the scenario outlined by the industry awareness approach. However, the actual case was found to be more complicated, as the residents' cultural adaptation to the nuclear industry was neither harmoniously occurring nor homogenously experienced.

There is a latent social cleavage in the area studied. This means that there is a hidden division or dividing line of members into two factions or groups, among which there is a potential for conflict. For instance, there is a discrepancy between women's and men's views on most issues outlined in the survey. Furthermore, from a political perspective, the findings suggest that residents in favour of the final disposal plan are most likely to be found among the supporters of the Coalition Party, the Centre Party and in some cases also the Social Democratic Party. Residents with a negative attitude towards the final disposal plan are more likely to be found among the supporters of the Green League and the Christian Democrats. The analysis of the data also indicates that the attitudes of those with higher incomes, levels of education, and occupational status are considerably more positive towards the final disposal than of those with lower incomes, educational levels, and occupational status. In some cases the differences are quite remarkable. Respondents with higher levels of income were notably more likely to deny, tolerate, or hesitate to identify the risks of nuclear waste disposal.

Both the Finnish Radiation and Safety Authority (STUK) and the nuclear industry have succeeded in establishing a fairly trusted position as an information provider in the localities, but the study identified that the same social division can be seen among receivers of the information. This means that there are significant numbers of local residents who do not trust these actors as reliable sources of information.

THREE ADDITIONAL PROJECTS TO THE MAIN PROJECT

In addition to the main project, three smaller supporting research projects were conducted. The first focused on the public meetings organized by the Ministry of

Employment and the Economy and Posiva Ltd. concerning of Posiva's proposed spent nuclear fuel repository expansion in Eurajoki, Finland. Four public meetings, held in 2008 and 2009 in Eurajoki, were analysed. Two of them were in accordance with the Act on Environment Impact Assessment Procedure (EIA) and two in accordance with the Nuclear Energy Act and its Decision-in Principle Procedure. The study showed that issues surrounding project information and project decision-making aroused the majority of interest amongst local residents. Regarding 'project decision-making', issues concerning decision-making at the municipal level, nuclear issues and energy policy were broadly discussed in the meetings. Issues raised in relation to 'project information' included the methods of implementation of the project, the reliability of information provided, and industry responsibility. Concern was voiced regarding public health and radiation issues, both for residents' personal safety and broader environmental impact. Perceived threats posed by potential radiation were addressed in the meetings; however, it was noted that the meetings did not address all security related concerns. Nuclear safety issues seem to fall between two different laws in which the EIA and decision-in principle public meetings are described.

Two further projects analysed the public meetings of the Environmental Impact Assessment (EIA) and Decision-in-Principle (DiP) processes regarding the Fennovoima Ltd Nuclear Power Plant (NPP) unit. As questions remain surrounding the nuclear waste management of Fennovoima Ltd, it was considered important to produce updated information on the nuclear power and waste debate for potential NPP siting communities.

SIGNIFICANCE AND APPLICABILITY OF RESEARCH RESULTS

The findings of the SEURA-project can be utilised in several respects. Firstly, the findings hold key relevance for authorities, decision-makers, industry and citizens as they offer an up-to-date picture of Eurajoki and neighbouring municipalities' residents' views regarding the socio-economic and socio-political impacts of the final disposal facility. Secondly, the research provides an opportunity to critically analyse the results of other social science research produced by the industry. It may also suggest some new research topics for further studies. Thirdly, the research produced some completely new findings, i.e., the use of the internet as a way of obtaining information regarding the final disposal plan. Fourthly, research findings indicated some residents perceive an explicit threat posed by the disposal facility and require further information. Fifthly, social science research on nuclear waste management is vital from the perspective of historical documentation and ensuring transparent decision-making. Finally, the texts published in English have created the possibility for international audiences to access the findings and follow the societal aspects of Finnish nuclear waste management.

Further analysis and application of the research findings is dependent on the key actors maintaining a central role in Finnish nuclear waste management policy.

The development of the Eurajoki municipality as an important concentration of the nuclear industry is also a regional issue; as such, regional and local authorities, decision-makers and residents alike can utilise the research findings. The new phase of Finnish nuclear power policy – particularly bearing in mind the public debate surrounding Fennovoima's spent nuclear fuel management – increases the relevance of the findings for the nuclear industry and the public both at the local and national level. Furthermore, the attention shown by the international media regarding the research findings reveals a broader interest in Finnish nuclear waste policy. The international significance of the findings can be cited in the publication of the research project articles in key international science journals.

Research findings contribute to the development of an overall understanding of the relationship between the nuclear industry and local communities. The identified social cleavage at the local level is an important issue, as the implementation of nuclear waste management requires residents' broad acceptance. Distinct attitudinal differences regarding gender and other demographic factors indicate social challenges requiring the attention of Finnish nuclear waste policy actors.

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Annex 1

List of research projects

	Time	Project	Organisation
1	2006–2010	Finnish research project on partitioning and transmutation	VTT
2	2006–2007	Scientific and technical basis of nuclear waste management (TEPE)	VTT
3	2007	Review on environmental remediation techniques of uranium mining and milling tailings	VTT
4	2010	Coordination of the nuclear waste management university education and further training	TKK
5	2006	DECOVALEX THMC project in 2006	STUK
6	2006	Activation of biological shield of NPP (BETAKT)	VTT
7	2006–2010	Long term integrity of copper overpack	VTT
8	2009–2010	Long-term integrity of copper overpack	VTT
9	2007–2010	Deformation Mechanisms of Nuclear Waste Canisters	TKK
10	2007–2008	Measurement of the corrosion rate of copper in gas phase at first stage of the final disposal	TKK
11	2006	Stress corrosion analysis of copper	VTT
12	2009–2010	Sulphide induced stress corrosion cracking of copper	VTT
13	2010	Corrosion of copper by water under oxygen free conditions	TKK
14	2007–2008	Bentomap: Survey of bentonite and tunnel backfill knowledge	VTT
15	2006	Reactive transport modelling in the disposal conditions (REKU)	VTT
16	2007–2008	Modelling in near field (LÄHITHC)	VTT
17	2008	Modelling of glacial erosion in bentonite buffer (GLASE)	VTT
18	2009–2010	Coupled behaviour of bentonite buffer (PUSKURI)	VTT
19	2009–2010	THM modelling of bentonite buffer (PUSKURI)	Numerola
20	2007–2008	THM modelling of bentonite buffer (PUSKURI)	JY/FL
21	2009–2010	THM modelling of bentonite buffer (PUSKURI)	JY/FL
22	2010	Dissolution of montmorillonite in groundwater simulants (MOPO)	VTT
23	2008–2010	Modelling the durability of concrete for nuclear waste disposal facilities	TKK
24	2006–2007	Time-dependent fracturing of rock mass and modelling of EDZ of a repository for spent nuclear fuel (CREEP)	TKK
25	2006–2009	Acoustic emission based rock stress measurement method	TKK
26	2009–2010	Rockmass classification, determination and visualization of rock quality and associated uncertainties using statistical interference and geological 3D and GIS software	TKK
27	2008–2010	Gases and biogeochemical processes in the crystalline bedrock (KABIO)	GTK
28	2006–2010	Deep subsurface geomicrobial processes - Molecular monitoring methods (GEOMOL)	VTT
29	2006–2008	Redox and water-rock-interaction (KIVES-REDOX)	HYRL
30	2006	Fractured bedrock and hydrogeology	GTK

31	2006	Modelling of sorption and reactive migration in tunnel filling materials and fractures in bedrock	HYRL
32	2006–2008	Grimsel Test Site	HYRL
33	2009–2010	Grimsel Test Site – Phase VI and Long Term Diffusion –project (LTD)	HYRL
34	2006–2007	Fracture flow, radionuclide transport and chemical interaction in bedrock	HYRL
35	2008–2010	Colloid formation and their relevance in promoting the transport of radionuclides (KOLKU)	HYRL
36	2006–2008	Imaging of rock porosity by tomographic methods	JY/FL
37	2009–2010	Imaging of rock porosity by tomographic methods and combining it with matrix diffusion modelling	JY/FL
38	2006–2008	Migration and retention of uranium in granitic rock (GEOCHEM)	TKK
39	2006	Migration and retention of uranium in granitic rock (GEOCHEM)	HYRL
40	2008–2010	Sorption of trivalent actinides on clay and (hydr)oxide minerals	HYRL
41	2006–2010	Disposal of spent nuclear fuel: ecological risk assessment in forest ecosystem	KY
42	2009–2010	Fate of radionuclide ¹⁴ C in soil-plant-atmosphere continuum	KY
43	2008–2009	Follow-up research regarding the socio-economic effects and communication of final disposal facility of spent nuclear fuel in Eurajoki and its neighbouring municipalities (SEURA)	JY/YTK
44	2010	Industry awareness and acceptance of final disposal of spent nuclear fuel (TEKY)	JY/YTK

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KYT2010 Kansallinen ydinjätehuollon tutkimusohjelma 2006–2010 – Loppuraportti			
Tiivistelmä Referat Abstract			
<p>Kansallinen ydinjätehuollon tutkimusohjelma KYT2010 käynnistyi vuonna 2006. Tutkimuskausi päättyi vuoden 2010 lopussa.</p> <p>Tutkimusohjelman keskeiset aihepiirit ovat 1) ydinjätehuollon strategiset selvitykset, 2) ydinjätteiden loppusijoituksen turvallisuuden arviointi ja 3) yhteiskuntatieteelliset selvitykset. Ydinjätteiden loppusijoituksen turvallisuuden arvioinnissa on kolme osa-aluetta, jotka ovat tekniset vapautumisesteet, kallioopera ja pohjavesi sekä radionuklidien vapautuminen ja kulkeutuminen.</p> <p>Tutkimuskaudella on ollut käynnissä noin 40 tutkimushanketta, jotka ovat ensisijaisesti liittyneet ydinjätehuollon turvallisuuden arviointiin. Valtion ydinjätehuoltorahasto ohjasi rahaa tutkimushankkeisiin yhteensä noin 7 miljoonaa euroa.</p> <p>KYT2010-tutkimusohjelmassa ovat olleet mukana seuraavat tutkimusorganisaatiot: VTT, Aalto-yliopiston teknillinen korkeakoulu, Helsingin yliopiston Radiokemian laboratorio, Geologian tutkimuskeskus, Jyväskylän yliopisto, Itä-Suomen yliopiston Kuopion kampus, Numerola Oy.</p> <p>Tutkimusohjelman työskentely on perustunut tutkimusohjelman johtoryhmän, kahden tukiryhmän, koordinaattorin ja tutkimushankkeiden keskinäiseen yhteistyöhön ja työnjakoon. Tutkimusohjelman johtoryhmässä ovat olleet edustettuina Säteilyturvakeskus, Teknologian tutkimuskeskus Tekes, Fortum Power and Heat Oy, Teollisuuden Voima Oyj, Posiva Oy ja työ- ja elinkeinoministeriö.</p>			
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Julkaisun nimi Titel Title KYT2010 Nationellt forskningsprogram om kärnavfallshantering 2006–2010 – Slutrapport	
Tiivistelmä Referat Abstract Det nationella forskningsprogrammet om kärnavfallshantering (KYT2010) startade 2006. Forskningsperioden slutade vid utgången av 2010. Forskningsprogrammets centrala temaområden är 1) de strategiska utredningarna om kärnavfallshanteringen, 2) bedömningen av säkerheten vid slutförvaring av kärnavfall och 3) de samhällsvetenskapliga utredningarna. I bedömningen av säkerheten vid slutförvaring av kärnavfall ingår tre delar vilka är de tekniska barriärerna, berggrunden och grundvattnet samt frigörelse och transport av radionuklider. Under forskningsperioden har det pågått ca 40 forskningsprojekt som i första hand har hänfört sig till bedömning av säkerheten vid slutförvaring av kärnavfall. Statens kärnavfallshanteringsfond har kanaliserat sammanlagt ca 7 miljoner euro till forskningsprojekten. Följande forskningsorganisationer har deltagit i forskningsprogrammet KYT2010: VTT, Aalto-universitetets tekniska högskola, Helsingfors universitets radiokemiska laboratorium, Geologiska forskningscentralen, Jyväskylän universitet, Östra Finlands universitets campus i Kuopio, Numerola Oy. Arbetet inom forskningsprogrammet har grundat sig på det ömsesidiga samarbetet och arbetsfördelningen mellan forskningsprogrammets ledningsgrupp, två stödgrupper, samordnare och olika forskningsprojekt. I forskningsprogrammets ledningsgrupp har ingått företrädare för Strålsäkerhetscentralen, Utvecklingscentralen för teknik och innovationer – Tekes, Fortum Power and Heat Oy, Industrins Kraft Abp, Posiva Oy och arbets- och näringsministeriet. Kontaktpersoner vid arbets- och näringsministeriet: Energiavdelning/Jaana Avolahti, tfn 010 606 4836	
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KYT2010 Finnish Research Programme on Nuclear Waste Management 2006–2010 – Final Report

The Finnish Research Programme on Nuclear Waste Management, KYT2010, took place between 2006 and 2010. Key themes of the research programme include strategic analyses of nuclear waste management, assessment of the safety of final nuclear waste disposal and sociological analyses. The assessment of the safety of final nuclear waste disposal comprises three sectors: technical barriers, bedrock and groundwater, and the release and transport of radionuclides.

The roughly 40 or so research projects underway during the research period have primarily been concerned with assessing the safety of nuclear waste management. This final report of the KYT2010 research programme presents the programme's objectives, organisation and research projects.

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