KYT2014 Finnish Research Programme on Nuclear Waste Management 2011–2014

Final Report

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KYT2014 Finnish Research Programme on Nuclear Waste Management 2011–2014 – Final Report

Tiivistelmä | Referat | Abstract

The Finnish Research Programme on Nuclear Waste Management, KYT2014, was launched in 2011. The research period closed at the end of 2014.

Key themes of the research programme include new and alternative nuclear waste management technologies, research into the safety of nuclear waste management and sociological research related to nuclear waste management. The assessment of the safety of final nuclear waste disposal comprises four sectors: safety case, capacity of buffer and filler materials, long-term integrity of the final disposal canister, other research on safety.

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.

The following research organisations have participated in the KYT2014 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, Numerola Oy, Ludus Mundi Oy and Tmi Pawel Simbierowicz.

Work in the research programme has been based on mutual cooperation and division of duties between the research programme steering group, three support groups, a coordinator and the research projects. The research programme steering group has included representatives of the Ministry of Social Affairs and Health, the Radiation and Nuclear Safety Authority, the Ministry of Employment and the Economy, Ministry of the Environment, Fennovoima Oy, Fortum Power and Heat Oy, Posiva Oy and Teollisuuden Voima Oyj.

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Foreword

The Finnish Research Programme on Nuclear Waste Management, KYT2014, took place between 2011 and 2014. The research programme continued the coordinated nuclear waste management research programmes of Finland's public administration, launched in 1989. Predecessors of the KYT2014 research programme were the following: publicly funded nuclear waste research programme JYT in 1989–1993, public administration's nuclear waste research programme JYT2 in 1994–1996, public administration's nuclear waste research programme JYT2001 in 1997–2001, Finnish Research Programme on Nuclear Waste Management KYT in 2002–2005 and Finnish Research Programme on Nuclear Waste Management KYT2010 in 2006–2010. Correspondingly, separate research programmes on reactor safety have been in progress.

This final report of the KYT2014 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. Kari Rasilainen and Aku Itälä of Technical Research Center VTT have participated in editing the entire report. The following persons from STUK have participated in writing chapters 3 and 4: Marko Alenius, Rainer Laaksonen and Ari Luukkonen (chapters 4.2, 4.3 and 4.4) and Petri Jussila (chapters 3, 4.1 and 4.4).

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

Helsinki, September 2015
Ministry of Employment and the Economy
Energy Department

Contents

1 Introduction

The Finnish Research Programme on Nuclear Waste Management, KYT2014, was launched in 2011. The research period closed at the end of 2014.

The principles of the KYT2014 research programme are based on the Nuclear Energy Act (990/1987) that emphasises the research needs of the authorities. According to the Nuclear Energy Act 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 assessment of the various ways and methods of carrying out nuclear waste management"* (53 b §).

The research programme aimed to find research topics considered central in terms of national expertise, which needed to be analysed due to their importance. Coordinated projects covering the entire programme period were planned for the topics considered most crucial. 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. 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).

Moreover, the KYT2014 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 and coordinated research projects have aimed to avoid any overlapping research, as well as to coordinate international projects, for instance.

The KYT2014 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. Two projects in KYT2014 programme participated in EU-projects. Carbon-14 took part in EU project CAST 2013–2014 and FinSotec took part in EU project InSotec 2012–2014.

2 Research programme organisation

In organizing and practical working of the research programme essential starting point is that the funded projects are chosen annually based on public call for project proposals.

2.1 Objectives and their attainment

The key objectives of the KYT2014 research programme are written into the framework programme.[1] Key themes of the research programme include 1) new and alternative nuclear waste management technologies, 2) research into the safety of nuclear waste management and 3) sociological research related to nuclear waste management. The research into the safety of nuclear waste management comprises four sectors: safety case, capacity of buffer and filler materials, long-term integrity of the final disposal canister, other research on safety.

Guidelines specifying the framework programme, compiled by the steering group for the annual call for project proposals, have been available throughout the whole research period. First and foremost, the guidelines have specified the topic of assessing the safety of final nuclear waste disposal. Guidelines specified general objectives as well. For instance, in 2013, the call for project proposals 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 and scientific merit, 4) profitability shown in KYT projects or other contexts, and 5) realism of costs and work.

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.

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1 TEM, 2010, Finnish Research Programme on Nuclear Waste Management KYT2014. Framework Programme for the Research Period 2011-2014, Publications of the Ministry of Employment and the Economy, Energy and the Climate 72/2010.

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.

Realism has meant the balance between costs and work as well as the balance between the project timetable and human resources.

During the research period there has been in total 39 research projects which have been either new projects starting in the middle of research period or they have been continuation for earlier ongoing projects. 21 of these projects have been ongoing the whole research period. The research projects have mainly been related to the research into the safety of nuclear waste management. The theme new and alternative nuclear waste management technologies had three projects and social science studies related to nuclear waste management had one project (2012-2014).

The research projects have been ongoing as follows: 30 projects in year 2011, 31 projects in 2012, 29 projects in 2013 and 28 projects in year 2014. The list of research projects is in Annex 1.

During the research period the State Nuclear Waste Management Fund channelled money to the research projects in total about 7 million euros. Annually the money spend for research was ca 1,7 million euros.

Table. Distribution of the VYR funding[2] between research topics 2011–2014.

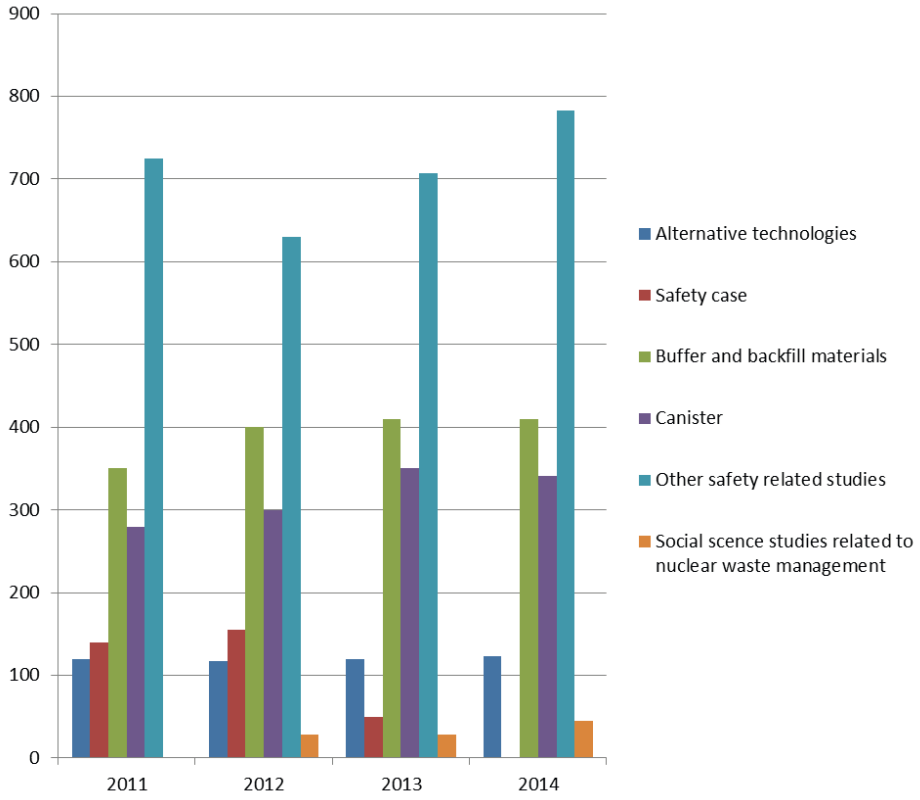
Research area/thousand euros	2011	2012	2013	2014
New and alternative technologies in nuclear waste management	120	117	120	123
Safety case	140	155	50	-
Buffer and backfill materials	350	400	410	410
Canister	280	300	350	341
Other safety related studies	725	630	707	783
Social science studies related to nuclear waste management		28	28	45
Total	1615	1630	1665	1702

Approximately 90% of the funding is directed to the safety research in nuclear waste management. From this share slightly less than half is directed to themes safety case, buffer and backfill materials and canister. The funding to new and alternative

2 In addition to VYR funding, research organisations themselves funded altogether their research projects by around 1 million euros annually.

technologies in nuclear waste management has stayed on the level of ca 7% and social science studies related to nuclear waste management on the level of ca 2-3%.

Figure 1. Distribution of the VYR funding between research topics 2011-2014.



The research projects of KYT2014 have published altogether 50 peer reviewed articles, 174 conference papers or working reports, and 47 theses. The titles of the publications are reported in the Annual review of KYT2014[3], [4], [5]. In annual reviews the projects have reported articles waiting for acceptance and theses in progress, but these are not taken into account in the following tables[6]. Summary of the publications is listed on Annex 2. Some of the publications have been based on the work done on KYT2010 programme preceding KYT 2014 programme. Correspondingly some of the work done in KYT2014 is going to be published in KYT2018 programme, the successor of KYT2014.

3 The Finnish Research Programme on Nuclear Waste Management, KYT2014 2006–2010. Annual review 2011. (in Finnish only).
4 The Finnish Research Programme on Nuclear Waste Management, KYT2014 2006–2010. Annual review 2012. (in Finnish only).
5 The Finnish Research Programme on Nuclear Waste Management, KYT2014 2006–2010. Annual review 2013 (in Finnish only).
6 The conference presentations are often published in peer reviewed journals. In these cases the conference presentations are not mentioned. (In other words publications are only documented once)

Table. Peer reviewed articles by research topic 2011-2014.

Publications by research topic/pcs	2011	2012	2013	2014
New and alternative technologies in nuclear waste management	-	-	-	-
Safety case	-	-	-	-
Buffer and backfill materials	-	-	3	4
Canister	1	4	-	3
Other safety related studies	8	6	8	11
Social science studies related to nuclear waste management	-	1	-	1
Total	9	11	11	19

Table. The number of conference papers and working reports by research topic 2011–2014.

Publications by research topic/pcs	2011	2012	2013	2014
New and alternative technologies in nuclear waste management	7	6	5	6
Safety case	4	4	1	2
Buffer and backfill materials	-	12	7	9
Canister	6	5	8	11
Other safety related studies	12	15	22	23
Social science studies related to nuclear waste management	-	3	4	2
Total	29	45	47	53

Table. Number of theses by research topic 2011–2014.

Theses by research topic/pcs	2011	2012	2013	2014
New and alternative technologies in nuclear waste management	2	2	4	2
Safety case	-	2	-	-
Buffer and backfill materials	1	4	1	3
Canister	-	2	1	1
Other safety related studies	5	6	4	8
Social science studies related to nuclear waste management	-	-	-	-
Total	7	16	10	14

Altogether six doctoral dissertations were finalised during the research period. One dissertation was done in the topic buffer and backfill materials (Suuronen (2014)), one was done in the topic canister (Savolainen (2012)), and four in the topic other safety related studies (Huittinen (2013), Kari (2015), Roivainen (2011) and Voutilainen (2012)). Doctoral theses record long-term work and at least part of the work

mentioned above has been already started before KYT2014 research programme. They may have also included projects outside KYT programme.

The following research organisations have participated in the KYT2014 research programme: VTT – the Technical Research Centre of Finland, Aalto University, University of Helsinki, the Geological Survey of Finland, University of Jyväskylä, University of Eastern Finland, Ludus Mundi Oy, Tmi Pawel Simbierowicz (Subcontractor of VTT 2013) and Numerola Oy.

2.2 Research programme evaluation

The research programme evaluation was implemented between 26 November and 30 November 2012. 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[7].

The research programme steering group reviewed evaluation results systematically in 2013. Operating instructions were further developed. Research project monitoring was developed and follow-up meetings were launched in 2011. To enhance the visibility of the KYT research programme and the co-operation have been postponed and will take place in subsequent programme periods. The development of national nuclear waste management education and training will be continued by the planning group of the course of nuclear waste management, although the pilot version of the course received KYT funding.

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 cooperation and division of duties between the research programme steering group, three support groups, coordinator, and research projects. The research programme steering group has convened normally five times a year (in year 2014 only 4 times since no new call for project proposals was realised on the last year of the research period).

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

7 Apted, M., Karlsson, F. & Salomaa, R. 2013. KYT2014 Review Report. Publications of the Ministry of Employment and the Economy, Energy and the Climate 10/2013, 29 p

has compiled annual recommendations to the Ministry on targeting of VYR funding to nuclear waste management research.

Research programme support groups have convened once or twice a year. During the research period the support groups were as follows: support group I Buffer backfill and canister, support group II Safety assessment and innovations, and support group III Society and man.

The support groups have conducted detailed annual assessments of the project proposals and prepared an annual summary report for the steering group on the basis of their evaluations. The support groups have monitored the progress of research projects. Separate support groups have been managing technical-scientific and social science research.

The Technical Research Centre VTT has been in charge of research programme coordination.

Representatives of the Ministry of Employment and the Economy, the Radiation and Nuclear Safety Authority, the Ministry of Social Affairs, the Ministry of Environment, and organisations in the nuclear waste management industry were appointed as members of the research programme steering group. Kaisa-Leena Hutri (Radiation and Nuclear Safety Authority STUK) has been the chairman of the steering group. Other steering group members are Mikko Paunio (Ministry of Social Affairs), Miliza Malmelin (Ministry of Environment), Sami Hautakangas (Fortum Power and Heat Oy), Veijo Ryhänen (Teollisuuden Voima Oyj), Marjut Vähänen (Posiva Oy), Jaana Avolahti (Ministry of Employment and the Economy). Mia Ylä-Mella (Fennovoima Oy) was appointed as a member of expertise.

Deputy members were Risto Paltemaa (Radiation and Nuclear Safety Authority STUK), Magnus Nyström (Ministry of Environment), Maiju Paunonen (Fortum Power and Heat Oy), Liisa Heikinheimo (Teollisuuden Voima Oyj), Lasse Koskinen (Posiva Oy), Jorma Aurela (Ministry of Employment and the Economy), Hanna Virlander (Fennovoima Oy).

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 KYT2014 research programme, a total of six seminars were arranged. Research topics were handled at four thematic seminars. They focused on one topic at a time and presented relevant viewpoints of research organisations and end-users of results. The research programme, all its topics, and research projects were discussed at the mid-term seminar of the programme period and at the final seminar.

Several stakeholders were informed about the thematic seminars, which were open to all interested parties. The seminars, usually attended by 10-20 people, attracted participants from outside the research programme, too.

Table. KYT2014 research programme seminars

Seminar topic	Time	Location
Bentonite buffer	31.1.2012	University of Helsinki, Laboratory of Radiochemistry
Safety Case	2.2.2012	Geological Survey of Finland, Espoo
Matrix diffusion	13.9.2012	University of Helsinki, Laboratory of Radiochemistry
Mid-term seminar of programme period	17.4.2013	Hotel President, Helsinki
Bentonite Buffer	19.8.2014	VTT, Espoo
Final seminar of the programme period	18.3.2015	Finlandia Hall, Helsinki

2.4.2 Follow up meetings

To facilitate the practical follow up of research projects the projects were divided into separate follow up groups by topic[8]. All projects belonging to follow-up groups (1)-(8) met their support groups 1-2 times a year. The goal of the meetings is to communicate latest research results and 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 year 2011 follow-up groups (1)-(8) convened once (8 meetings), in year 2012 and 2013 twice (16 meetings) and in year 2014 once (8 meetings).

2.4.3 Other contacts

The research programme website (kyt2014.vtt.fi) has been the primary medium for contacts and communication. All material published by the research programme is available on the website. For instance, presentations given in the seminars mentioned above are on the website.

The research programme has published annual plans and annual reviews every year. Interim reports have been compiled in 2011-2014 (3 per year). Bulletins have been prepared to communicate the decisions or discussions by the steering group.

The steering group has also been in direct contact with the research projects by inviting project managers to present their projects in steering group meetings. These project presentations were started in 2012.

8 In year 2014 there were nine follow-up groups. Support group I had follow up groups (1) bentonite buffer, (2) canister, (3) concrete and (4) rock mechanics. Support group II had follow up groups (5) transport of nuclides, (6) microbiology, (7) biosphere and (8) new and alternative technologies in nuclear waste management. Support group III had follow-up group (9) social science.

3 New and alternative technologies in nuclear waste management 2011–2014

According to the Framework Programme the authorities must have access to up-to-date information and expertise on alternative forms of geological final disposal undergoing research and development. From time to time, the management alternatives outlined for various types of nuclear waste are assessed and specified. In such a context, the possibility or need may arise to examine new technical solutions. Research into new and alternative technologies would enhance the security of implementing nuclear waste management in Finland, if the current primary option, geological final disposal, is not realized as planned or if new methods are developed e.g. to reduce the amount of waste generated. This type of research can best be implemented through participation in international cooperation. However, concrete national expertise is a prerequisite for the participation of Finnish research teams in international research programmes.

The projects funded in the programme covered some of the central areas presented in the Framework Programme. Three projects involved with advanced fuel cycles, partitioning and transmutation were involved during the programme period, namely, Advanced nuclear fuel cycles – new separation technologies (HYRL), Advanced Fuel Cycles – Computational Fuel Cycle Analysis (VTT), and Transmutation of nuclear waste in an ADS (Aalto).

The project of HYRL covered the empirical research work of one graduate student aiming at a doctoral thesis in the area of nuclear separation technology. The aim of the study was to develop new nanoporous metal phosphate ion exchangers for the separation of actinides from spent nuclear fuel or from the secondary waste solutions produced by new liquid elution methods. Another aim was to follow up and communicate information on the technological advances.

The project of VTT covered computational fuel cycle analysis in addition to the following up of the new international knowledge on the subject. The goal of the project was to achieve such capability and computational tools that different fuel cycle solutions can effectively be modelled aiming at considerable reducing of the amount and activity of the waste. In the project growth of the Olkiluoto repository capacity when using advanced fuel cycle technologies was computationally assessed. In addition, transmutation of minor actinides in EPR and boiling water reactors were modelled. Activities of international groups involved with advanced fuel cycles were followed up in the project.

The project of Aalto University involved a computational study of transmutation of spent fuel in a lead cooled acceleration reactor. The aim was to critically assess

transmutation techniques and to improve reactor analysis methodologies and their precision in order to compute nuclide concentrations of high burnup fuels. An international project was attended in which MYRRHA experimental reactor was simulated using new computational tools. The goal was to achieve a subjective assessment of the capability of the transmutation method in order to realistically assess the consumption possibilities of the most serious nuclides. Problems related to high burnup were also attempted to analyze in the project.

The characteristics of the projects involved in the programme have been compatible with the Framework Programme policy of follow-up of the international understanding through concrete hands-on activities. The projects have succeeded in producing expertise for the Finnish authority to compare between various ways and methods of carrying out nuclear waste management in Finland. Majority of the achieved knowledge is involved with the reducing of the amount and activity of the produced waste and the subsequent consequences on the implementation of the geological disposal.

The organizations involved have a long mutual cooperation history in the past national programmes. The international networking has been a central goal for the projects in order to follow up knowledge and to maintain expertise.

A central content of the projects has been the training of experts through hands-on work and following up international knowledge. The projects have produced several institutional and conference reports. One doctoral thesis is to be produced in the project of HYRL.

4 Safety research in nuclear waste management 2011-2014

Research into the safety of nuclear waste management was related mainly to three central topics: safety case, buffer and backfill materials, and canister. The coordinated projects focused on these topics have been long-term joint efforts done by several research institutes. The research programme has also included individual projects done by one research institute under the theme other safety related research.

4.1 Safety case

According to the Framework Programme, as the authorities are responsible for assessing the license applicant's safety case, a sufficient amount of high quality information, independent of the applicant, must be available to the authorities. Such information must refer to the safety case's underlying principles, philosophy and restrictions. Furthermore, a coordinated project has the aim of training new experts in Finland, qualified in compiling and assessing safety cases. A further goal is to devise new methods of assessing the long-term safety of geological final disposal. Through hands-on work, the idea is to understand the philosophy (based on safety case scenarios), methods of acquiring and generalizing information, the performance of practical analyses (based on different calculation models), and methods of assessing the reliability of results and the related uncertainties. For this purpose, work will begin on compiling a safety case for a final disposal solution, of the KBS-3 type, in crystalline bedrock.

The coordinated safety case project LS-TUPER originally consisted of several subprojects, which in addition to coordination, mentoring and synthesizing included the following areas: safety case methodology and presentation, alternative conceptual models and interpretations, scenario development, development of computational analysis model, development of uncertainty analysis methods, and complementary considerations. In general, these subprojects are compatible with the safety case areas presented in the Framework Programme. Unfortunately, the goals set for the project were not achieved and the project was ended before the end of the programme period. Consequently, all the aimed subprojects were not carried out.

The benefits of the prematurely ended project were less than originally planned. The very existence of the project and the analysis of its content and results have contributed to the expertise of both the project participants and the authority on the scope, problematics and importance of the subject. The report produced in the project provides with a general view of the safety case's role and, consequently, expresses one of the goals of the project to spread knowledge of the disposal safety case for wider audiences.

Generally, the national networking of the coordinated project was good. There were participants from three major national organizations and from two small enterprises. Project participants took part in EU research programme CROCK. A matrix diffusion workshop was held in which there were participants from Finnish and Swedish organizations.

New safety case experts were trained in the project. A special assignment on radionuclide transport in geosphere and a master's thesis on ground water flow modelling were produced in addition to one institutional report on the safety case's role. National workshops were arranged and international conferences were actively participated.

4.2 Buffer and backfill materials

In the KBS-3 concept the reliable assessment of buffer and backfill materials performance determines to large extent the credibility of the safety case. For that purpose the authorities must have sufficient amount of high-level expertise about the performance of these materials and its effect on the long-term safety. The need to focus research on buffer and backfill materials has been written in the framework program (KYT2014) aiming at for example the development of coupled modelling of processes ensuring performance and testing methods of bentonite microstructure.

Buffer and backfill materials performance related modelling and the testing methods of the bentonite microstructure were developed in a coordinated project "Assessment of bentonite characteristics" BOA. The coordinator of the project was VTT and the other participants were universities, research organizations and private enterprises. The participation time of organizations within the coordinated project varied from one year to four years.

The research plan of the coordinated project (BOA) agreed well with the research item areas listed in the framework program. The project was focused on characterization and modelling. The project both studied material properties and developed methods for the determination. During the project, research methods of bentonite microstructure in different scales and pore water testing methods were developed. Within the project solubility studies with montmorillonite and also studies with the formation of colloids of different materials were carried on. The strength properties of bentonite blocks corresponding to backfill blocks were determined in one sub project for both block to block interfaces and block to different materials interfaces.

The original project plan for BOA contained a significant investment on coupled modelling of bentonite, but the fraction of coupled modelling was reduced by the end of the project. On the other hand, in the other modelling branch of the project, namely phenomenological modelling of bentonite based on empirical results, empirical research methods were developed, such as tomography and material models and

determination of material parameters. The empirical model was successfully implemented in a commercial code. Although the project did not proceed substantially in describing the relationships between the buffer and backfill performance and bentonite microstructure or coupled processes, the characterization and modelling procedures developed are available for use in the next research projects within this area. The authority has been able to partially utilize the results from the project during the review work of the Posiva's construction licence application material.

The coordinated project, BOA, formed a nationally significant consortium and fulfilled well the goals of the framework program in relation to networking and integration of bentonite know-how. The project had, part of the time, a firm coupling to one EU-project (BELBaR) and during the project some international cooperation has been launched.

Several master's degrees were graduated during the project. Several post graduates for doctor's degree were working in project, and one doctor thesis was successfully defended during the project (Suuronen 2014). The project has generated lectures, posters and articles in excess to master's degrees. The project has raised the level of know-how and trained new experts within this issue. The scientific merits are the extensive development of different characterization methods for the bentonite and development work for the phenomenological model together with the development of parameter determination and the implementation of the model.

4.3 Canister

According to the framework programme sufficient amount of high-level expertise regarding the long-term durability of the final disposal canister, the key factors affecting this and the methods applied in assessing long-term durability must be available to the authorities. Canister research projects within the KYT programme have met this need.

KBS-3 concept is based on the multi-barrier principle which consists of mutually complementary engineered barriers and natural barrier. The final disposal canister is considered the most important barrier and its long-term performance is important to the safety of the concept. When assessing the performance of the disposal canister factors that have effect on the mechanical and chemical integrity shall be taken into account.

During the KYT programme canister research projects concentrated on the mechanical and chemical performance of copper material. Since 2012 canister research projects made up the coordinated project L-TICO (Long-Term Integrity of Copper Overpack). VTT was the project coordinator and other participants within this coordinated project came from the Aalto University. Two canister research projects regarding the mechanical integrity of the canister and four research projects regarding chemical integrity of the canister were implemented during the KYT programme. Three projects of these covered the entire KYT research period. In

addition to the cooperation between the canister research projects there was also cooperation with Swedish research institutes and a Japanese University.

Posiva's construction licence application for the encapsulation plant and underground final repository was reviewed by the authorities during the second half of the programme period (2013-2014). Research results regarding the mechanical and chemical integrity of the canister were utilized in the review work conducted by the authorities.

One aim of the KYT programme is to promote the training of a new generation of experts in the field of nuclear waste management. Canister research projects produced several master theses. Most of the master thesis workers continued working as researchers within the canister research projects. During the KYT programme one doctoral thesis (Savolainen 2012) was also conducted. These theses conducted during the KYT programme and new researchers fulfilled the aforementioned training aim well. Research results were also published in the seminars and conferences and in peer-review articles. Scientific merits of the canister research projects are e.g. creep testing of copper and its weldments and creep modelling, deformation and failure mechanisms of copper and its weldments, and corrosion mechanisms of copper.

4.4 Other safety related studies

Support group I projects

The need to target research on long-term behaviour of concrete structures in final disposal conditions has been registered in the framework program (KYT2014). This registration was made because concrete will be, in any case, needed in the final disposal of spent fuel, plant operational waste, and plant disassembly waste.

The long-term behaviour of concrete structures was studied in VTT and Aalto University projects "Durability of concrete release barriers in final disposal of operational waste". The research projects lasted the complete period of the framework program.

The target for the research projects was to increase the knowledge on durability of concrete barriers in final disposal conditions, and to create capabilities for evaluating more realistically the service life of release barriers. The projects had two main targets. Aalto University attempted to generate a concrete degradation model based on thermodynamics. According to the modelling hypothesis, the durability of concrete cannot be described only with chloride diffusion, but more diverse chemical factors need to be taken into account that have significance as initiators of iron reinforcement corrosion. Another target of Aalto University project was to find from literature and by experimental studies suitable parameter values that were needed in thermodynamic modelling. VTT's objective was to document, with diverse research analyses, the conditions of concrete samples immersed in various water type pools. The immersion started in 1997 on behalf of power companies (Fortum and TVO), and the target of analyses was to estimate potential degradation mechanisms

for concrete and use the results for the calibration of the straightforward chloride diffusion model. VTT also offered its analytical results as input for more complex modelling approach favoured by Aalto University.

Research programs of the projects covered well one of the specific safety research areas listed in the framework program. The results produced in the projects are of value to owner companies of Finnish power plants, and later on also to owners of other nuclear facilities. Licensees also funded partially these projects.

The two active parties in the studies are both nationally significant expert organisations in theory and experimentation of concrete and reinforced concrete studies. Regarding the international networking, study and congress journeys did take place.

The educational contribution promised at the application stage was one doctoral thesis. This thesis was finalised by an Aalto University project researcher at spring 2015 (Kari 2015). The thesis was compiled from three scientific articles and two conference articles. Analytical results produced by VTT have been published in VTT report series. The target of thermodynamic modelling work implemented by Aalto University was to create a modelling tool that covers more universally, than experimented in the long-term immersion tests, concrete degradation evolution potentially met in the operational waste repositories during operation and following 500 years after closure of the repositories. VTT's analysis program produced versatile results of the long-term concrete aging experiment that is exceptional also internationally.

Support group II projects

According to the Framework Programme, when assessing the safety of nuclear waste management in general, and the safety of geological final disposal in particular, information is needed from several disciplines and beyond the coordinated projects mentioned above. The research topics of these projects outside the coordinated ones vary considerably. Their topics were compatible with the topics mentioned in the Framework Programme, but did not completely cover all the areas explicitly presented in the Framework Programme.

The educational impacts of these projects were various. New experts have been trained in the projects and the total amount of theses is considerable. In addition, some of the projects have involved active work of senior researches or have resembled basic research.

Projects involving transport of radionuclides were the following: In situ long term diffusion experiments (HYRL), Release of 14C (VTT), Including heterogeneous rock structure in the modeling of matrix diffusion (JYFL), Chemical forms and sorption of radiocarbon in the geosphere (HYRL), and Sorption of trivalent actinides onto clay and (hydr)oxide minerals (HYRL). These projects e.g. provided with information on retardation of essential radionuclides in situ and in laboratory, developed laboratory and modelling methods to study matrix diffusion in a very small scale, and provided

with information on the behavior of chemical forms of radiocarbon in the geosphere and the release of radiocarbon from metallic decommissioning waste.

The projects involving radionuclide transport have produced information on the behavior of some of the most safety relevant radionuclides. The information has been available in the assessment of both the initial data and the results of the safety analysis. The new information on the behavior of radiocarbon has been useful in the assessment of the uncertainties of the safety case. In general, the projects' national and international networking has been fine. Some of the projects have shown integral mutual cooperation. The projects dealing with radionuclide transport have produced more than a dozen published or submitted scientific papers, several institutional reports and seminar presentations, and four theses. The two doctoral theses (Huittinen (2013) and Voutilainen (2012)) were initiated during the previous programme period.

Microbiological phenomena were examined in the following projects: Saline fluids, gases and microbes in crystalline bedrock (GTK), Characterization of deep groundwater microbial communities (VTT), Deep bedrock bioinformatics (Aalto), Risks of microbiologically influenced corrosion in the Finnish nuclear waste repository (VTT), and Microbial communities in the gas generation experiment (VTT).

These projects e.g. examined possible mutual relations and interactions of gases and microbes in saline ground waters, generation, development, delay, and possible safety relevance of gases and salinity in geosphere, produced information on the behavior, metabolic routes and related corrosion risks of deep ground water microbial communities, developed methodology and tools for bioinformatics, studied development of biofilms and microbiological corrosion risk in metallic decommissioning materials both in laboratory and in situ, and studied the generation rate of gas, degradation rate of maintenance waste, and enrichment of microbial species in the 15-year gas generation experiment in Olkiluoto LILW repository.

The information gathered in the microbiology related projects has helped to cope with the uncertainties related to the disposal safety case. The projects have improved knowledge on ground water chemistry and the effect of microbes on the engineered barriers. This information has been available in the assessment of the safety case. The nature of some of the projects has been basic research. The cooperation between the microbiological projects has been exemplary. National and international networking has been excellent. The projects have remarkably improved the national expertise, and their educational impact has been significant. The studies have contributed to two doctoral theses and to several lower degree theses.

A project called Use of empirical data to improve radioecological modeling applied to risk assessment of radioactive waste (UEF) covered phenomena in the biosphere. The project studied empirically the transfer of nuclides from soil to plants and animals and developed radioecological modelling methodology applicable to Finnish forest ecosystems. The important results of the project of UEF have been available when assessing the initial data and the results of disposal safety analysis. The project

was not interconnected with the other projects in the program. However, there has been some connection with the Posiva studies and the results have been published in international conferences. The project produced a doctoral thesis (Roivainen (2011) and three lower degree theses.

In addition, a project called KARMO – Mechanical Properties of Rock Joints (Aalto) started during the last year of the programme period. The project studies mechanical behavior of rock fractures with 3D printing technology and modelling. The project was of a preparatory nature for the next programme period. The results of the project can be available in the assessment of the safety and the implementation of the disposal.

5 Social science studies related to nuclear waste management

The purpose of sociological research, related to nuclear waste management conducted within the KYT programme, is to support decision-making and related preparations. Nuclear waste management can only be carried out subject to a licence, and is influenced by societal values and expectations. The views of various groups on nuclear waste management, and the final disposal of spent nuclear fuel in particular, were considered important topics. Those views are affected by various actors' independence and reliability as well as ethical perspectives and long-term duration of waste management.

Between 2012 and 2014, the research programme financed a sociological research project on different perspectives of the plans of nuclear waste management. On the basis of the research results the socio-technical challenges were defined as well as factors influencing those challenges. In addition, the dialogue between Posiva and STUK was assessed focusing on the safety and the risks.

The results lend themselves to use as part of decision-making and comparative assessments. 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.

6 Abstracts of projects

The abstracts of the research projects of KYT2014 program are presented in this chapter in accordance with project list in appendix 1. Abstracts have been written under the leadership of project managers.

6.1 Advanced fuel cycles – New separation techniques (Project 1)

Risto Harjula, University of Helsinki, Laboratory of radiochemistry

About the research

Partitioning and transmutation (P&T) of the long-lived radionuclides contained in nuclear waste is a part of the ongoing world-wide research on the topic of next generation nuclear technology. With new P&T technologies the aim is to maximize the fission energy gain from nuclear fuel and also to minimize long term radiotoxicity of spent nuclear fuel. Depending on the technique used, P&T is estimated to lower the radiotoxicity of nuclear waste in their repository by an order of 10 to 100 in comparison with direct disposal.

The separation techniques developed in P&T concept can be divided in hydrometallurgical and pyrometallurgical methods. Hydrometallurgical methods in development (UREX, TRUEX, DIAMEX, GANEX and SANEX) are upgrades or alternatives to the already in use PUREX-process, where organic extracting reagents are used. These new developments often concentrate on separating uranium and transuranium elements (Np, Pu, Am, Cm) onto clean fractions ready for transmutation. These advanced methods are still done in laboratory scale and the separations have shown to be very complex since it is difficult to separate transuranium elements from similar lanthanides.

The drawback on hydrometallurgical processes is often limited radiochemical stability of the used organic extracting reagents and therefore they might not be very suitable for processing of the high activity transmutation fuels. Also, hydrometallurgical processes form considerable streams of secondary active liquid waste and there is a loss of actinides for the end product for transmutation, and radiotoxicity in the final waste repository if not lowered as much as possible. There is also interest in using inorganic sorption materials for uptake and separation of actinides, which is the theme of our project.

In our project we take advantage of inorganic ion exchange resins. These materials can be used in chromatography columns, they are resistant to radiation damage and in ideal case, reusable. In addition, the materials developed in this project can also

be used as an addition to the already existing liquid extraction methods to reduce active waste volume, independent of the advances of transmutation technologies.

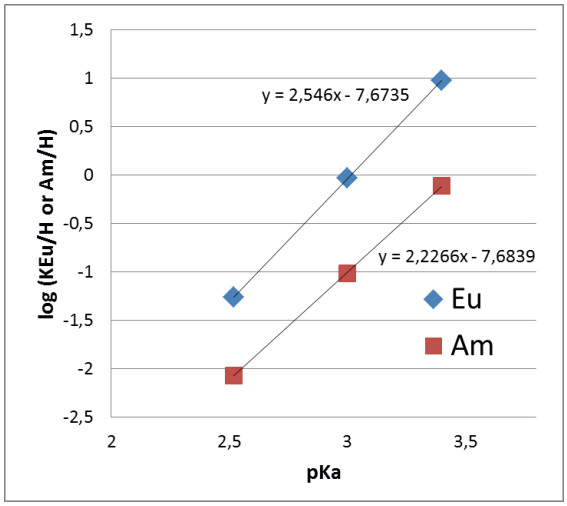
Methods used and main results

In this project, several inorganic ion exchange materials, mainly zirconium phosphates (ZrP) and zirconium titanium mixed phosphates (ZrTiP) were investigated for the use of actinide-lanthanide separations in acidic conditions. The most promising materials investigated were layered crystal structural α-zirconium phosphates (α-ZrP). Unlike other studied ZrP, TiP and ZrTiP materials, the α-ZrPs can be readily synthesized as grainy materials suitable for column use, without use of additional binding materials.

With the use of nuclides Eu-152 and Am-241 as models for lanthanides and actinides, respectively, in column separation experiments a successful separation was shown, which was one of the main goals in the project. However, impractical volume of eluting acid was required for the separation. Notably only nitric acid with varying concentration was used for the whole process, and no additional reagents of extractants typical for other separation methods were needed. For some ZrPs synthesized the suitable pH region for uptake and separation was around 1, as for some it was around 2 to 3.

One remarkable main result is the possibility to change α-ZrPs selectivity towards actinide and lanthanide ions based on the synthesis. Even though several α-ZrP materials synthesized in different ways had the same chemical composition and crystal structure, they had different acidic properties and selectivity for Eu and Am. The bigger the pKa, the bigger the selectivity for both, but with a greater effect on Eu (Figure). An important topic to further investigate will be which factors in the ZrP synthesis have an effect on the acidity and selectivity.

Figure 2. Three different α-ZrP selectivity constants (log kEu/H and log kAm/H) as a function of acidity (pKa).



All of the syntheses were done in aqueous solutions. The synthesis of ZrPs is relatively well known in literature, as ZrPs have been used for different purposes in the past, e.g. ion exchange of lighter elements. Titanium and mixed metal phosphates were synthesized in an analogous manner to ZrP. The applicability of a product for Ln/An separation was tested as batch and column experiments. With batch experiment, distribution coefficients for model nuclides Eu and Am were determined. The distribution coefficient tells how the nuclide in question is split between the exchange material and the solution, in equilibrium. Distribution efficients suitable for Eu/Am separation was requirement for a material to be studied further. With column experiments, optimal conditions for the actual separation were investigated. Radioactive nuclide analytics was done with gamma spectroscopy. In addition, inactive experiments were done with stable europium using mass spectrometric analysis (ICP-MS).

For the characterization of the materials studied, multiple methods were used: electron microscopy (FESEM), x-ray spectroscopy (PXRD), infra-red spectroscopy (FTIR), thermogravimetry (TGA). By using these characterization methods we resolved suck properties as the crystal structure, zirconium-phosphorous ratio, macroscopic shapes of the crystals, acidity, crystal water mass.

Use of the results

This project along with its inclusion of the doctoral training will develop expertise of this area in Finland and helps Finnish research institutes to get involved in international P&T collaboration. In medium term, the materials developed in the project could be applied to the processing of already existing liquids e.g. at spent fuel reprocessing plants. In long term, the materials can be applied to the new P&T concept fuel cycles. The Finnish expertise in medium and low-level activity nuclear waste processing (e.g. Fortum, TVO) is assessed to have use in international P&T research at least on the topic of minimizing the secondary wastes in spent nuclear fuel processing.

6.2 Advanced Fuel Cycles – Computational Fuel Cycle Analysis, KEPLA (Project 2)

Silja Häkkinen, VTT

Research Topic

Advanced fuel cycles are all about effective, safe and economic use of nuclear fuel. Some of the specific objectives of advanced fuel cycles are for example reducing the volume, heat production and radiotoxicity of nuclear waste, and proliferation resistance. The goal of this project is to acquire competence and computational capabilities to effectively simulate different fuel cycle options in order to significantly reduce the volume, heat production and radiotoxicity of nuclear waste. This project

has concentrated in scenario calculations and minor actinide burning in thermal light water reactors. International research on the field has also been followed in the framework of the project.

Scenario calculations

Scenario calculations were started in 2011 by acquiring the COSI6 [1] simulation software from CEA. The software is capable of modelling the whole nuclear fuel cycle from mining to final disposal. The first COSI calculations were done in 2012. The first study involved modelling the possible transition from thermal light water reactors to fast reactors in Finland. The main result was that the plutonium stock accumulated in Finnish thermal reactors is sufficient for starting the required fast reactors. Also, the volume of plutonium and minor actinides were reduced when they were recycled in fast reactors. The results of the calculations were reported in IEMPT2012 conference.

In 2013, the scenario calculations were continued by calculating the heat production of nuclear waste in different scenarios in Finland. Since heat production of nuclear waste is the most restrictive factor in the packing density of the disposal capsules, it would be possible to increase the capacity of the Olkiluoto repository by reducing the amount of heat generating transuranic elements by means of partitioning and transmutation. Recycling of plutonium and the minor actinides significantly reduced the maximum heat production of nuclear waste compared to the scenario where only light water reactors were used. The results of the calculations were presented in GLOBAL2013 conference.

The proliferation resistance of the different scenarios in Finland was evaluated in 2014 based on the earlier calculations. The proliferation risk was estimated using Charlton's method [2] that gives a single value between zero and one, corresponding to the risk related to a particular scenario. The evaluation was only preliminary because it wasn't possible to calculate all the attributes related to the value. According to these indicative results, recycling of plutonium and minor actinides in fast reactors improves proliferation resistance. On the other hand, partial recycling may degrade proliferation resistance compared to an open fuel cycle in light water reactors. The results of the calculations were reported in IEMPT2014 conference.

Minor actinide transmutation in light water reactors

Minor actinide transmutation in thermal pressurized and boiling water reactors has been studied in this project using CASMO-SIMULATE and Serpent programs. In order to model MOX fuel correctly, a new version of SIMULATE was acquired during this project.

In 2011 and 2012, an EPR reactor was modelled using 100 % and partial MOX fuel loading in the core. The calculations were done using CASMO-SIMULATE software. The burning of minor actinides in the EPR core was unsuccessful in both cases. In 2013, an error was found in the initial minor actinide quantity of the minor actinide

bearing fuel. Therefore, the calculations were repeated once more in 2013. In the new calculations, slow burning of minor actinides was observed. The results were similar to those in earlier calculations of other institutes.

In 2013, burning of minor actinides was modelled with Serpent in the upper part of a boiling water reactor, where it was possible to get a fast neutron spectrum by appropriate choice of fuel and cladding. At this point, the calculations were limited to zero burnup and two dimensions. Actinide fission to absorption ratios and parameters related to reactor safety were calculated. Two types of fuel with hafnium cladding were used. The first fuel was PuZrN and the second fuel was MOX fuel with 40 % plutonium content. The fission to absorption ratios were significantly better for these types of fuels compared to conventional UOX. However, parameters related to reactor safety were questionable especially for the PuZrN fuel.

The boiling water reactor calculations continued in 2014. The same MOX fuel with hafnium cladding from the earlier calculations was studied. Additionally, MOX fuels with different, much smaller, plutonium contents and conventional zirconium cladding were considered. The small increase in plutonium quantity did not significantly improve the fission to absorption ratios. The ratios were clearly better for the fuel with hafnium cladding. However, the parameters related to reactor safety were significantly poorer. In order to reliably estimate the parameters related to reactor safety, the calculations have to be extended to three dimensions in the future.

International research

In this project, also international research in the field of advanced nuclear fuel cycles has been studied. The project personnel have attended the annual meetings of OECD/NEA working group WPFC and the expert group AFCS. In addition, the personnel have participated in GLOBAL and IEMPT conferences which are concentrated on advanced fuel cycles. Travel reports have been written of these journeys. The use of COSI6 software has been studied in France under the guidance of CEA in three years. In 2011 and 2014, a report on the development of international research on the field was written together with HYRL project “Advanced Fuel Cycles – New Partition Techniques”. Cooperation was also organized with the Aalto University project “Transmutation of Nuclear Waste in ADS Reactors”. The cooperation was mostly implemented in the form of meetings where the current matters of the projects and new research ideas were discussed.

References

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W. S. Charlton et al., ”Proliferation Resistance Assessment Methodology for Nuclear Fuel Cycles”, Nuclear Technology, 157, p. 153-156, 2007.

6.3 Transmutation of nuclear waste in an ADS reactor, FLUTRA (Project 3)

Pertti Aarnio, Aalto University

Introduction

An alternative for once through nuclear fuel cycle is recycling. It requires partitioning and, at least, rudimentary transmutation. In FLUTRA-project we have studied transmutation of actinides and important fission products. Our main emphasis has been in the transmutation of high burnup (60 MWd/kgHMI) BWR fuel in accelerator driven fast MYRRHA reactor. In addition, we have modelled transmutation in BWR, PWR and CANDU reactors. As an alternative for nuclear fleet consisting both PWRs and CANDUs we have also considered DUPIC (Direct Use of PWR spent fuel In CANDU) fuel cycle.

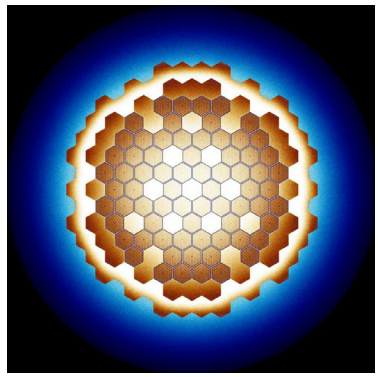
We characterized the spent fuel using CASMO4E/SIMULATE code package, which was used to calculate the amount of different nuclides, their axial distribution and decay heat production.

Burning minor actinides and fission products in MYRRHA

MYRRHA is driven by a 600 MeV 3.5 mA proton beam hitting a lead bismuth target at the middle if the core (Figure) producing spallation neutrons. We have modelled the proton beam and its interactions using FLUKA high energy transport code. Once the neutron energies fall below 20 MeV they are handed over to reactor code Serpent for further transport.

When subcritical, k_{eff} around 0.95 to 0.97, the reactor is kept in operation by the proton beam only. We found in our simulations that one proton produces about 11 fast spallation neutrons and the energy amplification factor is 36 corresponding to the thermal power of 76 MW. The simulations were carried out at pin, assembly and full core geometries as well as in critical and subcritical conditions.

Figure 3. Neutron collision densities in MYRRHA.



We calculated the transmutation of minor actinides in homogenously MA doped MOX fuel (25 % Pu) and, of course, found an increase in their absolute amount. However, after normalizing it against the same energy produced in an LWR the MA burnup was significant. The burnup is shown in the following table.

Table. Burnup of MA in MOX calculated using FLUKA/Serpent code at fuel burnup of 100 MWd/kgHMI

Isotope	Np-237	Am-241	Am-243	Cm-243	Cm-244
Burned	38.2 %	41.2 %	36.5 %	48.0 %	28.4 %

The important fission products ^{99}Tc and ^{129}I were burn in a heterogeneous geometry replacing the center pins of fuel assemblies with I and Tc. The incineration was slow; the burnup of 20 % took 1200 days.

Burning fission products in thermal reactors

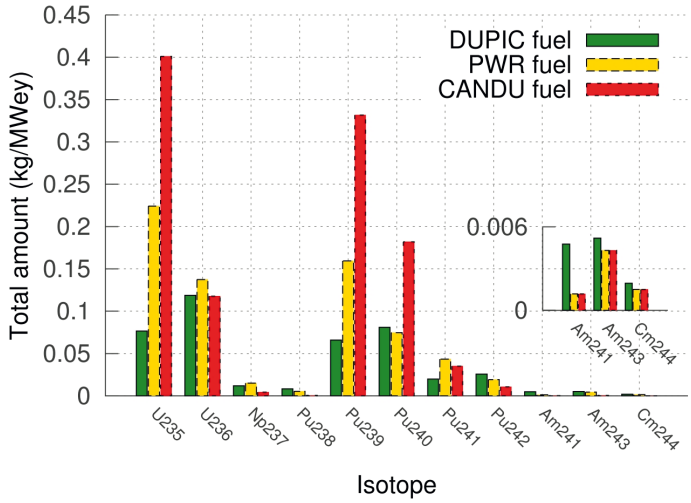
In addition, we simulated heterogeneous incineration of ^{129}I and ^{99}Tc in thermal reactors using the isotope ratios calculated with CASMO4E. We use metallic technetium and iodine as cerium oxide. The PWR burnups of ^{129}I and ^{99}Tc in 1200 days were 16 and 13 per cent, respectively. The BWR burnup rate of iodine was essentially the same as in the PWR. However, due to a softer and higher neutron flux of a CANDU its corresponding value was 25 %. Technetium was not burned in a BWR or CANDU.

DUPIC fuel cycle

DUPIC fuels is produced in the so called OREOX (Oxidation and Reduction of Oxide fuel) process in which the spent fuel is first decladded and then the fuel repeatedly goes through oxidation and reduction at high temperatures until a complete oxide powder is formed. The powder can then be sintered to CANDU sized pellets. The volatile and most of the semi-volatile materials get removed due to the high temperatures used in the process.

We have calculated the amount of important actinides produced in three fuel cycles: PWR, CANDU and DUPIC. In the DUPIC cycle all spent PWR fuel, and it only, is reused after OREOX process in CANDU reactors. The result is a significant reduction of most actinides, with the exception of heavier actinides due to the higher total burnup (see Figure). In addition, the amount of spent fuel and natural uranium are saved by 60 and 18 per cent, respectively.

Figure 4. The amount of important actinides in the spent fuel of DUPIC, PWR and CANDU once through fuel cycle.



Education

An important contribution of the FLUTRA project is also the education of new nuclear experts as well as more sustainable recruiting of new students. Six BSc's have graduated from the project and five special assignments, with the requirements equal to the BSc, has been done.

6.4 Safety case for final disposal: coordinated project LS-TUPER (Projects 4–7)

Markus Olin, VTT (ed.)

Goals and Safety Case

The main goal was to learn the processes needed in carrying out complete Safety Case for a KBS-3 type final repository for spent nuclear fuel. The major learning mechanism was by doing self, while also following the literature and work done by license applicators (Posiva, SKB) was planned. The project was targeting mainly to computational safety analysis methods, which is supported by some important topics mainly is scenario development.

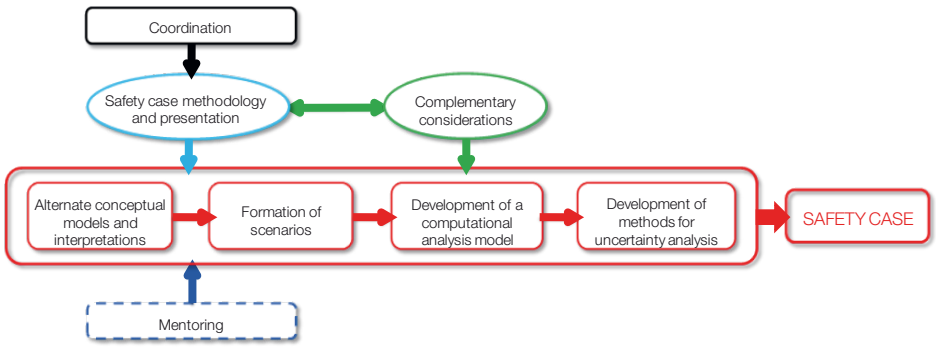
KYT2014 programme included options for coordinated project in three different areas, of which one was safety case for spent fuel final disposal. In Finland the knowhow for safety case was identified to be limited to certain aspects and mainly used for license applier's needs.

A safety case consists of large set of documents (typically thousands of pages) totally be provided by license applicant; it aims to show the fulfilment of all long term safety requirements. Therefore, it is not needed and not even possible to produce a whole parallel safety case either by regulator (STUK) or scientific community (KYT). However following is needed to

- Train new experts and scientists
- Learn by doing on some specific carefully chosen topics
- Propose new or alternative approaches or models
- Inform about the research done on the topic – in Finnish and in an easily understandable way.

The plan to carry out the project is shown in the following figure, while the results finally obtained are under chapter results.

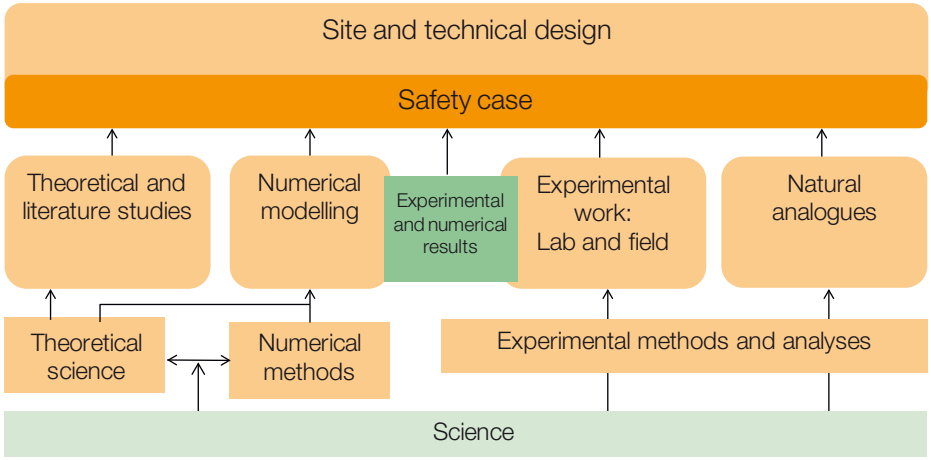
Figure 5. A sketch of LS-TUPER project.



Utilisation of the results

New experts were trained for safety case work, for both performing and evaluating, in Finland. In parallel, some new ideas for analysing long term safety of geologic final disposal of spent nuclear fuel were proposed. All results of the project were readily available for the use of regulator. The concept of safety case was clarified to larger audience. However, no new own computational model was succeeded to develop.

Figure 6. Safety Case stands on science.



Results

In coordination task internal, domestic and international collaboration was planned and coordinated. The goal is in developing natural ways of working, which enable the change of information and integration of the results optimally from the point of view of the project's targets. The major collaborative project was the EU CROCK, while something was done with the EU BELBaR, too.

In Methodology and ways of presentation in Safety Case some recent Safety Cases were reviewed. On that basis of own approach for carrying out a Safety Case project, ways of thinking and limitations was planned to be formed. However, after vivid discussions in the project team, no real breakthrough was created.

Alternative conceptual model and interpretations were studied: spent fuel, copper canister, buffer, and bedrock, and interfaces between them. New points of views were especially searched. The starting point was in colloid and sorption models. In addition, in cooperation with VTT's nuclear fuel scientists, better approaches for IRF problematics were considered (Instant Release Fraction). Karita Kajanto finalised a special assignment in the supervision of Veli-Matti Pulkkanen: goal was to study the effect of fracture geometry on the transport numerically by COMSOL

Scenario development was started by a review on the subject followed again by vivid discussion inside the project group. However, no real breakthrough approach was found.

Development of a modular, easy to use and cheap *computational model* for estimation of safety was aimed. However, both modelling approaches (Monte Carlo based and adaptive multigrid) appeared to be difficult to implement on the available resources.

The goal was to estimate *uncertainties* caused by complexity of the final repository system and long time periods by mathematical and conceptual methods. This topic

started slowly, but finally Karita Kajanto finished her Master of Science thesis about the subject; in the thesis the effect of stress state of bedrock on the fracture aperture and via it to flow field was studied.

In the supplementary studies and ways to apply these were investigated. The work was started by a short analysis of the copper in some previous safety analysis works. A review of metallic copper natural analogues and of the environments of metallic copper occurrence was written. The topics were natural analogues of engineered barriers and migration phenomena together with hydrogeological history of the final disposal site; some of these may be have importance even in the licensing of the Olkiluoto site.

Supervision of work or *mentoring* of scientists is an essential part of a LS-TUPER kind of strongly training oriented project. Merja Tanhua-Tyrkkö participated Erasmus Mundus programme (IMACS- International Master in Advanced Clay Science) clay course at the Poitiers University: Molecular modelling, Thermodynamic functions: experimental and theoretical approaches and Modelling of fluid-clay interactions in the environment. Veli-Matti Pulkkanen supervised both Karita Kajanto's special assignment and Master of Science thesis. Markus Olin as project manager and professor was guiding all VTT scientists in the project.

The synthesis was planned to show how in the project via various steps is finally get on to own Safety Case. However, the project ended before real work on synthesis even started.

6.5 Assessment of bentonite characteristics: coordinated project BOA (Projects 8-15)

Markus Olin, VTT (ed.)

Safety importance

The bentonite buffer is one of the most essential components in the KBS-3 concept:

- it limits the mass flow to and from the copper canister
  - to prevent corrosion and
  - in the case of canister failure, it retards transport of radionuclides to bed rock
- it protects the canister in the case of large dislocations of bed rock.

In the KBS-3 method, the reliable estimation of functioning of buffer and backfilling materials protecting the copper canister and spent fuel therein is a bottom line issue for safety. For that target, know-how about the behaviour and long-term functioning of these materials, from the point of view of long-term safety is needed. Our aim was to produce just that data and knowledge, the work being divided into three major topics: THM, THC and THMCB (Thermal-Hydrologic-Mechanical-Chemical-Biological).

Block Shear Experiments

"Block Shear" has been investigating the shear resistance between various tunnel backfill interfaces in the KBS-3V. The project has been active during 2012-2014 and mainly involved with testing at the Geotechnical laboratory at Aalto University. In the past, not much attention has been given to the interface shear resistance between tunnel backfill materials; however, it plays an important role in controlling the swelling of the buffer, especially during the early stages of saturation of the repository where a saturated backfill could swell and uplift the dry-backfill. Thus, in order to ensure the saturated density criteria of the buffer (1950–2050 kg/m^3) at all times a detailed modelling with reliable interface shear resistance parameters was needed. Therefore, the aim of the "Block Shear" was to determine the shear behaviour for modelling and also to get insight into the shearing behaviour of various backfill interfaces in KBS-3V under different likely scenarios.

Typical tunnel backfill material interfaces from KBS-3V were tested in a direct shear box under different conditions. The effect of more likely variables such as interface water content, groundwater salinity of the interface water and temperature, on the interface shear resistance was investigated. Proposed typical backfill materials such as precompressed Friedland clay blocks, bentonite granular materials like pellets and granules were used in testing.

The internal shear strength of bentonite pellets and granules decreased with increasing water content. The results showed that the interface shear resistance increased with increasing salt concentration of the interface water. This could be attributable to the formation of the rough surface due to high salt concentrations. When the water content in the interface were studied following observations were made: (i) when the water contents of materials were less than 40%, the shearing took place at the interfaces, and (ii) when the water contents increased above 40%, the shearing predominately took place within the granulated bentonite and pellets (i.e. no more failure at the interface). Only marginal differences were observed in the interface friction when the block to block interfaces were tested over a range of temperature between +20°C to +80°C.

Mineralogical characterization

The mineralogical characterization is a primary tool in estimating the use of bentonite material as buffer and backfill material. There are big differences between different types of bentonite deposits. The knowledge of the bentonite mineralogy is useful when interpretations of the chemical analyses are made. Bentonite material is composed mainly from smectite minerals (usually montmorillonite) together with minor amount of accessory minerals (e.g. quartz, feldspars, micas, phosphates, carbonates, sulphides, sulphates and iron oxides). The purification process of bentonite material removes the major amount of accessories and the purity can be confirmed by mineralogical methods. Traditionally X-ray diffraction method (XRD) has been used in bentonite research.

This study was focused to improve analysis methods, especially electron optics methods, developing the sample treatment. Fine grained sediment material bentonite dilutes to the water and thus makes it difficult to prepare good sample preparation. The use of the scanning electron microscope (SEM) in high vacuum requires solid samples for analysis. Electron microscope is useful in estimating the amounts of the accessory minerals of the sample, enabling also visual study of individual grains. By improving the sample preparation techniques was managed to prepare good sample preparations for electron microscope and electron microprobe (EPMA, electron micro probe analyzer) studies. EPMA was used to measure detailed the chemical composition of the material. By combining the different analysis methods we were able to find out the quality and composition as well as the amount of accessory minerals in the bentonite material. Also the chemical composition of the smectite mineral was analysed. The advantage of electron optic methods was emphasized in VTT's case study where bentonite material was sealed inside the copper canister for 15 years in aerobic and anaerobic environment.

Applications of X-ray tomography

A non-invasive method for measuring the three-dimensional displacement field and water content distribution in a wetting and swelling bentonite using X-ray tomographic imaging is developed. The detailed and well characterized experimental data obtained by the method is used in developing and validating models devoted to assessing bentonite buffer behaviour.

A non-invasive method for measuring the three-dimensional displacement field and water content distribution in a wetting and swelling clay using X-ray tomographic (CT) imaging is developed. The method is based on comparison of X-ray tomographic images of the clay sample in the reference state and in the wetted and deformed state. Deformation field can be measured for clays that contain sufficient amount of details visible in tomographic images so as to allow tracking by a 3D image correlation algorithm. The water content distribution can be determined from the different tomographic image between the wetted state and the reference state. The results of deformation analysis and of water content analysis based on the CT-method were compared with those from numerical solution for an axially compressed cylindrical rubber sample, and from gravimetric measurements of sliced samples, respectively. In both cases, the results given by the CT-method are in close agreement with those obtained by the reference methods. The method has been applied for monitoring the evolution of 3D deformation and water content distributions in cylindrical purified bentonite samples wetted in a constant volume (4D study). The measurements were carried out using a high-resolution microtomographic device (SkyScan 1172) and image voxel size 24 µm. The 3D wetting and deformation data obtained by the CT techniques will be organized in a data bank to be used in validating numerical models of moisture transport and hydromechanical behaviour of bentonite buffer during initial wetting phase.

Bentonite colloids

The University of Helsinki (Radiochemistry) has developed methods for bentonite colloid measurements and has studied sorption of radionuclides onto those colloids. Ionic strength and pH have a significant influence on the stability of bentonite colloids and radionuclide interaction with colloids and clay minerals, contributing to the colloid-mediated radionuclide, especially actinide, migration in environmentally relevant conditions for SNF repository.

The bentonite erosion resulting via gel phase in the formation of colloids may have a direct impact on the overall performance of the repository. The potential relevance of colloids for the increase in radionuclide transport is highly dependent on the formation of stable and mobile colloids in different chemical environments and their interaction with radionuclides.

In KOLORA subtask, chemical bentonite erosion, colloid formation and stability as well as radionuclide sorption on bentonite, Na-montmorillonite and bentonite colloids were investigated. The main tasks were to develop and test experimental arrangements, prepare materials and apply different characterization and determination methods. Bentonite erosion mechanisms, generation and stability of inorganic colloids were determined using MX-80 type bentonite powder and pellets. Solutions of different ionic strength and pH were Allard, low salinity granitic and OLSO, saline Olkiluoto reference groundwater and NaCl and CaCl2 electrolytes.

In batch type experiments, bentonite powder or pellets were placed in a sample tube where solution is added. The samples were stored with and without agitation and colloid generation and stability was followed as a function of time by analysing pH, particle size distribution, particle concentration and Zeta potential applying the photon correlation spectroscopy (PCS) method. Sr-85 and Eu-152 sorption was studied on powdered bentonite and bentonite colloids as a function of ionic strength and pH using the batch method to obtain information of the amount of the sorption. The macroscopic sorption behavior of Np(V)-237 on reference mineral corundum (α -Al_2O_3), Na-montmorillonite and bentonite colloids was studied by conducting experiments as a function of pH and neptunium concentration to get source of data for further studies using specific methods. The chemical nature of the complex between Np-237 and mineral surface was investigated by Zeta potential measurements and in situ ATR-FT IR using a flow cell and EXAFS spectroscopic experiments. The erosion and stability of bentonite colloids depended strongly on the ionic strength of the medium and the valence of the cation. The colloidal dispersion has remained stable in low salinity solutions so far for four years and noticeable colloid erosion occurred only in the most diluted solutions. The bentonite erosion was significantly increased with the slow agitation. Sr-85 and Eu-152 adsorption onto the bentonite colloids and montmorillonite was highly pH dependent, adsorption increasing with increasing pH. In situ ATR FT-IR spectroscopy was applied successfully the first time on montmorillonite and bentonite colloids to identify the dominating surface

reactions. Np(V) adsorbed onto montmorillonite and corundum as an inner-sphere complex though the bond is highly reversible.

Microstructure

In the topic, studies are carried out on the nanostructure of bentonite and purified montmorillonite at the University of Helsinki, Department of Physics. The main method was X-ray scattering (small-angle X-ray scattering SAXS, and wide-angle X-ray scattering WAXS/XRD), which is an excellent tool to get averaged information on the stacking and structure of clay platelets in samples of about one cubic millimetre in size. X-ray microtomography was also utilized in combination with SAXS (or XRD) in order to relate the macroscopic and microscopic structures and their orientation. The parameters to be determined in SAXS studies include the distribution of interlamellar distances and water layer thicknesses, the number of clay platelets in the stacks, and the fraction of delaminated discs. X-ray scattering intensities or density profiles of the clay stacks may be computed from the coordinate sets and compared to the experimental data. Combined in-situ SAXS and microtomography studies on drying or wetting clay compacted clay were done. Tactoid and microcrack reorientation along the compaction axis was observed in purified montmorillonite. This affects water mobility and is of interest from the point of view of buffer properties of bentonite. SAXS studies on clay colloids were carried out in co-operation with the University of Helsinki, Laboratory of Radiochemistry.

Modelling

Numerola Oy has applied their own mathematical methods, Numerrin, both to analyze 3D tomography data and to solve transport models based on that data. In Numerola a new numerical solver was implemented and validated for simulating thermal, hydrological and mechanical behavior of bentonite.

A new numerical THM-solver was implemented for simulating thermal, hydrological and mechanical behavior of bentonite. The mechanical part of the solver is based on a phenomenological model of Markku Kataja describing bentonite as an elastoplastic material with large deformations and moisture dependent mechanical parameters. The parametrization of the mechanical model is based on several wetting and swelling experiments and this parametrization works now for a wide range of water contents. The mechanical solver was tested by simulating to X-ray tomography experiments, and the comparison showed qualitative agreement of simulated and measured displacement field. The hydrological component of THM-solver describes moisture transport as diffusion of two moisture phases (water vapour and liquid water) and mass transfer between the phases. Comparison of simulations to X-ray tomography experiments yielded qualitative agreement with respect to simulated and measured water content profile. The new heat transfer solver is based on a heat conduction model with a moisture dependent heat conduction coefficient. The bentonite chemistry model developed in collaboration with VTT during KYT2010

was updated and corrected. This model simulates the evolution of sodium, calcium and chloride in the bentonite.

THC experiments and microbiology

The electrodes for measuring Na^+, Ca^{2+}, and SO_4^{2-} ions have been developed and tested. The stability of the electrodes can be improved by using liquid junctions between the membrane and conductor wire, instead of solid junctions. The liquid junction was tested for the Na electrode and the calibration results were promising. H+ sensitive IrOx electrodes were used to measure pH in compacted bentonite. The pH in the experiment with 0 P_{CO_2} remained nearly constant throughout the 5 month period. On the other hand, the pH dropped to near 6 with 0.3 P_{CO_2} and to 5.5 with 1 P_{CO_2}.

The cationic form affects many essential properties of montmorillonite. However, the cation exchange selectivities for montmorillonite have mostly been studied at room temperature. In this work, the cation-exchange selectivity coefficients and cation-exchange isotherms were determined in batch experiments for montmorillonite at three different temperatures (25, 50, and 75°C). The analysing accuracy of tested methods appeared to be an issue. However, any notable temperature dependence was not observed. The final reporting will be done after having the all analysis results by ICP-MS HR.

The dissolution study of Ca – and Na montmorillonite in groundwater simulants (2 g/L) indicated that the nature of the smectite mineral did not change over 140 days. However, the experimental conditions, more or less, modified the structure (e.g. the layer stacking of montmorillonite; the partial dissolution of the smectite). The partial dissolution could not be detected by XRD but was evidenced by chemical data, and can be considered as a possible contributor to the stacking faults of the montmorillonite. Depending on the experimental conditions, the logrates (mol g-1 s-1) for dissolution varied between -10.64 and -12.13.

Bacteria and fungal diversity studies were done of compacted bentonite experimental set up where bentonite had been compacted in the copper cylinders and maintained in both aerobic and anaerobic conditions for 15 years. A great diversity of microorganisms, also active microorganisms were detected. Sulphate reducers were as well present and they were more abundant in anaerobic experiment than in aerobic. SEM micrographs revealed the presence of fungi, which were later identified by molecular methods.

Conclusions

Our work is an example of a coordinated domestic project, which is, however, collaborating with several international projects such as EU BELBaR. Bentonite has appeared difficult to understand in the context of spent fuel disposal by the KBS3 method. However, we have obtained good results by applying several methods in studying the properties of montmorillonite. All these methods are directly applicable

in studies of the bentonite buffer in a large application area, such as from dry to saturated bentonite, from initial state to post glacial conditions, and from compacted bentonite to dilute colloidal solutions.

6.6 Corrosion of copper by water under oxygen free conditions (Project 16)

Jari Aromaa, Aalto University, School of Chemical Technology, Department of Materials Science and Engineering

Research topic and the main results

The objective of the study was to obtain definitive information on whether copper corrosion is possible in an oxygen-free ultrapure water by hydrogen evolution as cathodic reaction, and to continue to determine whether the reaction is possible in a simulated groundwater. Based on this data the corrosion likelihood and potential consequences on the final disposal were estimated.

The results showed evolution of hydrogen in the vacuum chamber, observed as an increase in the upper chamber pressure as well as increased hydrogen content of the palladium foil. The hydrogen concentration increase did not correlate with the observed corrosion of copper.

The quartz crystal microbalance (QCM) experiments suggest that in a closed system, where oxygen is consumed by the corrosion reactions, the corrosion rate is less than a tenth of the corrosion rate compared to aerobic conditions. The corrosion rates measured by the QCM correspond to the rates published in the groundwater measurements, 10–30 µm/year. This is about ten times more than measured in the bentonite. Under anaerobic conditions the copper corrosion is most likely not due to hydrogen evolution, but due to residual oxygen.

Significance of the results in nuclear waste management research

The aim of the study was that the results would help decision makers to evaluate better the corrosion resistance of copper in the final disposal. If hydrogen is not formed, the copper corrosion resistance evaluation criteria do not require any changes. If hydrogen is generated, the effect of hydrogen on the corrosion rate of copper shall be assessed. If hydrogen is formed in cathodic reactions of copper corrosion is evidently necessary to examine formation of the new, hitherto unidentified compounds of copper.

The results show that the copper is corroded under anoxic conditions, but the correlation between evolution of hydrogen and corrosion of copper has not been demonstrated. The connection of hydrogen evolution and copper corrosion was studied by Boman et al. and they have reported that there is no correlation (The 5th International Workshop on Long-term Prediction of Corrosion Damage in Nuclear Waste Systems, Asahikawa, Hokkaido, Japan, October 2013). On the other hand,

because the copper corrosion rate is greater during immersion in aerobic and anaerobic conditions and in the gas phase than in bentonite, studies of the corrosion rates in the initial stages of exposure are still necessary.

The research methods

Corrosive environments were ultra-pure water and synthetic saline and oxygen-free groundwater as described in Posiva Working Report 98-61.

The most important research system was the vacuum chamber, where it was possible to measure the evolution of hydrogen by means of pressure change. In the corrosive environments used in this work, the sole source of hydrogen is hydrogen generation during copper corrosion. The test apparatus consisted of two chambers, separated by a palladium foil allowing hydrogen permeation only. Both chambers were equipped with connectors and pressure sensors to create vacuum and pressure measuring during the tests. Copper corrosion reactions took place in the lower chamber, which was nitrogen purged and remaining oxygen removed by vacuum. The upper chamber was evacuated, and the pressure in the chamber increases were attributed to corrosion reactions as hydrogen passes through the palladium membrane. The lower chamber was built in two versions, the first was made of stainless steel and the copper sample was within a glass vessel, and the second was made of copper with potential measuring electrodes.

Another method used in the study was a quartz crystal microbalance, which enables the measurement of copper corrosion by continuous change in weight. The method was used in determination of corrosion rates and monitoring their changes in nitrogen purged solutions.

Furthermore, the hydrogen content of palladium foils was determined and the corrosion was estimated by changes in solution pH, conductivity, redox potential and changes in the concentrations of copper.

6.7 Mechanical strength of copper canister (Project 17)

Hannu Hänninen, Aalto University, School of Engineering, Department of Engineering Design and Production

Introduction

Copper canister will have a non-homogeneous microstructure and it will contain discontinuities such as small defects which both cause stress concentrations in the canister structure. These discontinuities and regions affect markedly the deformation mechanisms of the copper canister during the long operation time and all these effects have to be known in detail to predict the safe lifetime of the canister. One of the most important factors is the variation of the grain structure (size and texture) both in the weldments and in the different parts of the canister. The other

important factor is the variation of the degree of deformation in the different parts of the canister and between the surface layer and the interior of the canister wall (effects of machining) and in the weldments, which results in residual stresses and deformation localization during long-term exposure. Deformation localization is important both in creep and environmentally enhanced cracking, stress corrosion cracking (SCC).

Research objective and the main results

The aim of the project was to clarify the mechanical properties of the different parts (base metals and welds) of the copper canisters and to understand the mechanisms of macroscopic and microscopic plastic deformation in its non-homogeneous structure. The project addressed topics mainly for the field long-term structural integrity of the copper canister as well as manufacturing technologies of the copper canisters and the necessary requirements for the manufacturing and closure welding of the canisters. The following topics were examined:

- Properties and structures of the FSW weldments of copper canisters;
- Effects of discontinuities and weld defects on mechanical properties of copper canisters;
- Deformation localization in the copper canisters in various loading situations;
- Hydrogen effects on mechanical and creep properties of copper;
- Stress corrosion cracking mechanism of copper based on vacancy accumulation in copper due to oxidation in the final disposal conditions.

Effect of the results on nuclear waste disposal and other related research

In short term, the results are exploited by the ongoing creep and corrosion research, which may not be able to give reliable results without mechanistic understanding of the controlling mechanisms of the phenomena. The plastic deformation mechanisms of the non-homogeneous copper material (weldments and discontinuities) have to be understood in order to understand the deformation and creep behavior. With mechanistic understanding of the role of various discontinuities in grain and deformation structures guarantees the reliable prediction of long-term integrity of the copper canisters and gives the necessary data for the modeling of the canister deformation and creep during the final disposal. The research is extremely important for definition of the quality requirements for the canister manufacturing, analysis of the criticality of the discontinuities (allowable defects and their sizes as well as their acceptance criteria) for safety analysis of the canister and the whole final disposal process. The role of hydrogen absorption in copper and its effects on mechanical and creep properties is at the moment very little understood subject affecting the safety of copper canisters and needs a careful study. This is done in joint action with KTH and KIMAB, Sweden. Exclusion of the possibility of stress corrosion cracking of copper canisters in the safety analysis must be based on the mechanistic understanding of

the phenomenon and not only on short-term laboratory test results as the present situation is.

Experimental methods

The Laboratory of Engineering Materials of Aalto University has obtained modern friction stir welding machine for making the necessary specimens for the project using the own laboratory equipment and getting the experience on weldability of copper canister materials (forged, pierce & drawn and extruded copper). In the welding experiments also the common defects are fabricated for the structural integrity studies. On the same time the experience is gained how to avoid typical weld defects forming in the FSW welds of thick-wall copper canisters. Optical strain measurement 3D-system based on digital image correlation (DIC) has been taken in use for analyzing the macroscopic deformation of the copper welds. A new surface pattern was developed to increase the resolution of the deformation localization measurements to the level of 1 µm and also the possibility to study the deformation mechanisms and deformation localization at elevated temperatures is now possible. Laboratory of Engineering Materials has complete capabilities for the residual stress measurements of copper canister (X-ray diffraction, center hole drilling, ring-core and contour methods). For small specimens an in-situ mechanical test system is built which allows the measurement of local deformation by using digital image correlation in non-homogeneous microstructures, where the deformation localizes to certain regions such as process zones (fine grain size and oxide particle stringers in FSW welds). Two modern FE-SEM/EBSD/EDS equipment systems allow the highest resolution orientation microscopy and extremely high-resolution residual strain distribution determination of the critical regions of the copper canisters. Combining of the macroscopic and microscopic deformation mechanisms in the non-homogeneous structures of the copper canister allows the understanding of the deformation mechanisms of the different regions of the canister. Based on these results the long-term deformation of the whole copper canister can be predicted and modeled reliably.

Recently copper corrosion in anoxic conditions in pure water has been claimed to take place by hydrogen evolution. Part of this hydrogen is expected to absorb in copper. However, the most important source of hydrogen in copper is the sulfide corrosion which is the main form of copper corrosion during the long-term deposition. Hydrogen uptake is expected to occur as a result of corrosion and therefore hydrogen effects on the mechanical properties (ductility/toughness and creep properties) of copper have to be known. Effects of hydrogen on copper are not yet understood very well and research results are scarce. In this study hydrogen diffusion and trapping as well as hydrogen-induced void formation in copper and hydrogen effects on deformation mechanisms and localization as well as creep of copper were explored. Hydrogen behavior in copper is also modeled by modern *ab-initio* modeling methods taking into account the hydrogen interactions with impurity atoms (S, P, O, Ag, Ni)

and vacancies in copper. As a result of these studies the effects of hydrogen absorbed in copper on mechanical and creep properties of copper in the conditions of final deposition are understood. In 2014 co-operation was started with KTH to study hydrogen absorption in copper in corrosion experiments performed under gamma radiation which simulates the real conditions of the copper canister prevailing under final disposal under first 1000 years time period.

6.8 Material Integrity of Welded Copper Overpack, MICO (Project 18)

Juhani Rantala, VTT

The research project has concentrated in determining the mechanical strength and ductility of canister copper and its weld. The work has involved a remarkable amount of experimental creep testing by using various methods like uniaxial creep testing, notched bars and multi-axial tests on CT specimens. The methodology in uniaxial creep testing has been to use acceleration by increasing only temperature, which is a conservative method and therefore more reliable in life assessment. On the basis of in-house and public data a LCSP creep model was developed as a result of a PhD thesis. The model has been used in FE analysis for calculating the stresses and strains and their time history in the canister. The FE analysis has shown that the stresses in the most stressed parts of the canister will relax quickly and therefore relaxation testing of copper has been started in order to develop a relaxation model. The relaxation testing is still ongoing. Both pneumatic bellows rig and a servo-hydraulic testing machine have been used, but in the future the testing will be done in a servo-mechanical machine, which is more stable. The FE analysis of the canister will be repeated when the relaxation model has been developed and verified. In the analysis also defects will be modelled to represent the joint line hooking and the oxide particle zone.

The purpose of the multi-axial tests has been to clarify the effect of multi-axiality on the life of copper. The VTT view is that multi-axiality will shorten life. Tests with CT-specimens have been conducted for both FSW and EB welds. During the experimental testing also optical strain measurement has been tested, which can be used for verification of strain predictions produced by the FE analysis. Microscopy (LOM, SEM, EBSD, TEM) has been done as necessary and mostly for the multi-axial samples. Most of the SEM and EBSD microscopy has been done in collaboration with Aalto University.

The interaction of creep and corrosion by the ground water has also been tested, but no interaction was observed in those parts of the CT specimens which have the highest mechanical stress. In other parts of the specimens pitting corrosion was observed, but this was interpreted to be a result of galvanic coupling. These tests

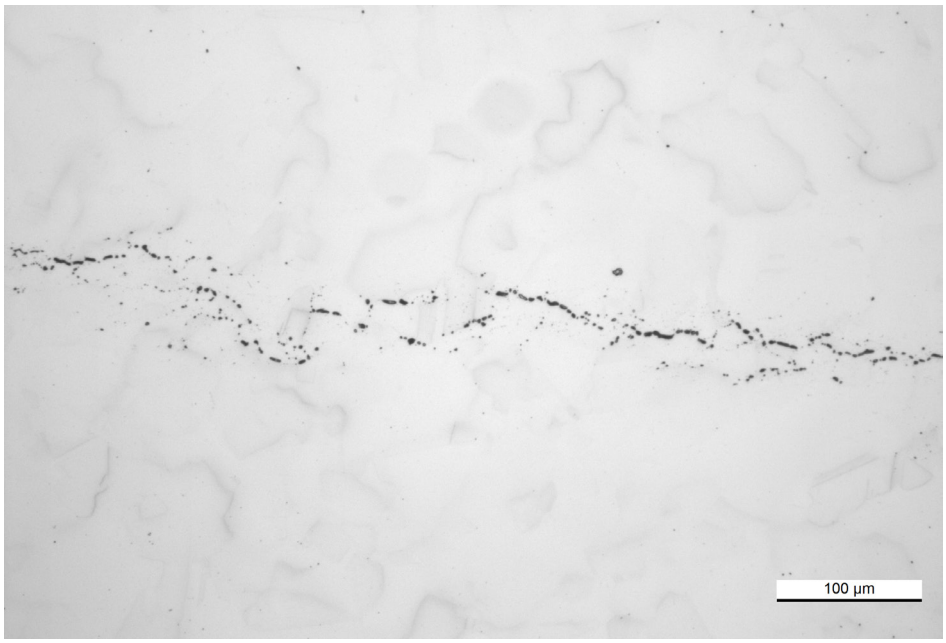
have been proposed to be continued in the next research programme but by avoiding the galvanic coupling.

Also creep brittle pure copper (OF) has been used as a reference material in order to find out whether it would be possible that the ductile phosphorous doped copper (OFP) could become brittle at long times. So far on the basis of an analysis made on the basis of literature data it seems the rupture ductility of OFP copper reduces only little at long times, but there is a lot of scatter.

A remarkable result after the testing of a CT specimen for 50000 hours was that when the notch of the specimen was the natural notch formed next to the FSW weld, the oxide particle zone inside the weld about 8 mm ahead of the notch had cracked (Figure) without the weld material immediately ahead of the notch being cracked although a lot of creep cavities was observed ahead of the notch tip. The stress level in the oxide particle zone was only about 13% of the stress ahead of the notch tip. The result is remarkable because it shows that the oxide particle zone is prone to cracking. The tested lid had been welded at a time when the welding was carried out in air. The present procedure is to do the welding in inert gas, which probably will strongly reduce the amount of oxide particles. The cracking of these welds needs to be tested.

The LCSP creep model has been used for life assessment of the canister by assuming for simplicity that the stress level experienced by the material would remain constant for 100000 years and that the bentonite would remain dry. By these assumptions the stress level should be 152 MPa in order for the material to fracture at the end of the chosen time period. By using a more realistic assumption that the bentonite will be wetted and therefore the temperature of the canister would drop, then at 152 MPa the life of copper would be 250000 years. In reality a stress this high would relax quickly and the life would increase remarkably. However, with many other materials relaxation can lead to premature fracture at relatively small strains, but there is no experimental data about this for copper.

Figure 7. Cracking in the oxide particle zone in a CT specimen after 50000 hours at the temperature of 175°C.



6.9 Sulphide-induced embrittlement of CuOFP, CUHA (Project 19)

Timo Saario, VTT

Disposal of spent nuclear fuel is based on the multiple barrier concept of which the copper canister forms a significant part. In 2007 a Japanese research team showed that sulphide can cause stress corrosion cracking of pure copper in oxygen-free water. In KYT 2010 research program a sub-project called "Sulphide induced stress corrosion cracking of copper" yielded a result referring that sulphide containing groundwater can cause sulphide induced embrittlement in copper. CUHA project was established to further evaluate the risk involved with this phenomenon.

Oxygen and sulphide are the most important species that can cause copper corrosion in the disposal conditions. The role of sulphide in terms of copper corrosion is increasing after the oxygen is depleted in various reactions, thus making sulphide the only species that is capable of oxidizing copper. In addition, the sulphide will not be consumed like oxygen but sulphide is present in low concentration in the repository throughout the disposal process. The results of this study can be used to evaluate the risk of sulphide-induced copper embrittlement in disposal conditions. This research has been a continuation of the previously conducted research on

sulphide induced corrosion on copper in the KYT-project "Sulphide induced stress corrosion cracking of copper".

The obtained main results include:

1. The five week long exposure experiments in Olkiluoto-type artificial groundwater with sulphide additions of 200 mg/l in room temperature yielded a small (about 12%) reduction in yield strength and fracture strain of copper. The sulphide exposure had virtually no effect on the ultimate tensile strength.
2. The creep test results showed that the difference between exposed (exposure time of five weeks in artificial groundwater with 200 mg/l sulphide) and reference samples in creep fracture strain increased while stress level decreased (and creep testing time increased).
3. No signs of sulphur ingress was found on the fracture surfaces of tensile and creep samples.
4. There was no indication of stress corrosion cracking of OFP copper according to the results obtained from in situ tests (slow strain rate tests, SSRT) in Olkiluoto-type artificial groundwater with sulphide addition of 200 mg/l at room temperature.

Figure 8. The effect of sulphide concentration on mechanical properties of OFP copper. Five week exposure time at room temperature. Abbreviations displayed in figure include: FS = fracture strain, YS = yield strength and UTS = ultimate tensile strength.

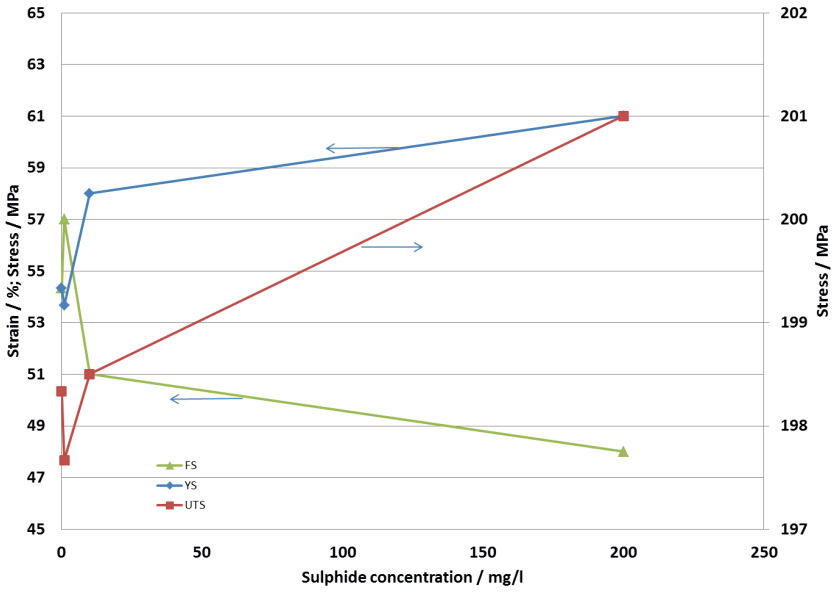
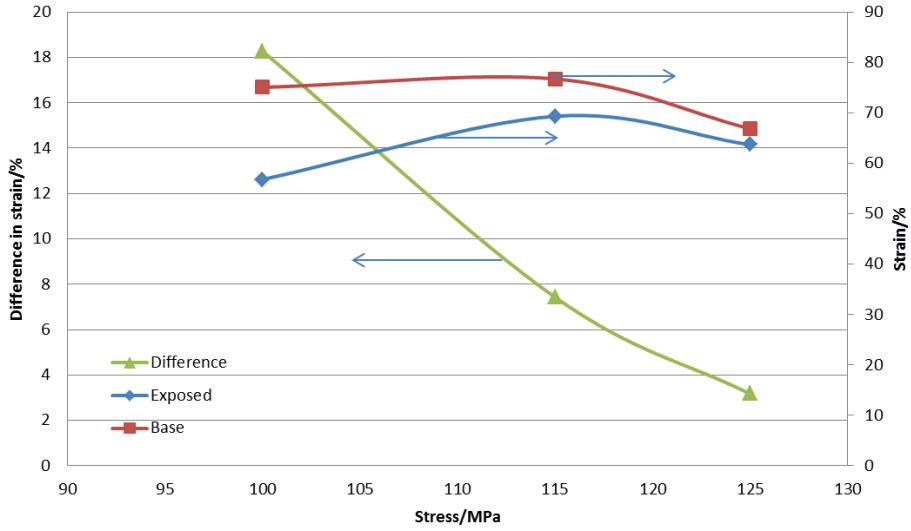


Figure 9. The effect of five week pre-exposure in 200 mg/l sulphide containing artificial ground water on creep fracture strain of OFP copper on different stress levels at T = 215°C. Red and blue lines indicate base and exposed samples, respectively. Green line indicates the difference in fracture strain between base and exposed samples.



Several experimental setups and analytical methods were used during the research. Autoclaves used during the exposure of the samples enabled precise controlling and monitoring of the water chemistry. SSRT experiments were conducted as in situ in the same autoclaves. Creep and tensile testing were conducted as ex situ. Tensile and creep tests were conducted according to the standards SFS-EN ISO 6892-1 and SFS-EN ISO 204, respectively. Samples were afterwards analysed by using SEM and EDS analyses, as well as an optical microscope.

6.10 Corrosion of copper canister (Projects 20–21)

Corrosion monitoring under disposal vault conditions (Project 20)

Timo Saario, VTT

Corrosion rate of technical barriers for activity release from spent fuel storage has to be known with sufficient accuracy. This research work consisted of a literature study focusing on potential on-line in situ measurement techniques for corrosion rate. The results were utilised in the project "Effects of microbial activity on different forms of copper corrosion", in which the on-line in situ ER-technique was used to monitor the corrosion rate of copper.

Effect of microbial action on different corrosion processes of the copper canister, MICCU (Project 21)

Leena Carpén, Pauliina Rajala, Malin Bomberg, VTT

Background

The disposal of high-level radioactive waste is based on the usage of multiple barriers. These barriers, from the used nuclear fuel, are the disposal canister, bentonite buffer, backfill materials in the tunnels and lastly the surrounding bedrock. The disposal canister plays a major part of these barriers. The copper canisters will be placed in the bedrock, at the depth of 400–450 meters and they should have lifetimes exceeding 100 000 years to prevent the release of radioactive nuclides to the surrounding environment.

The failure mechanisms of copper canister have been considered by means of models developed based on literature, of corrosion tests simulating the disposal conditions and of copper items found in nature. However, there are only few studies done in the presence of microbes and none in Finland. The activity of microbes attached to surfaces and the properties of formed biofilms are essential factors when considering the possibility of microbiologically influenced corrosion (MIC). Under the biofilm the conditions may differ remarkably from the surrounding solution and thus induce circumstances where corrosion is locally increased. In addition to the microbes attached to the surface of copper, also the microbiological activity in bentonite or in its vicinity producing the corrosive ingredients for copper, like acetate, nitrogen compounds or sulfides, can change the conditions and thus substantially accelerate the corrosion. These corrosive products may migrate to the vicinity of the copper canister especially when the bentonite buffer is unevenly swelled or compacted.

The presence of corrosive components in the disposal environments has been aimed to predict with so called MIC model (CCM-MIC). However, this model has not been tested or verified experimentally. Taking into account all the uncertainties

commonly connected to the understanding of the microbial behaviour the verification of the model is quite required.

The results of this study can be utilized in the long term copper durability focus area by evaluating the corrosion rates of copper as well as the validity of the developed models.

Aims

1. Design and construction of experimental arrangement to study microbially induced corrosion
2. To estimate the reliability of MIC model (CCM-MIC)
3. To evaluate the effects of microbial activity to the corrosion rates and mechanisms of copper in Finnish disposal environment
4. To find out what microbes will attach to the copper surface and the properties of the biofilms

Results

The goal of the project was to evaluate the possible effects of microbiological activity on the corrosion behaviour of copper canister in Finnish disposal environment. Experimental arrangement to study microbially induced corrosion enabling on-line electrochemical measurements was designed and constructed. The testing arrangements were constructed so that the testing environment corresponds as close as possible to the real disposal conditions. Microbes were seen to be attached to the surface of copper forming biofilms in biotic environment. Also the corrosion rates calculated from the electrochemical measurements showed higher values in biotic environment compared to the abiotic. According to the redox measurements the conditions in biotic environment were reducing where as in the abiotic the environment seemed to be slightly oxidizing. Rod-shaped bacteria and iron sulphide, FeS were detected on the copper surface after the exposure in the biotic environment. In abiotic environment copper (I) oxide, Cu_2O had formed on the copper surface.

In a thesis work nitrogen metabolisms at the disposal circumstance was studied to be able to experimentally evaluate the CCM-MIC model. Nitrogen compounds, ammonium and nitrate clearly increased the growth of microbes. The minor amount of these compounds in the ground water can be the restrictive factor for the growth of microbial communities.

Summary

The test arrangements and the enrichment as well as the inoculation were successful. Microbes attached and formed biofilm on the copper surface in biotic environment. The corrosion rates calculated from electrochemical methods were higher than those in abiotic environment. The native microbes in groundwater are able to produce several components potential to cause stress corrosion cracking to copper. In addition, these compounds can be utilized by other microbial groups.

6.11 Durability of engineered concrete barriers under final disposal conditions – Subproject 1 (Project 22)

Jari Puttonen, Aalto University

Research theme and main findings

The technical barriers made of concrete are required to be serviceable much a longer period of time than we have experiences from existing concrete structures. The service life requirement also exceeds the time period covered by design standards. As the concrete structures undergo complex and interacted physiochemical phenomena during their ageing, the service life estimation has to be based on recognised ageing phenomena and their modelling and simulation.

As a main result of the study, a general thermodynamic model was introduced for managing the ageing phenomena of concrete structures in disposal conditions. The model was verified with long-term experimental results. The thermodynamic model considers ion-ion and ion-cement hydrate interactions as well as the properties of the gas phase in multi-ionic transport in concrete. A statistical methodology was presented for evaluating the effect of uncertainties on the results.

The model was applied to the simulation of the long-term ageing of concrete structures in the facilities of low- and intermediate-level nuclear waste. The different sets of environmental conditions during the lifetime of the disposal facilities were considered in the calculations. The results showed that the concrete studied performed satisfactorily during the simulated period of 500 years, but the corrosion of the steel reinforcement cannot totally be excluded. The research also pointed out the limits of traditional diffusion models and accelerated tests in evaluating the ageing of concrete. The importance to develop an ability to define more accurately the time needed for the initiation of the steel reinforcement corrosion was also revealed by the research. This requires a more targeted research in the future. The thermodynamic simulation method also makes it possible to observe and understand the latent factors involved in the deterioration of the concrete, which makes it possible to influence the selection of materials in the design phase or to find proper corrective measures. The method can be applied to different concrete mixes by defining their case-specific initial values.

Importance of the results for the research of nuclear waste management

The results improve an existing knowledge about the long-term durability of concrete in nuclear waste facilities and also increase capability to estimate more realistically the service life of concrete structures in disposal conditions. The results of the study can directly be exploited in the ageing management of concrete structures in Finnish and comparable disposal concepts. Generally, the results can be applied

to reinforced concrete structures in nuclear power plants and in other applications of civil engineering. Therefore, the results can be directly exploited, for example, in the projects of SAFIR -programme (Safety of nuclear power plants – Finnish national research programme), where the concept of service life management of concrete structures in nuclear power plants is studied. Finnish Radiation and Nuclear Safety Authority (STUK) can also use the results for the evaluation of disposal concepts. As the production of theses has been integrated into the research, the project has been supported the education of new experts to the field.

Research methods

A numerical solution of the model was carried out with a commonly available software. Experimental research was used for verifying the model. The experiments performed for the concrete specimens under simulated disposal conditions in a joint research programme by TVO and Fortum Power and Heat Oy, launched in 1997, were exploited in the evaluation of the model. A number of versatile laboratory analyses were carried out for the specimens in Aalto University and VTT (Subproject 2). In addition, other experiments for the needs of the model were performed in Aalto University.

6.12 Durability of engineered concrete barriers under final disposal conditions – Subproject 2 (Project 23)

Eila Lehmus, VTT

Scope and objective

The 500 year service life requirement for the technical barriers in nuclear waste repositories represents a significant challenge to concrete engineering. The required service life is much longer than present experience with reinforced concrete structures. Reinforced concrete structures are now and probably also in the future much used in the technical barriers of final repositories for low and middle active nuclear wastes as well as for activated reactor pressure vessels and the internal parts of reactors.

The objective of the BetKYT project – Durability of engineered concrete barriers under final disposal conditions, was defined as “elevating the level of knowledge on the durability of concrete barriers in final disposal conditions, i.e. below the ground water, and to acquire data for modelling the performance of concrete over time. Based on these results the aim is to develop methodologies for evaluating the service life of such barriers.”

Project

An important step toward the development of concrete that performs well in aggressive underground environments is the understanding of the deterioration

mechanisms of concrete by these aggressive media. This knowledge will lead to an increase of the service life of the concrete structure. The study of concrete that has been reacted under controlled laboratory conditions is the foundation to understand the sequence of mineralogical reactions that occur and relate the findings to exposure conditions and concrete mix design definition.

Nine aggressive water solutions, as well as clean tap water, were prepared for the immersion of concrete specimens. The compositions of the solutions were chosen in order to simulate varying levels of ground water aggressiveness. The chemicals and concentrations of test solutions are presented in the following table.

Table. Aggressive chemical compounds added to solutions.

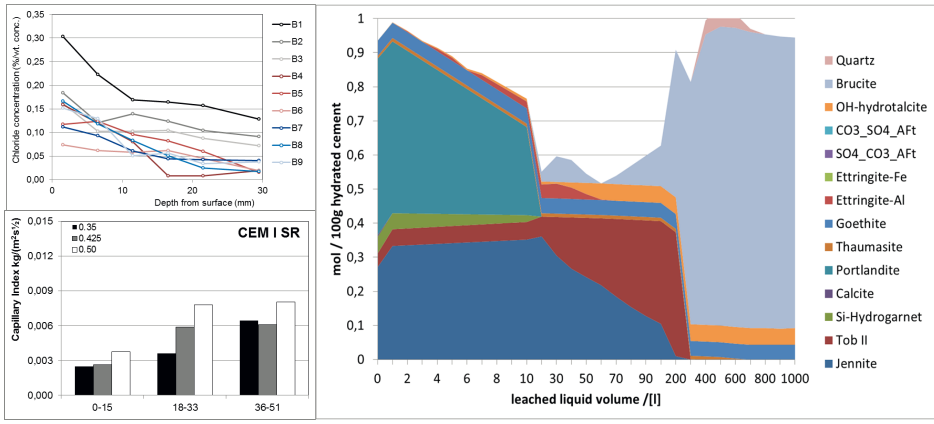
Solutions	Chemical	Aggressiive component (mg/l)
L1		SO_4 - 20
L2	Na_2SO_4	SO_4 - 500
L3		SO_4 - 1000
L4		Cl - 50
L5	NaCl	Cl - 1000
L6		Cl - 10000
L7		SO_4 – 20; Cl – 50; Mg -5
L8	Na_2SO_4 + NaCl + + $MgCl_2 \cdot 6H_2O$	SO_4 – 500; Cl – 1000; Mg -100
L9		SO_4 – 1000; Cl – 10000; Mg -300

Concrete samples were stored in these solutions for approximately 13 years after which samples were taken for analysis. Testing focused on three specific aspects: i) Chemical changes in concretes that were quantified by measuring the chemical profiles (chloride, sulphate, sulphur, and magnesium) and the element distributions using SEM-EDS; ii) Changes in mineralogy and hydration of the concretes using TG/DTA and X-ray diffraction; iii) Changes in microstructure measured using electron microscopy, and in porosity based on capillary water uptake tests.

Results and analysis

An analysis of the concrete mix designs used in this study shows that almost all concrete mixes are, based on current standard requirements, able to fulfil a 100 years' service life for the most aggressive of the solutions used (L9). The requirements for concrete durability are based on the chemical attack to the cement hydrates, and therefore do not take into consideration the effects of chloride ingress and carbonation, and the influence of these on reinforcement corrosion. The choice of binder type for the concrete mix design is strongly influenced by the aggressive loading and types of deterioration the concrete will be subjected to. The sequence and variation of exposure conditions are also of importance as the characteristics of concrete can be changed due to the interaction with the environment.

Figure 10. (a) Chloride profiles for all concretes in solution L6; (b) Average capillary index for CEM I SR concretes in solutions L2 and L3; and (c) Geochemical modelling of leaching of concrete in CEM I SR in solution L9.



The results of the extensive testing program, despite focusing on different aspects of the concrete performance/interaction with the solutions, corroborate each other. The chemical profiles measured show that, disregarding the surface layer in contact with the solution, the penetration of magnesium and sulphate into concrete has not occurred. Estimated initial content values are still observed from the profiles. In the case of chloride ingress, we observe greater penetrations for concretes made with CEM I binder (up to 30 mm considering a critical chloride content of 0.07% by weight of binder), than the other binder mixes (up to 10-15 mm considering a critical chloride content of 0.07% by weight of binder). A conservative critical chloride content value has been chosen. Very little information is available on the values that would initiate reinforcement corrosion, especially in submerged aggressive solutions. Total porosity and capillary absorption results observed are in the range expected for the concrete qualities in question. These results also confirm, indirectly, that no deterioration is occurring in the concretes studied. The results have been further analysed by chloride ingress modelling and using a holistic approach by geochemical modelling. Chloride ingress modelling, although based on limited data, shows that taking into account the designed service life CEM I type concretes are not recommended for environments in which there are chlorides with the risk of reinforcement corrosion. Geochemical modelling confirms that the concretes are at a very early age of deterioration when considering leaching and expansive reactions. It shows that equilibrium with the solution has not been reached. Leaching has not propagated for long enough to allow the precipitation of Aft phases. In addition, thermodynamic equilibrium predicts the presence of brucite in concretes of the binder type S3, but according to the test results the samples did not contain brucite.

As a summary, the concrete with good quality exposed to low-intermediate aggressive simulated ground water for 11-13 years show no signs of deterioration.

6.13 Mechanical Properties of Rock Joints, KARMO I (Project 25)

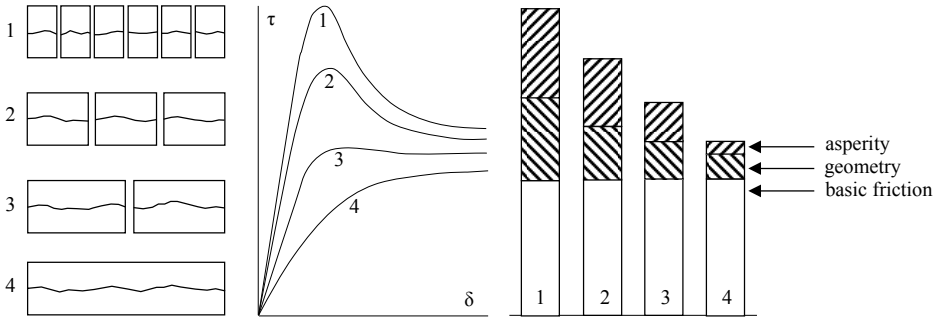
Lauri Uotinen, Aalto University, Department of Civil and Environmental Engineering

KARMO I research project was conducted during the last year of the KYT2014 research programme. It functioned as a preparatory pilot research for the later KARMO II (2015-2016) and KARMO III (2017-2018) research projects. The KARMO research continuum aims to develop a method to determine the mechanical properties of rock joints for numerical modelling purposes using laboratory scale replica series. KARMO I (2014) research project aimed to develop the photogrammetric method to record the rock joint geometry, to digitize the rock joint into a printable mold, to print the molds and cast the concrete surface replicas and test the replicas and derive from the results how the components of the shear strength change with scale.

Research Topic

The mechanical properties of rock joints depend on the testing scale (Figure). Laboratory samples are 50…200 mm long and the naturally occurring joints may be up to tens of meters long. Natural, undisturbed rock joints are hard or impossible to test, so the goal is to develop a method how small scale joint replica shear tests can be used to derive the mechanical parameters of natural joints for numerical modelling. Modelling is used for example to estimate the displacement potential of rock mass in the spent nuclear fuel repositories.

Figure 11. Scale (left) effect to shear strength (middle) three components (right) asperity, geometry and basic friction. Modified from Bandis (1990) and Barton & Bandis (1990).



Research Methods

The research began with a literature survey on photogrammetry. The discovery of suitable parameters for the fast prototypers was done empirically starting from the simplest possible method and progressing iteratively towards the desired quality

level. The point cloud was converted to a printable 3D model using photogrammetric mathematical software and programming. The concrete casting recipe and method were developed empirically. The shear box testing was carried out in the Rock Mechanics laboratory of Aalto University. The data was analyzed using stochastic methods.

Results

Subproject 1 – Replicating surface geometry using 3D technology

Two fast prototyper extrusion printers Printrbot Simple v4 and Ultimaker Original v3 were obtained. Raphaël Yorke assembled and tested the machines. The Bachelor's Thesis studies the theoretical basis of the photogrammetric method using a literature survey and it describes the photogrammetric method developed during the research. The thesis contains an overview of the working principles of the fast prototypers and how they may be utilized as a part of the manufacturing process. A rock joint was captured and replicated into a printable 3D model.

Subproject 2 – Shear box testing of the surfaces and analysis of the results

Eero Korpi conducted his Bachelor's Thesis research in 2012 about the Shear Box Test for Rock Joints. In spring 2014 he continued the research by obtaining and splitting a rock boulder and by piloting the 3D manufacturing method. The thesis describes the work from obtaining the large sample to testing the replicas in the laboratory. A survey to the latest rock joint research is presented, mechanisms how the scale effect works are described, sampling plan, digital post-processing and the manufacture into replica molds are presented. The study ends in analysis of the pilot test results, discussion and conclusions. Research assistants Antoni Kopaly and Daniil Iakovlev casted the research series and performed the shear box testing and documented the results for the third subproject.

Subproject 3 – Analysis of Scale Effect, Geometry and Asperity

Doctoral Student Ari Hartikainen developed an algorithm to transform the measured point cloud into a printable 3D replica mold. Doctoral Student Lauri Uotinen analyzed the preliminary results of subprojects 1 & 2. The results will be published in ISRM2015 congress 10.–13.5.2015 in Montréal, Canada.

The raw data from the replica series became available midway December 2014 and only two of the results are supporting the initial assumptions. The rest of the shear tests show no clear peak strength and the sample pairs compress vertically in the beginning of the tests. It seems that the observed behavior may be caused by too much imprecision with the matedness of the surfaces. The problem may be caused by the photogrammetric recording process, manufacturing process or it can be an error in the testing equipment or in the testing method. The issue will be resolved

during the KARMO II continuation research in more detailed analyses. The most significant findings of KARMO I study will be reported as a scientific article entitled *"A method to downscale joint surface roughness and to create replica series using 3D printed molds"*.

Significance and Utilization of the Results

KARMO I research project demonstrated that the proposed photogrammetric replication process can produce surface replicas for the shear box test. The pilot tests and two tests from the main series are in line with the initial assumptions and produced the expected scale effect, but further studies are needed to increase the reliability of the proposed method. The initial results are most important for the KARMO II continuation study. The photogrammetric replication process is independent of the use purpose and it may be used to capture any surface geometry irrespective of size.

The results of the KARMO research continuum are published for researchers and officials, and they may be used to evaluate the suitability of different crack acceptance criterions or the acceptance of joint displacement analysis results. The majority of the results will be used as initial data for numerical modelling when analyzing the mechanical response or displacement of rock joints.

Research Group

Prof. Mikael Rinne (Rock Mechanics) acted as the Project Manager. Raphaël Yorke carried out the first subproject Bachelor's Thesis. Juha Antikainen directed and Eero Korpi carried out the Master's Thesis continuing his Bachelor's Thesis from 2012. Ari Hartikainen developed a method to transform the captured joint surface into a printable 3D solid and Antoni Kopaly and Daniil Iakovlev casted and tested the replica series. Lauri Uotinen acted as the Coordinator and carried out the analyses of the results and acted as the main author in the conference paper and in the scientific article.

6.14 C-14 release under disposal conditions, Hiili-14 (Project 26)

Kaija Ollila, VTT

The final objective of this project is to investigate the release of ^{14}C from activated steel under disposal conditions in the cement-based repository. The speciation of ^{14}C in both aqueous and gaseous phase is important for safety analyses. Especially the proportion of organic species is important due to its high mobility. These studies are a part of the project CArbon-14 Source Term (CAST, WP Steels) under the Seventh Framework Programme of the European Atomic Energy Community (EURATOM).

The studies were initiated with leaching experiments with inactive steel materials as solid phases, in which the release of ^{12}C is measured. The form of ^{14}C in irradiated steels is not known. If the nitrogen is originally present as nitride then the resulting ^{14}C may be present as carbide (and/or carbonitride). The first inactive materials were Loviisa steel, which is identical with the steel components of the core of the Loviisa reactor, and Japanese high carbon steel. The carbon contents were 0.08 % and 1.2%, respectively. The analyses with optical microscopy and SEM/EDS showed that Loviisa steel has its carbon mainly as interstitial atoms in the austenitic steel lattice (Figure). No Fe3C was observed. The main form of carbon is carbide in High carbon steel. These two materials were sawed to small fragments with a band saw and purified afterwards with acetone and isopropanol.

Simulated groundwaters were chosen as the leaching solutions for the experiments. The compositions of the simulated groundwaters were based on an analysed groundwater sample from Loviisa site (LPVA5). The compositions were adjusted with the help of an equilibrium model (EQ3) to be stable under Ar atmosphere (O_2 less than 1 ppm and low CO_2 content). Elements with low concentrations and disturbing elements (e.g. redox sensitive elements, Mg, Si) were excluded. The recipes of two simulated groundwaters, pH 8.5 and 12.5 (both with and without HCO3-) were modelled (Table). The alkaline pH simulates the effects of cement in the repository conditions. The pH of 8.5 is used as a reference.

The batch experiments were conducted in polypropylene vials in the glove box (Ar). The ratio of the mass of the solid phase to the volume of the leaching solution was varied (50 g/100 mL, 25 g/100 mL and 15 g/100 mL). Samples were taken from the leachants after sequential leaching periods for the analyses of the total contents of dissolved organic (DOC) and inorganic carbon (DIC). The analyser (Analytic Jena) first measures the total content of carbon (TC). Subsequently, the inorganic carbon is changed to CO_2 by acidification and bubbling, and brought to the reactor, from which the CO_2 is led to the NDIR detector and measured (DIC). The organic carbon is calculated from the difference between TC and DIC. Measurements for pH and redox (E_h) were performed in selected samplings.

Figure 12. SEM picture of the surface and microstructure of Loviisa steel.

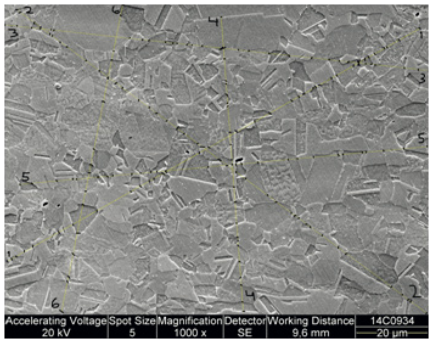
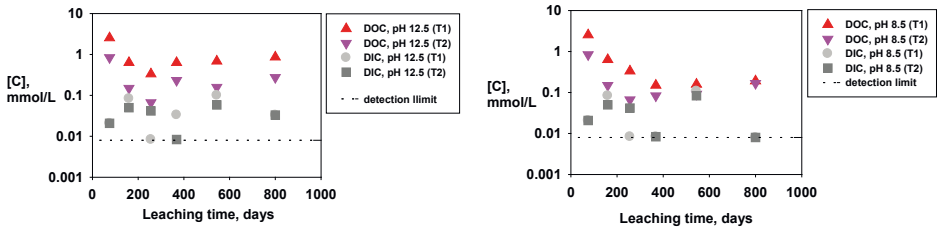


Table. Compositions of the simulated groundwaters. Units in mg/L.

	CA85	CA125	HC85	HC125
pH	8.5	12.5	8.5	12.5
Na^+	2680	2200	3440	3440
Ca^{2+}	630	500	-	-
K^+	25	25	25	25
HCO_{3^-}	-	-	104	104
$SO4^{2-}$	560	560	560	560
Cl^-	4840	4037	4840	4840
Br^-	18	18	18	18

Figure 13. Measured contents for dissolved inorganic (DIC) and organic (DOC) carbon in simulated groundwater in the leaching experiments of crushed Loviisa steel (left) and crushed High carbon steel (right). T1 represents the tests with m/V=50 g/100 mL and T2 the tests with m/V=25 g/100 mL.



Relatively high concentrations of organic carbon (DOC) were measured, especially at the early stage of the experiments. The aqueous phases were replaced with fresh simulated groundwater after the first sampling. The concentrations decreased afterwards. Possible contamination of the solid phase by machine oil during the grinding process or inadequate purification, and their effects on the results could not be excluded. Additionally, separate tests revealed that some contamination may originate from the filter material in the first sampling. The filter material was changed after that. Generally, higher contents for DOC compared with DIC were measured in the leaching solutions with both steel materials (Figure). The DOC contents increased slightly with pH. Lower contents for DOC were measured in the experiments with High carbon steel.

Because of possible contamination phenomena, new leaching experiments with new carefully characterized steel and iron powders were planned. A better crushing method, which would not contaminate steel material, was not available. The new steel powders were prepared with gas atomization method and heat treatment at

VTT. Before the preparation, the microstructure of Loviisa steel was analysed with optical microscopy, SEM/EDS and XRD. The composition was identical with the AISI321 steel. The composition of the AISI316Ti steel was otherwise identical but it has a lower carbon content. Because of the better availability, AISI316Ti steel powder was acquired as raw material for the preparation of the steel powder. The carbon content was increased with gas atomization method. The composition and microstructure was checked after atomization and the grain size was made equal with Loviisa steel with heat treatment.

Carbide has been suggested to be a possible form of ^{14}C in irradiated steel, when the nitrogen is originally as nitride. Therefore, Fe (4% C) powder, in which the main form of C is carbide, was chosen to be another solid phase. This powder has also been prepared with gas atomization method. The form of carbon was analysed with XRD using the Rietweld refinement of the spectrum. Iron carbide (cementite) was shown to be the major form of carbon (70 weight%). Additionally, a small batch of Fe3C powder was received from a component manufacturer from USA. This powder is not impurity-free. It has 92,4 % Fe_3C, but all C is as Fe_3C.

The leaching experiments with these new steel powders will be initiated in the near future as a part of the CAST project. The chemical forms of the organic carbon (speciation) will be studied in these experiments.

6.15 In-situ long term diffusion experiments in Grimsel (Project 27)

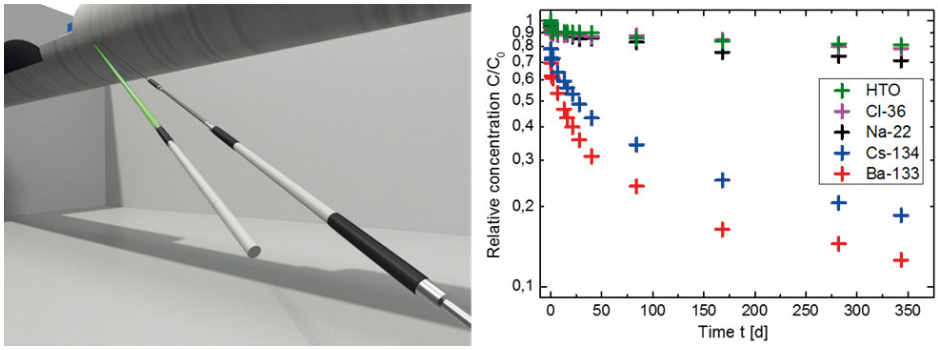
Marja Siitari-Kauppi, Mikko Voutilainen, Jussi Ikonen, Juuso Sammaljärvi, Lalli Jokelainen, Eveliina Muuri, Laboratory of Radiochemistry, University of Helsinki

In the project, carried out in the Laboratory of Radiochemistry, University of Helsinki, we studied migration of radionuclides in bedrock and their sorption on it. Aim of the project has been to develop measuring and analysis techniques as well as mathematical models which can be used to assess the impact of in-situ conditions on the transport properties of bedrock compared to laboratory results. We have studied the uncertainties of radionuclides retention parameters used in the current safety assessment calculations. The project has been carried out in collaboration with the University of Jyväskylä, the "Pore structure characterization of rock with tomography combined with matrix diffusion modelling" – project, the French University of Poitiers and Geological Survey of Finland, and it is closely connected to the Grimsel Test Site (GTS)-Phase VI framework programme in the Long Term the Diffusion (LTD) – project.

During the KYT2014-programme second in-situ experiment (Monopole2 LTD) was planned and launched in the Grimsel test site. Radionuclides (tritiated water, Cl-36, Na-22, Cs-134 and Ba-133) and stable selenium were used. The experimental set up consist of two parallel drill holes (see Figure) which are about 10-15 cm distance from each other. For this in-situ through diffusion experiment meter long section

was isolated from both of the holes. In the first section the tracer solution (total volume: 3 l) and in the second the pure synthetic ground water are circulated. We are following continuously the decrease of concentration in source hole and increase of concentration from the monitoring hole. The data from source hole provides immediate information on retention of the radionuclides as it is demonstrated in the following figure. In addition, elevated concentration of tritiated water has been observed in monitoring hole a half year after initiation of the experiment.

Figure 14. Experimental setup of the through-diffusion measurement in Grimsel test site (on left). The concentrations tritiated water (HTO), Cl-36, Na-22, Cs-134 and Ba-133 in source drill hole on relation to initial concentration have been decreasing during the first 344 days as expected (on right).

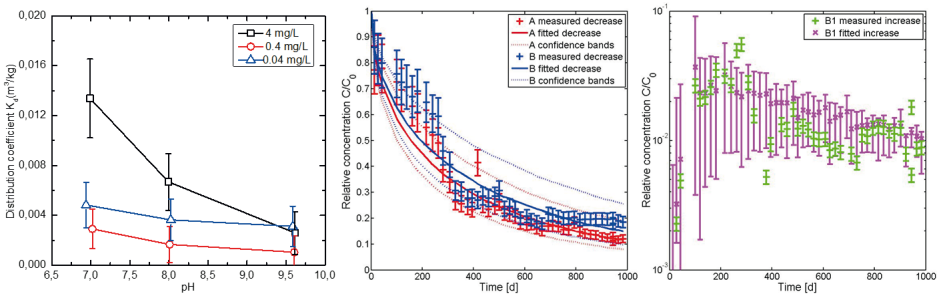


In addition, we focused on analysis of the first in-situ experiment which was performed during 2007-2009. We analyzed the concentration of sorbing radionuclides (Na-22 and Cs-134) inside the rock samples and contributed to modelling of the in-situ experiment (Jokelainen et al., 2013, Soler et al., 2014). Furthermore, we leached out non-sorbing nuclides (tritiated water and stable iodine), which has been diffused there during in-situ experiment, from the rock matrix (over cored subsamples; diameter 30 cm) and modelled the results in cooperation with the University of Poitiers using numerical Time Domain Diffusion modeling (TDD) (Ikonen et al., 2015). We also continued the laboratory experiments to support the in-situ experiments and determined diffusion and distribution coefficients needed in the analyses. During the years 2012–2013 we further developed the C-14-PMMA, when J. Sammaljärvi developed the heating polymerization method for C-14 labelled methyl methacrylate impregnations (Sammaljärvi et al., 2012).

In the second in-situ experiment inactive selenium is also used. To this end, we performed diffusion and sorption experiments on Grimsel granodiorite rock cores in laboratory using selenium. In comparison, similar experiments were conducted also in the Kuru gray granite, properties of which has been studied extensively in previous projects. Sorption experiments were performed in oxic conditions for

crushed rock as a function of grain size, pH and the concentration (see Figure), and as a function of pH also in low oxic conditions. Selenium diffusion experiments were performed in two 30 × 30 × 30 cm^3 sized rock blocks as through diffusion experiments. The decrease of selenium concentration was followed in the source hole and the increase of it in monitoring holes located at different distances from the source hole. The results of the experiment were analysed using TDD simulations (see Figure). As a result we got the effective diffusion coefficient of (2.5 ± 1.5) × 10^{-12} m^2/s and the distribution coefficient of (1.0 ± 0.5) × 10^{-4} m^3/kg for selenium for Grimsel granodiorite. The equivalent result from sorption experiment using crushed rock was about a factor of two larger which was explained by the difference in the specific surface areas between the two sample types; intact versus crushed rock.

Figure 15. Distribution coefficient of selenium was measured as a function of pH and concentration in oxic conditions for Grimsel granodiorite (on left). Results from diffusion experiment were determined from decrease of concentration in source holes (middle) and from increase of concentration in monitoring hole (on right).



When analyzing the results of the first in-situ experiment, it was found that heterogeneity of Grimsel granodiorite has a significant effect on migration of caesium. To this end, we developed the TDD method further in collaboration with the University of Poitiers and University of Jyväskylä and included possibility of heterogeneous sorption to it. In order to apply the method, one needs to know the mineral specific porosities of minerals and distribution coefficients of studied radionuclide. The porosities were determined previously by C-14-PMMA method and the distribution coefficients of caesium were determined for main minerals in Grimsel granodiorite as a part of this project. As expected, distribution coefficient of caesium was found to be strongly mineral dependent; high for biotite and lower for other minerals. The results of caesium sorption experiments were also modelled using PHREEQC program. By combining the mineral specific distribution coefficients and porosities to 3D structure obtained by X-ray tomography we observed that the heterogeneity of the rock texture has a significant impact on migration of caesium.

6.16 Chemical forms and sorption of radiocarbon in geosphere (Project 28)

Janne Lempinen, University of Helsinki, Laboratory of Radiochemistry

The research topic of the project is the chemical behavior of radiocarbon in geosphere and its sorption onto fracture minerals, especially calcite. Radiocarbon (14C) is one of the key radionuclides, along with 129I and 36Cl, in the assessment of radiation doses to humans and biota due to the final disposal of spent nuclear fuel. The project was started in 2013.

The speciation of radiocarbon in geosphere is the most important source of uncertainty in assessing its transport. Carbon is present in groundwater mainly as dissolved carbon dioxide and methane, and to a lesser extent as heavier hydrocarbons. Of these chemical species methane is not retained by the bedrock and is thus transported along the groundwater flow. Radiocarbon as dissolved carbon dioxide, on the other hand, can be sorbed onto calcite and iron (hydr)oxide minerals.

The starting point of the speciation studies of radiocarbon is the speciation of carbon in geosphere. The prevailing species are methane and carbon dioxide, the latter of which reacts to form carbonic acid and carbonate and bicarbonate ions. At repository depth radiocarbon as carbon dioxide is expected to reduce to methane, which is the prevailing species of carbon in groundwater. Moving upwards through the Olkiluoto bedrock there is a sulfate-rich layer at the depth of 200-300 m, where the concentration of methane decreases by two orders of magnitude and the concentration of carbon dioxide increases correspondingly. Therefore, the methane may be oxidized to carbon dioxide in the sulfate-rich layer as sulfate reduces to sulfide. However, this reaction is not spontaneous and requires microbial activity.

The changes in the speciation of radiocarbon are investigated by allowing a solution of a composition similar to groundwater to reach equilibrium with a suitable reducing agent such as iron. The experiments are done in headspace vials in a nitrogen atmosphere to reach experimental conditions that are reducing enough. After the equilibration, isotope-labelled bicarbonate is added to the samples and concentrations of methane and carbon dioxide are monitored using gas chromatography – mass spectrometry. The stable isotope 13C is used in the experiments as analogue for radiocarbon. Gas chromatography – mass spectrometry enables the determination of concentrations and carbon isotope ratios of the gases.

Studies on the sorption of radiocarbon have so concentrated on the isotope exchange between groundwater and fracture calcite. The dissolved carbon dioxide is present mainly as bicarbonate ions in the pH range of the groundwaters (7-9). In groundwater at solubility equilibrium with fracture calcite the dissolution and precipitation of calcite proceed at equal rates and the concentrations of calcium and bicarbonate remain constant. The radiocarbon in bicarbonate ions can also be precipitated as calcite in such conditions and thus forms a solid solution with

fracture calcite. The activity of radiocarbon in solution therefore decreases until the carbon isotope ratio is equal in the groundwater and on the surface of calcite. The reaction equation of the isotope exchange is

$CaCO_3$ (calcite) + $H^{14}CO_3^-$ (aq) ⇔ $Ca^{14}CO_3$ (calcite) + HCO_3^- (aq).

The sorption of radiocarbon onto calcite by isotope exchange was investigated in batch experiments. The rate of the sorption was studied in solution containing calcium chloride and sodium chloride where calcium concentration was between 0.2 and 100 mM and in fresh (ALL-MO) and saline (OL-SO) synthetic reference groundwaters. The activity of radiocarbon decreased exponentially as a function of time in all the solutions. The decrease of the activity became faster with increasing calcium concentration. Doing an exponential fit on the activity data in each solution as a function of time the half-life of the isotope exchange was determined. The half-life decreased from hundreds of days as a function of calcium concentration in the range of 0–10 mM. At higher calcium concentrations the half-life was constant, approximately 4.7 days. The typical calcium concentration in brackish and saline groundwaters is in the range of 15 to 100 mM.

The results can be exploited in the performance assessment of the final disposal of spent nuclear fuel. The carbonate radiocarbon has been shown to be retained by carbon isotope exchange between groundwater and fracture calcite. The results that are gained on the speciation of radiocarbon will enable a better understanding of the behavior of radiocarbon in geosphere.

6.17 Pore structure characterization of rock with nanotomography combined with matrix diffusion modelling (Projects 29–31)

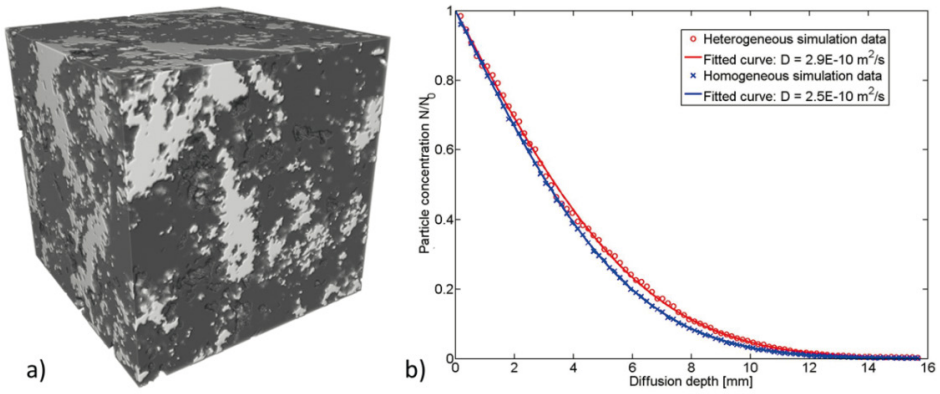
Jussi Timonen, Jukka Kuva, Joni Parkkonen, Department of Physics, University of Jyväskylä
Mikko Voutilainen, Laboratory of Radiochemistry, University of Helsinki

During the KYT2014 – programme our group executed two closely connected projects, that investigated the three dimensional pore- and mineral structure of rock using x-ray tomography and the C-14-PMMA method, and used information obtained this way in diffusion simulations. First of the projects, including heterogeneous rock structure in the modelling of matrix diffusion, took place during the years 2011-2012, and the second, Pore structure characterization of rock with nanotomography combined with matrix diffusion modelling, took place during the years 2013-2014. These projects were part of a research entity advancing the long term safety of final disposal of nuclear waste. The projects were executed in collaboration with the “In situ long term diffusion experiments in Grimsel” -project in the University

of Helsinki, the French University of Poitiers and Geological Survey of Finland, and they are also closely connected with the Long Term Diffusion (LTD) – project in the Grimsel Test Site (GTS).

During the years 2011-2012 a project started during the KYT2010-programme investigating the three dimensional pore- and mineral structure of altered Sievi tonalite and Grimsel granodiorite was continued. The structure of these rock samples was characterized using the x-ray tomography equipment of the University of Jyväskylä and the C-14-PMMA method developed in the University of Helsinki (see Figure). Diffusion in these structures was modelled using the Time Domain Diffusion (TDD) simulation method developed in the University of Poitiers. We were interested in the effect of heterogeneous structure on diffusion. A significant effect was found in the study (see Figure). TDD method was also developed in the project and a feature which allows the simulation of simple chemical sorption processes via a linear sorption coefficient was added. Heterogeneity of rock has been found to have an even stronger effect on the transport of sorbing elements. Transport modelling of caesium in Grimsel granodiorite was started in this project and after Mikko Voutilainen transferred to Helsinki after finishing his doctorate thesis, the work was continued as part of the "In situ long term diffusion experiments in Grimsel" project.

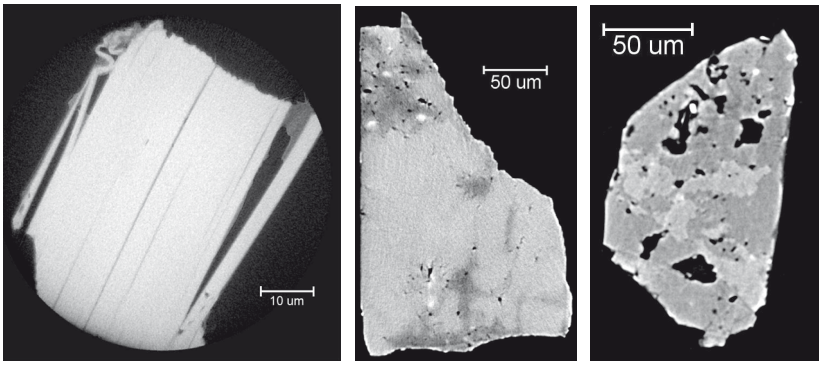
Figure 16. The effect of the heterogeneous structure of altered Sievi tonalite (a) on the diffusion coefficient determined from a simulated in-diffusion profile (b) was found to be about 16 %.



During the previous projects it was found that the resolution of x-ray microtomography is not enough to detect the inner pores of minerals within tight crystalline rock. Therefore in the project executed during the years 2013–2014 the rock samples were imaged with a nanotomography device that can reach a resolution of 50 nm. Two samples were chosen for the study, the first of which is from Olkiluoto, close to the planned final deposition site, and the other of which is from Sievi, which was included in the location survey done in the 90s. Because we were interested in the

alteration-induced changes in the pore- and mineral structure, the samples were chosen together with a researcher from the Geological Survey of Finland, taking into account the alteration of their minerals. The Sievi sample was more heavily altered than the Olkiluoto sample. Four minerals from the Olkiluoto sample and three minerals from the Sievi sample were chosen for nanotomographic imaging and a total of 15 imagings and reconstructions were done with nano- and microtomography devices. Based on these images deductions and observations were made of the inner structures of the minerals and the effect of alteration on them. The structure and elemental composition of the minerals was also studied with electron microscopy (SEM/EDS).

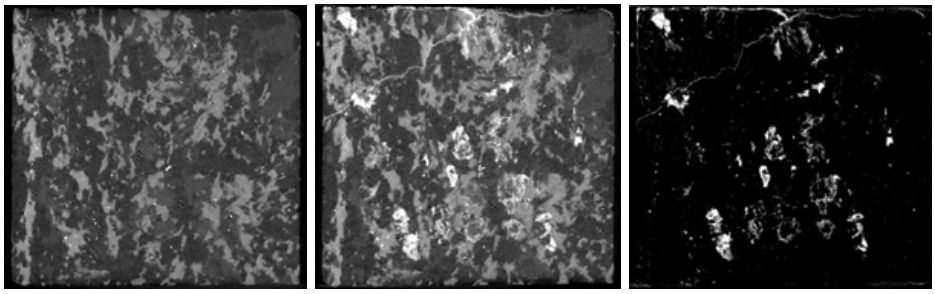
Figure 17. Left panel: Olkiluoto biotite sample imaged with nanotomography, showing the lamellar structure. Middle panel: Mildly altered Olkiluoto feldspar imaged with microtomography. Right panel: Strongly altered Sievi feldspar imaged with microtomography.



In the latter project we also investigated the transport of caesium in Olkiluoto veined gneiss and Grimsel granodiorite using x-ray microtomography. Caesium was used as a tracer because it is an important element in the Posiva safety analysis and heavy enough compared to typical elements of minerals to be distinguishable from tomographic images. Caesium has also been found to have a heavily mineral-dependent sorption coefficient, so the experiment could also give information on the effect heterogeneous structure has on transport. In the experiments six samples, sized 1×1×1 cm^3 were imaged before and after immersion in supersaturated CsCl solution. By comparing the acquired images information on caesium transport and transport routes was obtained (see Figure). The samples were manufactured so that five of the six faces of a cubic sample were sealed with epoxy resin, so that caesium could only enter the sample from one face. This allowed us to investigate the transport velocity of caesium by varying the contact time with CsCl solution. The first samples were re-imaged after 141 days contact time and the second samples

after 249 days contact time. In both cases caesium had migrated throughout the Olkiluoto sample and only a few millimeters into the Grimsel sample. As expected, caesium was seen mostly in biotite. The Olkiluoto samples have lots of high porosity areas, which also had lots of caesium (Figure). The Grimsel samples do not have such areas and therefore did not contain big caesium concentrations.

Figure 18. Olkiluoto veined gneiss sample before immersion in CsCl (left panel) and after 249 days of immersion (middle panel). Difference of the images (right panel) shows the infiltration of caesium into the sample.



6.18 Sorption of trivalent actinides on clay and (hydr)oxide minerals (Project 32)

Nina Huittinen, University of Helsinki, Laboratory of Radiochemistry

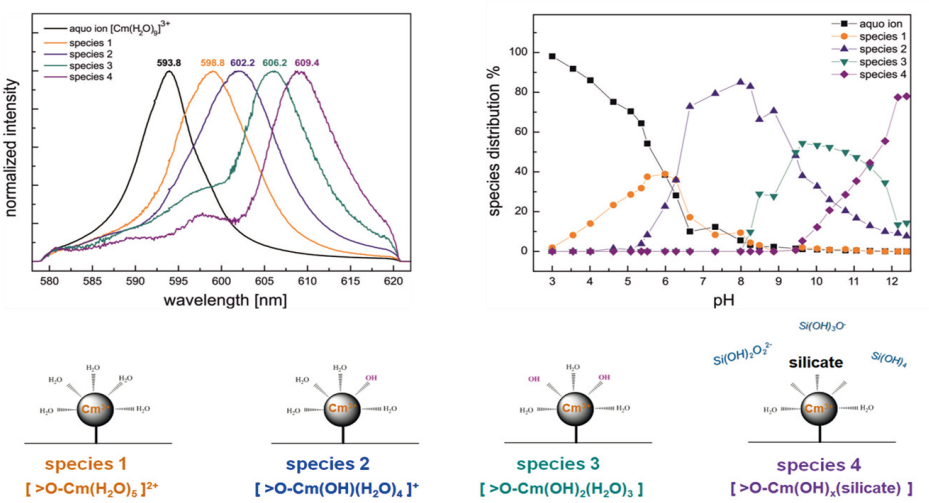
The aim of the project was to understand sorption processes and mechanisms taking place between trivalent metals found in nuclear waste and clay and (hydr) oxide minerals. The primary goal of the work has been to create new atomic scale data for mechanistic modeling of radionuclide transport in geological environments and to introduce new methods with which to study the sorption mechanisms. The main research methods in this work, laser-induced luminescence spectroscopy (TRLFS) and nuclear magnetic resonance spectroscopy (NMR), have not been used before in research related to nuclear waste disposal in Finland. Thus, the results obtained on the sorption behaviour of actinides on an atomic- and molecular scale have generated new, unique information that can be used for nuclear safety assessment purposes.

The project was carried out in close cooperation with the Karlsruhe Institute of Technology (KIT), Institute of Nuclear Waste Disposal (INE), and with the National Institute of Chemical Physics and Biophysics in Tallinn, Estonia. The main results of the study are presented below according to the methods used for the research.

TRLFS spectroscopy

Laser-induced luminescence spectroscopy has been used to study the speciation of the trivalent actinide, curium, on the clay mineral kaolinite ($Al_2Si_2O_5(OH)_4$). Kaolinite is present in granitic bedrock as a secondary fracture mineral and it has been used as a model mineral phase in the present work for more complex clay minerals like montmorillonite, present in the buffer backfill in spent nuclear fuel repositories. In the investigations on curium sorption on kaolinite, four different curium species, in addition to the curium aquo ion, were identified in the investigated pH-range. All experiments were conducted under argon atmosphere to exclude the influence of atmospheric carbon dioxide on the curium speciation through formation of curium-carbonate complexes. By analyzing the curium luminescence lifetimes, the following surface complexes could be identified in the kaolinite suspension: ≡O-Cm^{2+}(H$_2$O)$_5$ (species 1), ≡O-Cm(OH)$^{+}$(H$_2$O)$_4$ (species 2), and ≡O-Cm(OH)$_2$(H$_2$O)$_3$ (species 3). The fourth curium complex present in the alkaline pH range (pH > 10) could, after some additional experiments, be ascribed to a ternary curium silicate complex (≡O-Cm(OH)x-silicate) formed on the kaolinite surface between curium and silicates in solution originating from the dissolution of the mineral itself.

Figure 19. Below: The identified Cm-complexes on the kaolinite surface. Above: The emission spectra of the Cm species (left) and the species distribution as a function of pH (right).



NMR-spectroscopy

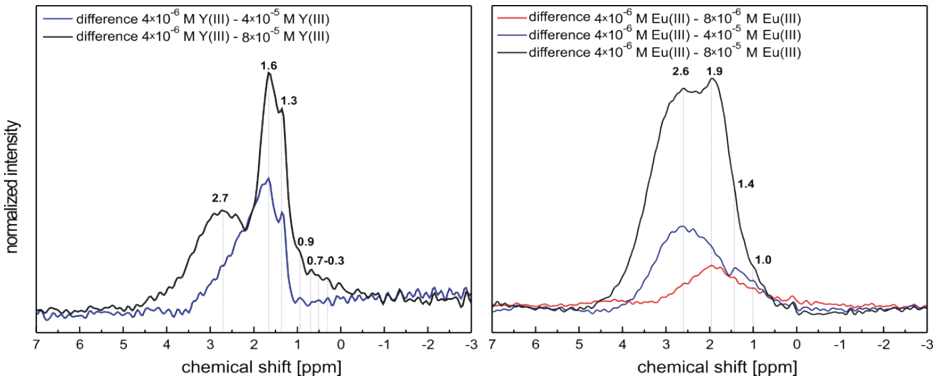
The specific sorption of europium and yttrium on kaolinite was investigated with nuclear magnetic resonance spectroscopy. The trivalent metal ions served as analogues for the trivalent actinides that could not be used in the NMR spectroscopic investigations. The NMR method was used to characterize the nature and the amount

of OH-groups present on the kaolinite surface, as well as to investigate how and to what type of surface groups the trivalent europium or yttrium ion is adsorbed. When a trivalent metal is adsorbed on the kaolinite surface, the hydroxyl proton dissociates according to:

$$\equiv S\text{-}OH + Y^{3+} \rightleftharpoons \equiv S\text{-}OY^{2+} + H^{+}$$

Upon metal sorption on the mineral surface some of the proton signal is lost, which can be detected in obtained 1H NMR difference spectra where proton spectra from yttrium/europium containing kaolinite samples were subtracted from the proton spectrum of the pure kaolinite surface.

Figure 20. 1H NMR difference spectra of yttrium (left) and europium (right) containing kaolinite samples.



The decrease of the proton signal as a function of metal ion concentration can be seen for protons with a chemical shift at 3.5-2.3 ppm, 1.6 ppm, 1.3 ppm, and 0.7-0.3 ppm. By comparing these proton signals to already existing results for yttrium sorption on α-alumina, singly coordinated Al-OH groups on the kaolinite edge surfaces can be seen to participate in the sorption reaction. These hydroxyl groups show up at chemical shifts below 1.6 ppm. The broad peak at 3.5-2.3 ppm indicates trivalent metal ion adsorption on the gibbsite-like basal plane of the mineral. This surface is in many studies considered inert toward metal ion sorption. From the difference spectra one can conclude that the sorption reaction occurs predominantly on the more basic Al-OH groups on the kaolinite edges, while the gibbsite like basal plane serves as a sorption site at higher metal ion loadings.

Summary

In this research, new molecular level information on the sorption mechanisms of trivalent actinides on clay minerals present in the final repository has been obtained.

Furthermore, new research methods have been introduced, that have not been used previously in nuclear waste disposal related research in Finland. Data obtained in the NMR investigations show in combination with TRLFS results that trivalent metal ion uptake on the kaolinite surface occurs on different types of hydroxyl groups as multiple different species that depend on the suspension pH and present dissolved mineral components. Several scientific publications have been published from results obtained in this project. The project ended in 2012 with the dissertation of M.Sc. Nina Huittinen.

6.19 Characterization of microbial communities of deep groundwaters, GEOMIKRO (Project 33)

Merja Itävaara, VTT

Microbial induced risks at the final disposal site of nuclear wastes may be due to several geobiological processes and interactions of chemical, physical and environmental factors. GEOMICRO (KYT2014) project is continuation of the earlier GEOMOL (KYT2010) project which also concerned development of deep sampling methods, optimizing and development of molecular methods for detection of microorganisms from deep groundwater samples originating from different geological and geochemical sites and depths in the bedrock. The aim has been to increase understanding about microorganisms inhabiting deep ground waters and especially fracture zone waters. Microbial role in deep bedrock aquifers has been poorly known until recently. Geogases, especially hydrogen and methane has great impacts on the presence and activity of microorganisms. Hydrogen which can be formed by several abiotic and biotic processes, is considered as the major source of energy for deep subsurfaces microorganisms and seem to be formed continuously by splitting of water molecules due to natural uranium and serpentinization. In addition iron can catalyse reactions where hydrogen can be formed in deep geological environments. Geogases hydrogen and methane in addition to sulphur and nitrogen compounds form also important sources of energy. Several biogeochemical processes connected to these geobiological cycles also may cause hydrogen sulphide formation which is highly corrosive agent if formed in the repository.

This project was a collaboration project with SALAMI and GEOBIOINFO projects in the KYT2014 research program.

Methods and results

Several deep microbiological sampling techniques has been developed in collaboration with scientists from VTT and GTK. The sampling sites are: Outokumpu deep borehole (2.5 km depth, sampling at several depths), 2. Nummi-Pusula (200 m depth), natural uranium site, 3. Olkiluoto (ca 20 boreboles 300-1000 m), 4. Kuhmo (600 m), 5. Pyhäsalmi cave (depth of the boreholes ca 2 km depth).

Microbial diversity and functional analysis

Biomass was concentrated by filtrating (0,2 umm filters). Microbial cells were disrupted to release nucleic acids and the presence of total community was analysed based on (DNA) or active microorganisms (rRNA, mRNA). Phylogeny and diversity of bacteria and archea were studied using 16S rRNA marker genes and the fungal species were identified using 18S rRNA and ITS genes. Fingerprinting technique PCR-DGGE or Tag 454 pyrosequencing was used to study species diversity and their functionality. Quantitative information on microbial groups was generated based on qPCR of target genes e.g functional genes encoding specific genes to detect e.g. sulphate reduction (dsrB), oxidation of methane (methane mono-oxygenase (pmoA), oxidation of methanol (mxaF), production of methane (mcrA), fixation of carbon dioxide (accC), nitrate reduction (narG), ammonia oxidation (amoA) were used to study geomicrobial processes.

Microbial cell numbers and diversity

Microbial cell numbers decrease in relation to depth in ground waters. The number of cells/ml water typically vary between 100.000 to 10.000 in the samples, but at deeper depths e.g. at 2.5 km cell amounts may be as low as 1000 cells/ml of water. Microbial communities varied considerably at different geological sites and depths. Deep ground waters inhabit a great diversity of different species of bacteria, archaea, fungi and viruses which vary depending on geology, gases and organic compounds. Iron and sulphur oxidizers (Epsilonproteobacteria, Gallionella sp.) typically inhabit the more shallow depths which may also cause iron and sulphur dissolution from the rock and increase sulphur compounds in waters. Outokumpu deep borehole was occupied by hydrogen consuming carbon dioxide fixing microorganisms Hydrogenophaga sp. as major community at the shallow depths. Sulphate reducers were detected at all sites and at all depths, but their species diversity varied. Sulphate reducers and methanogens are competitive microorganisms but at deeper depths sulphate reducers are outcompeted by methanogens. Fungal communities are less studied at geological sites, but we have found a great diversity of fungi present in the deep ground waters. Fungi are known to be able to form long filamentous hyphae capable of penetrating even concrete and may be considered when risks are estimated for the construction materials at the repositories.

Risks due to microorganisms

If bentonite barrier fails, evolution of methane from the earth crust at the final disposal site may cause biological corrosion risks to the copper containers used in deposition of the radioactive waste and, especially at sites where there is strong sulphate formation. In the present work we studied the activity of microorganisms in different conditions by using fluorescent viability stain. The results showed that generally the activity of microorganisms in deep groundwater is very low due to lack of electron donors and acceptors. However, very fast activation may occur e.g. due

to methane release from the earth crust or change in the composition of electron acceptors and donors. There are several species which can consume methane as carbon source and in the consortium with sulphate reducers they can form sulphides, which are corrosive agents. We studied this effect in the laboratory by monitoring activation of microorganisms by adding viability stain after addition of methane and sulphates and could also detect induction of gene expression (increases in mRNA) of the sulphate reducing community in the population (Rajala et al. 2015). Outokumpu deep borehole has been the major site to declare the effect of geochemistry and gases on microbial communities where we also have studied carbon cycling and effect of organic carbons (Itävaara et. al. 2011; Kietäväinen et al. 2013, 2014; Purkamo et al. 2013; 2014; Nyyssönen et al. 2014).

Metabolites in deep groundwaters

We also have studied organic compounds of deep ground waters using GC-MS and GCxGC-TOFMS techniques, instead of DOC and TOC which provide total organic carbon content of the groundwater. In Outokumpu deep borehole we could detect various small molecules. Short chain alcohols, carboxyl and dixarboxylacids (C4-C18) were detected, in addition to aminoacids, benzene and sulphate derivates (amides and acetates) and phenolic compounds. Samples from different depths contained different compounds suggesting relations to different the microorganisms

Free floating or attached microbes

Microbes are known to be attached on the surfaces of rock and tend to form biofilms. Most of our studies are mainly based on investigations of free floating microorganisms due to problems of studying biofilms in deep groundwaters. However, in Outokumpu deep borehole we have incubated copper and rock samples for a half a year to grow microbes at the surfaces. Results showed that the time was too short for biofilm formation but however some bacteria (Proteobacteria, Actinomyctes) and archaea could be detected.

6.20 Saline fluids, gases and microbes in crystalline bedrock, SALAMI (Project 34)

Lasse Ahonen, Riikka Kietäväinen, Ilmo Kukkonen and Arto Pullinen, Geological Survey of Finland (GTK)

The theme and key findings of the research

The project SALAMI (2011–2014) studied the interactions between deep saline fluids, gases and microbes in crystalline bedrock. The project was carried out in co-operation with VTT's Geomikro project and with Aalto University's GEOBIOINFO project. The common target of these projects was to study biogeochemical processes of the deep biosphere that may be relevant for the long-term safety of the nuclear

waste disposal. The 2.5 km deep scientific drill hole in Outokumpu was used as the main study site. Essential results of scientific relevance dealing with isotope geochemistry were published in international scientific journals.

A characteristic feature of saline groundwaters within crystalline bedrock is their water isotope composition which clearly differs from that of precipitation, fresh surface waters and near-surface groundwaters. Water-rock interaction, together with the changing isotope composition of precipitation due to the climate variation may explain the observed isotope ratios. Results obtained from the Outokumpu Deep Drill Hole indicate that the climate was 3–10 °C warmer than present during the groundwater infiltration, indicating a very long groundwater residence time in the bedrock. The study of evolution of saline waters was further elaborated by studying dissolved noble gases in water. Certain noble gas isotopes (^{4}He, ^{21}Ne, ^{40}Ar) build-up due to the decay of radioactive elements (U, Th, K) naturally occurring in the bedrock. By means of measured concentrations and isotope compositions, time required for their accumulation can be calculated, provided that diffusion phenomena have not affected the amount and composition of the gas. By studying the isotope ratios we could conclude that helium was formed by radiogenic process within the bedrock. Residence times required for the build-up of helium concentration, observed in different samples from the Outokumpu Deep Drill Hole, vary from 4 to 58 million years, with an average of about 30 million years. According to the present results, helium is built-up in radiogenic process in the crust *in situ*, while mantle-derived helium was not observed. Deep samples from Outokumpu did not show any helium concentration gradient indicative of diffusion from the deeper part of the crust. On the other hand, diffusion and transport of gases from the uppermost part of the crust to the atmosphere may take place. Within a time range of tens of millions of years, upward diffusion of helium was estimated to affect the concentration down to the depth of about 500 m in crystalline rock with an average porosity of 0.5 %. Methane flux was studied in the Outokumpu Deep Drill Hole by measuring the methane concentration on the collar. Based on the measurement, about 30–40 liters of methane is released from the borehole each day.

In order to elucidate microbial and biogeochemical diversity, research was also conducted in Kuhmo and in the Pyhäsalmi mine. The latter proved to be an excellent target, especially in terms of the representativeness of the samples, because pressurized samples could be collected from boreholes drilled from the mine down to the depth of more than 2 kilometers. A distinct difference between the gas composition of Outokumpu drill hole and the Pyhäsalmi mine was the low methane concentration in the latter. Microbes were found in all studied deep groundwater samples.

An essential part of the research was the identification and understanding of energy sources and utilization paths of subsurface microbial processes. Fermentative pathway utilizing photosynthetic energy from the ground surface was considered implausible based on the isotopic composition of deep methane which clearly differs

from typical microbial methane produced via fermentative pathway. Geochemical energy fluxes released from anoxic mineral transformation reactions remain negligible in proportion to the geological time scale. The most potential source of energy to take into consideration is the (dissolved) gas phase, in which hydrogen plays a key role. Some rock-type specific hydrogen producing reactions are known (e.g. serpentinization), but they evidently have local importance only. In the light of the observed behavior of helium, significant diffusion of hydrogen from any deep source cannot be considered a plausible process.

Significance and applicability of the results, connections to other research

Biogeochemical fluxes of energy and nutrients are of special importance for the near-field performance of a nuclear waste repository. The results constitute a data source available in performance analysis of the repository near field, especially related to canister corrosion and buffer processes. Biogeochemistry of bedrock fracture waters essentially deals with redox chemistry, i.e., electron transfer processes. In a wider perspective, understanding of the geomicrobiological processes and demonstration of the causalities constitute an important part of the safety case. Research continuum, networks created and the current extension to an EU co-operation promote the usability of our research results as an important and useful input of the safety case.

This research project has actively promoted interdisciplinary know-how and interaction, both in national and international levels. The connection with and national membership in the ICDP program (International Scientific Drilling Program) utilized through the technical support, as well as that received from Posiva Oy expanded the research possibilities at the Outokumpu Deep Drill Hole. International scientific co-operation was done with GFZ-Potsdam (Germany), University of Waterloo (Canada), Instituto Nazionale di Geofisica e Vulcanologian (INGV, Italy), and Deep Carbon Observatory – network (DCO). DCO co-operation has provided a vital link to create contacts to the leading research groups of carbon geochemistry. On the other hand, knowledge on the Finnish deep drilling research and know-how has been delivered to a wider scientific community. Swedish COSC deep drilling project has been followed, as well as participated in preparation of an international initiative for deep drilling primarily for microbiological research. In 2011 – 2013 SALAMI project was carried out parallel with the DEEP LIFE project financed by the Academy of Finland, and in co-operation with VTT and the University of Helsinki, Department of Geosciences and Geography.

Research methods

Methodologically the SALAMI project was firmly based on sampling, measurements and observations in crystalline bedrock, including deep borehole and mine environments. Devices were developed with an emphasis to extract representative gas and microbe samples from deep boreholes. In order to obtain pressurized

samples, a new sampling device was developed together with Lapela Oy and tested in Outokumpu. An evacuation line was constructed for collecting the gas phase from the pressurized fluid. Methane flux was measured in Outokumpu by collecting the gas in a closed chamber on the borehole collar and analyzing the concentrations by TDLAS+IR analyzer. Temporal variation of gas composition was studied at Outokumpu by on-line monitoring of the pumped fluid from 970 m for four weeks.

Tube samplings were performed to study variations in biogeochemical parameters with depth (in Outokumpu down to 1000 m and 2500 m, in Kuhmo RO-KR10 down to 600 m). Pressurized sampling was carried out in co-operation with ICDP technical support group from six depths. Long-term pumping was carried out at four levels (2300, 1820, 970 and 180 m) in Outokumpu. In the Pyhäsalmi mine, fluid samples were taken from several drill holes starting from different depth levels from 1080 to 1430 m. The drill holes were plugged by mechanical packers and equipped with valves, into which sample collection devices were installed.

In the Outokumpu Deep Drill Hole, two microbe trap experiments were done by installing specially designed traps at the depths of 500 and 976 m. Both experiments lasted for about half a year. The traps consisted of corrosion resistant steel cylinders which contained flow-through chambers filled with various substrate materials to collect microbial biomass. Substrates used included crushed rock, polished rock, glass slides and in the second test also copper plates.

Samples were analyzed using various geochemical and isotopic methods. Water samples were analyzed for cations and anions, including sulphide, pH, alkalinity, dissolved organic and inorganic carbon (DOC, DIC), and electrical conductivity (EC) (Labtium Oy, TVO Nuclear Services and Ramboll Analytics Oy). In addition, stable isotope composition of water as well as strontium, bromine, chlorine and lithium isotopes of water samples were analyzed from Outokumpu (GTK, University of Waterloo). A flow-through cell equipped with appropriate probes was used to monitor pH, EC, Eh and dissolved oxygen of the water during pumping. Dissolved gases were determined by gas chromatography (Ramboll Analytics Oy). Isotope compositions of hydrocarbons and hydrogen were determined by mass spectrometry (U. Waterloo, Hydroisotop GmbH and Isotech Laboratories Inc.). Isotope compositions of noble gases were determined by noble gas mass spectrometry at GFZ-Potsdam.

Modelling tools were needed in the interpretation of results, especially in geochemical thermodynamics in which PHREEQC code was the main tool.

6.21 Deep Bedrock Bioinformatics, GEOBIOINFO (Project 35)

Juho Rousu, University of Helsinki (2011–2012), Aalto University (2013-2014)

Objective and most important results

The project developed new bioinformatics methods and tools for studying the deep groundwater microbial community structure, functional genes and interactions with the geochemical environment.

The main results of the projects are the following:

1. Bioinformatics workflow for reconstructing metabolic networks from metagenomic data.
2. Web-based interface for visualizing the metabolic networks of the microbial communities
3. Computational methods for analysing the taxonomical distribution of functional genes and metabolic pathways.
4. Computational methods for analysing the statistical associations between microbe groups and geochemical variables
5. Knowledge of the statistical sufficiency of present metagenomic data for metabolic reconstruction.

Significance and future use of the results

The developed methods and tools make it possible to analyse and visualize the metabolism of deep groundwater microbial communities as well as the statistical associations between the microbial community and the geochemistry. The tools have been used to analyse the metagenomic data from deep drill holes provided by the GEOMIKRO and SALAMI projects. The tools are at the disposal of the research community even after the end of the project.

From the point of view of long-term storage of nuclear waste, the significant question whether microbial activity may cause corrosion of copper capsules in the given geochemical environment, requires a detailed knowledge of the structure of the metabolic networks of the microbial community. This requires the collection of large metagenomic (DNA) datasets from the relevant geochemical environment and reconstruction of the metabolic networks from the data. Based on the results of this project, significantly larger datasets are needed, if gapless metabolic networks are desired; the networks reconstructed from the current data remain too gappy from the perspective of the proper analysis of the limits of metabolic activity in the microbial community.

A suitable subgoal for future research would be to set the aim to reconstruct a gapless metabolic network for the microbial community encountered at the site of the nuclear waste storage. A such network could give an answer to the question: what metabolites can the community produce in theory in the given conditions.

Methods

The most important methods developed in the project are summarized below.

Bioinformatics workflow includes the processing of DNA data into metabolic networks:

1. Metagenome assembly: From DNA reads longer contigs are built
2. DNA-protein matching: protein counterparts are searched for the coding regions of DNA from Uniprot database
3. For proteins corresponding to enzymes and EC or KEGG orthology (KO) code is attached

Web-tool gives an user interface to visualize the metagenomic samples from different viewpoints

1. Basic statistics of the samples
2. Metabolic pathway visualization and statistics (KEGG maps)
3. Functional gene content analysis of the data (PCA and Sparse PCA analyses)

Statistical association analysis between microbial groups and geochemical variables

1. SCCA (Sparse canonical correlation analysis): finds small groups of microbes that correlate linearly or non-linearly with the geochemical profiles
2. KCCA (Kernel canonical correlation analysis): finds non-linear correlations between microbial groups and geochemical profiles.
3. Optimization of parameters and model evaluation: cross-validation and permutation tests
4. Visualization: correlation scatter plots and clustergrams

Figure 21. Bioinformatics workflow for metagenomic metabolic network reconstruction

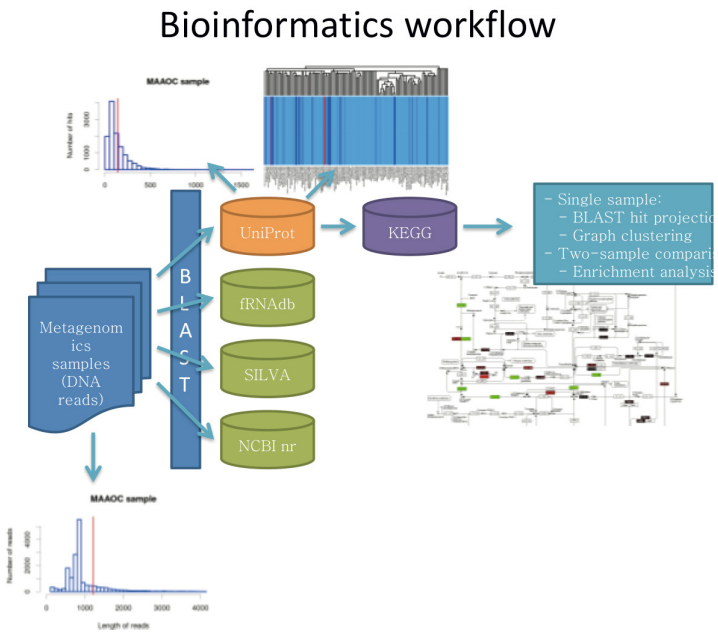
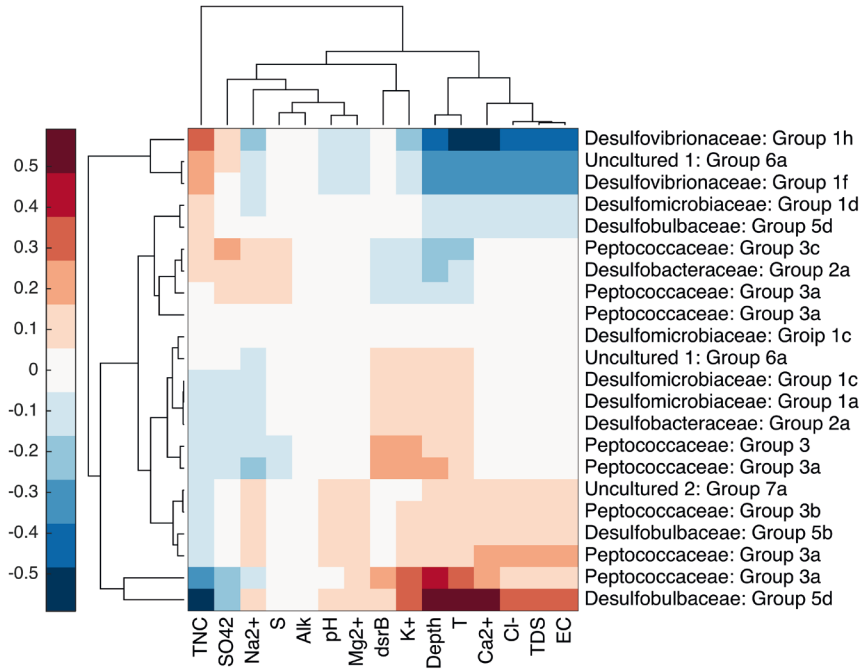


Figure 22. Clustergram for the visualization of correlating microbial groups and geochemical variables (Example: linear SCCA).



6.22 Risks of microbiologically influenced corrosion in the Finnish nuclear waste repository, REMIC (Project 36)

Leena Carpén, Pauliina Rajala, Malin Bomberg, VTT

In this project formation of biofilms and the risks of microbiologically influenced corrosion to the metallic decommissioning waste material in Finnish disposal environment was evaluated through experiments in the laboratory and on the site.

During the operation of nuclear power plants part of the structures become contaminated due to the neutron absorption. This operating waste includes especially the parts from the primary cooling circuit of reactor, like the primary nuclear vessel, internal fuel and control rods. Also contaminated decommissioning waste is produced during the dismantling the plant. Pipes, valves, filters and insulators e.g. are such waste, where the specific activity exceeds the free limit of 10 kBq/kg. The low level waste is packed into 200-liter drums and compressed to half of its volume using a hydraulic press. The waste is packed into concrete containers and transported to the repository using a radiation-protected vehicle. In the VLJ repository, the concrete containers are placed into the silos, excavated into the bedrock to the level of 60–100 meters below the ground surface. The open spaces between the containers and the walls of silos will be filled with concrete. Waste packages, concrete structures, other fillings, bedrock and part of biosphere will restrain the release of radioactive substances.

Corrosion rate of steels and the migration of compounds are typically very low and insignificant in concrete environment due to the high pH. The weathering of concrete, changes in oxygen content and the flow of ground water in and into the concrete silos and reaching to waste packages will expose the waste materials to different corrosion mechanisms. As a consequence of the corrosion as well as due to degradation of organic waste gases are formed which can cause water-bearing openings to the structures of the repository. As a final effect the radioactive nuclides may be released and thus carried through the groundwater causing radiation load to the neighboring area of the repository.

It is known that microbiological activity can induce corrosion as well as contribute to the integrity of concrete. Microbes can attach to the surfaces and can form together with exopolysaccharides and other organic material biofilm. Under the biofilm the amount of dissolved oxygen, inorganic and organic compounds as well as the pH can be totally different than in the surrounding liquid. Microbiological activity can accelerate among others general and localized, galvanic and intergranular corrosion. In anaerobic conditions especially sulfate reducing bacteria (SRB) are known to cause microbially induced corrosion. On the other hand, active microbes have been detected in deep bedrock environment at the disposal site. The metabolites produced

by these microbes can significantly reduce the pH of the environment which in turn affects the durability of concrete.

The effect of microbial activity on corrosion of decommissioning waste and the release of radioactive nuclides are not yet sufficiently known. In this project the effects of microbially induced corrosion and the properties of the biofilms have been studied both at the disposal site of the operating waste repository (VLJ-repository in Olkiluoto) and at the laboratory. In order to estimate the corrosive effects of microbial activity experiments with and without microbes were performed. In this project a novel experimental arrangement has been created by which it is possible to study the microbially induced corrosion. Electrochemical methods enabling monitoring of mechanisms and corrosion rate of general and localized corrosion in anaerobic environments in real time have been applied in this project. Also gravimetric method and microscopy has been utilized in corrosion studies. Corrosion products have been analysed using EDS or XRD methods.

The composition of the biofilm has been evaluated utilizing the methods of molecular biology. The biofilm forming community (bacteria, archaea, fungi) has been mapped by ChIp-sequencing analyses and the relevant microbial groups for corrosion standpoint were quantified with quantitative PCR.

The project was composed of following sub tasks: 1) To design and construct the test arrangement 2) To study the microbially induced corrosion of decommissioning waste 3) To study the properties of the biofilms formed on the surfaces of decommissioning waste 4) Reporting.

The technical aims of the project were:

1. To develop reliable experimental arrangement to estimate the microbially induced corrosion of decommissioning waste materials in Finnish repository conditions.
2. To determine the effects of microbial activity to the corrosion rate and corrosion mechanisms of decommissioning waste materials in Finnish repository conditions
3. To define the microbes attached to the surfaces of decommissioning waste materials and the properties of the biofilm formed during the course of time in laboratory conditions as well in the field conditions

The most notable results during the duration of the program:
Four laboratory experiment series were performed within years 2011-2014 (preliminary experiment, 1.laboratory experiment, 2. laboratory experiment and biocide experiment) and field test with the total time of 3 years. In this project experimental set-up that enables real-time monitoring of corrosion indicating electrochemical parameters under modified atmosphere was successfully developed. In addition, a set-up that enables simulation of different scenarios and accelerated microbial functions was developed.

In the preliminary experiment the behaviour of carbon steel was studied in two different temperatures. The main goal was to determine the best experimental methods to apply for the studies at the laboratory scale. According to the results of this study the following experiments were performed in situ temperature and using biocides to establish the abiotic environments. One significant scientific results of this preliminary study was that temperature had critical effect on the species composition of sulfate reducing bacteria. In addition a high proportion of iron consuming native bacteria from the groundwater in the biofilm and their possible role in corrosion were important findings.

In first laboratory experiment the corrosion of carbon steel and two different stainless steels was studied in different simulated environments using the natural groundwater as a basis. Some of the environments were highly alkaline simulating the environment where concrete is present. In some environments the flow of nutrients was simulated by adding glucose illustrating the nutrients being liberated from the degradation of organic waste. Methane was added to some environments to simulate the nutrient load originating from the flow of methane rich ground waters. Substrate additions produced changes in microbial community and according to electrochemical measurements also enhanced carbon steel corrosion. On the other hand, addition of concrete inhibited the formation of biofilm and decreased also corrosion. However, in the presence of concrete the corrosion tends to be more localized.

In the second experiment the aim was to accelerate corrosion by an enrichment of sulfate reducing bacteria isolated from the native drill hole water from the repository site. This was found successful and the corrosion rate of carbon steel was at the same level than with the glucose added as a nutrient. Noteworthy was also the fact that in this experiment the biofilm formed on the one type of stainless steel was clearly deviating from the others and in this specimen also starting pits could be detected after the exposure.

The effect of concrete on the corrosion and on the biofilm formation on the surfaces of carbon and stainless steel was studied at the field test. The corrosion rate of stainless steel was very low at the studied environments. Whereas, high corrosion rates and similar corrosion behaviour of carbon steel was achieved in environment without concrete that had been seen in the earlier studies in the actual bore holes. Without concrete the corrosion rates of carbon steel was clearly more intensive both in abiotic and especially in biotic environment where the corrosion rate was almost 500 times that of concrete environment.

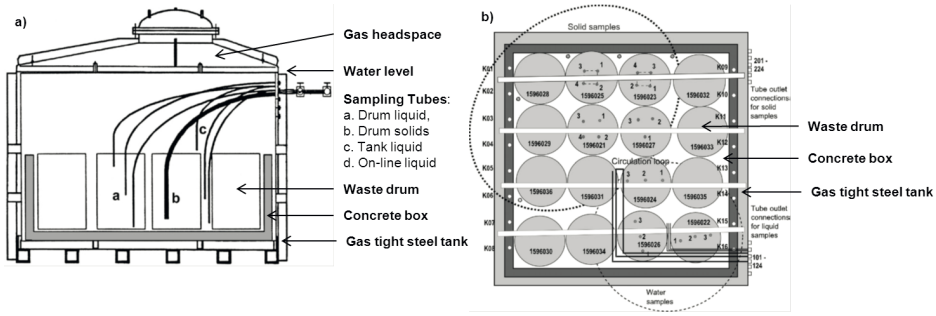
6.23 Microbiology of Gas Generation Experiment (Project 37)

Merja Itävaara, Minna Vikman, Kaisa Marjamaa, VTT

Topic

Finnish nuclear power plants produce 150-350 m³/year of low (LLW) and intermediate level (ILLW) radioactive wastes. The activity of low level waste is below 1MBq/kg and it typically contains high quantities of organic matter (YVL 8.3/ 29.6.2005, http://www.tvo.fi/Ydinjätehuolto). In order to simulate the geological disposal safety issues of low and intermediate radioactive wastes Teollisuuden Voima Oyj initiated the Gas Generation Experiment (GGE) in the VLJ repository in Olkiluoto. The experiment was started in 1997 and is ongoing with continuous monitoring of gases, conductivity, pH and temperature which are essential variables in safety analysis. Microbiological risks due to geological disposal of low level radioactive wastes are biocorrosion of metals and gases generated in biodegradation processes of organic materials which would increase migration of radionuclides to biosphere.

Figure 23. The Gas Generation Experiment (GGE) from side (a) and from top (b). 16 drums (vol. 200 l) containing low radioactive wastes originating from the Olkiluoto nuclear power plant were set into the concrete box inside 20 m³ gas tight acid proof steel tank (Figure modified from Small et al. 2008).



This project aimed to estimate the biodegradation degree of the waste materials since 1997 until 2013 and studying microbial diversity and changes during time inside the tank. Because cellulose was the major waste material (around 40%) we aimed to enrich cellulose degraders and to study their growth requirements.

How much LLW waste has been biodegraded during 16 years?

Gas generation started up during the first year of experiment. In addition to methane formation also volatile fatty acids detected by capillary electrophoresis indicated active microbial degradation process. Volatile fatty acids are microbial metabolites

which are formed as intermediate during anaerobic biodegradation of organic matter.

Due to alkaline pH of the tank water we assumed that ca 40% of the biogas formed was CO_2 which was almost completely adsorbed in the tank water. CO_2 in the head space of the tank has been less than 1% during the experiment. The biodegradation degree was calculated based on organic carbon content of the waste materials and biogas (CH_4+CO_2) formed during incubation time (16 years). Biodegradability has been performed before starting up the experiment to estimate the maximum gas generation from "biodegradable materials of the LLW (Itävaara, 1997). Based on all these calculations we estimate that less than 7% of the cellulose-based wastes has been biodegraded.

Microbial diversity in the GGE

Molecular method was used to study microbial diversity changes during time between 1998 and 2013. Microbial communities were analysed by extracting DNA from the microbial biomass of the frozen water samples. In addition microbial communities were analyzed by quantitative PCR and Tag 454 pyrosequencing.

Microbial diversity and activity in different parts of the tank varied considerably and was in connection to the content of the drums (cellulosic material/other organic material) and the amount of organic matter (drum lid level/bottom of the tank). The highest quantity of total bacteria, sulphate-reducing bacteria and methanogens were detected in the drums containing mostly cellulose-based material. The composition of the microbial diversity also showed considerable changes during the years. Several microbial groups could be directly connected to the biochemical reaction-transport-model GRM (Small et al., 2008), especially methanogens and microbes involved in hydrolysis of cellulosic materials were detected. Methanogens represented both acetate utilizing acetoclastic and hydrogen utililizing hydrogenotrophic methanogens. In addition sulphate reducers considered to be involved in biocorrosion were detected (*Desulfovibrionaceae*, *Syntrophobacterales* or *Desulfobulbaceae*). Archaeal population represented similar microbial communities found in typical anaerobic digestion process.

Microbes involved in degradation of cellulose-based polymers were studied by isolation and identification of the (hemi)cellulose hydrolysing microbes. Microbes were isolated in anaerobic conditions, both directly from the water samples or via enrichment in culture media containing cellulose or hemicellulose (xylan) as carbon source. Isolation of microbes was carried out using azurin cross-linked (AZCL) xylan or cellulose agar plates, where blue halo around the colonies indicate xylanolytic or cellulolytic activity, respectively.

The cellulose-based materials are composed of cellulose polymers and in the case of e.g. many paper products, also hemicelluloses. Thirteen hemicellulose (xylan) degrading bacteria strains were isolated and identified from the water and solid samples collected from the waste drum filled mainly with cellulose-based waste.

No xylanolytic bacteria were found from the water sample collected from the space between the drums, suggesting that growth of these bacteria is promoted by the presence of the cellulosic wastes. The identification was carried out using 16S ribosomal RNA gene sequences of the strains, which indicated that the isolated bacteria belong to Paenibacillus sp. and Clostridium sp., and six of them represent potentially new species. The (hemi)cellulolytic growth of the *Paenibacillus* strains at different temperature and pH was studied using the AZCL-agar plates, showing that the strains exhibit xylanolytic growth at 10–30 °C, one of them also at 5°C and one at 40°C. The growth pH of studied strains was between 5 and 9. This indicated that the prevailing conditions in GGE (8-10°C, pH 6–7) support the xylanolytic growth of these bacteria. The enzymes produced by these bacteria were studied by cultivating the strain in semi-liquid media with xylan or cellulose as carbon source, and studied the soluble and cell bound enzymes with standard spectrophotometric enzyme assays. The results showed that the enzymes produced by these bacteria hydrolyse both polymeric xylan and low molecular weight xylose derivatives, with short term (10 min assay) pH and temperature optima of the xylan hydrolysis being ca at 40-60°C and pH 7. The genomes of selected strains were studied via DNA isolation and high throughput sequencing (Illumina HiSeq). The analysis of assembled genomes showed that the genomes contain multiple enzyme genes potentially involved in the ability of these bacteria to utilize xylan nutrition. Although none of the studied strain showed ability to efficiently degrade cellulose in the tested condition, the genomes of the studied strains also contained enzyme genes associated to cellulose degradation. This suggests that in vivo in GGE these bacteria can also be involved in hydrolysis of cellulose polymers of the waste materials.

Referencest

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6.24 Project: Use of empirical data to improve radioecological modeling applied to risk assessment of radioactive waste (Project 38)

Jukka Juutilainen, University of Eastern Finland

Research topic and main results

The aim of the project was to improve radioecological modelling applied to Finnish forest ecosystems and the use of such modelling for assessing the risks of radioactive waste. Transfer of radionuclides from soil to plants and animals was a particular focus. The project consisted of two sub-projects.

The first sub-project aimed at investigating whether radioecological models and their accuracy could be improved by using non-linear instead of linear models. This sub-project was based on the observations of our project funded by the KYT2010 Programme, showing that transfer of elements into plants is non-linear: the transfer is most efficient when concentration in soil is low, and it cannot be described by the concentration ratios that are commonly used in radioecological models and are assumed to have a constant value independent of concentration in soil.

In a traditional linear model based on a constant concentration ratio, plant radionuclide concentration increases linearly with soil radionuclide concentration, independent of the total concentration of the element in soil. In our non-linear model, in contrast, plant radionuclide concentration is influenced by both the radionuclide and the total concentrations of the element in soil. In simulations modelling concentration in spruce needles, more than fivefold maximum differences were observed between the traditional linear model and a model based on a non-linear equation. The linear model can therefore either underestimate (when total element concentration in soil is low) or overestimate (when total element concentration is high) the uptake of radionuclides into plants, in comparison to the non-linear model (which most likely better describes the real-life process). A third modelling approach developed in this project gave results that were closely similar to those given by the non-linear model. This can be considered the most important result of the theoretical part of the project, as the new modelling approach is simple, and the necessary parameter values are generally available. The model based on non-linear equations, in contrast, would require empirical measurement of parameter values for different elements (radionuclides) and different types of plants. It also seems that the uncertainty of model predictions could be reduced by using models not based on linear concentration ratios.

The second sub-project used experimental ecosystems for studying transfer of uranium, cobalt, molybdenium, lead, thorium and zinc from soil to boreal forest plants and animals (earthworm, snail). The experimental data supported the finding, previously observed in natural forest settings, that transfer of elements from soil to plants is nonlinear. Concerning transfer of elements into animals, the experimental

ecosystems produced valuable new data, as uptake into animals cannot be easily investigated in natural settings. Both earthworms and snails are important parts in the food webs of terrestrial ecosystems. Somewhat surprisingly, the transfer of elements from food and soil to animals was found to be similarly nonlinear as transfer from soil to plants. This is presumably related to the fact that animals take up essential elements more effectively when concentration in food is low (aiming at a constant concentration in tissues). The uptake of uranium was almost linear (constant concentration ratio), which may be related to the fact that it is not an essential element. However, the behaviour of lead was similar to that of essential elements. Modelling of radionuclide transfer into animals may require using different approaches for different elements.

Publications and theses: Five articles in international peer-reviewed journals have been published during the project. One manuscript is almost completed and 2 are being prepared; these will be submitted during 2015. In addition, a review article is planned for 2016. The doctoral thesis of Päivi Roivainen was completed in 2011, and Tiina Tuovinen will defend her thesis during 2015. A Master's thesis and a Bachelor thesis were completed during the project, and a Master's thesis is expected to be completed in 2015.

Significance of the results for radioactive waste management, links to other research

Knowledge of the transfer of radionuclides from soil to plants and food webs is needed in biosphere modelling associated with safety analysis of the disposal of spent nuclear fuel. The project has produced data relevant to Finnish ecosystems, and improved understanding of the nonlinear nature of transfer of elements from soil to biosphere. The results have a potential to improve the accuracy of radioecological modelling.

The results of the project are helpful for improving models that are used in assessing risks to human health and wildlife from radioactive waste. The results can also be used assessing environmental impacts of uranium ore prospecting and possible uranium mines, development of radioecological models in general, and risk assessment of other environmental contaminants (such as heavy metals).

Methods

Data collected in an earlier KYT2010 project were utilized in simulations done in this study. These data included element concentrations in soil and plants in two forest sites in North Savo.

Empirical Langmuir-type nonlinear equations, derived from the KYT2010 project data, were used in this project to predict concentration in plants for selected radionuclides relevant to nuclear waste. The concentrations predicted by these equations were compared to a linear model, for which concentration ratios were derived from the same data. A third model fitted with the observations almost as

well as the Langmuir type equation, and was also included in the comparisons. This third model assumes that the total concentration of an element is constant in the plant, independent of concentration in soil (consistently with to our empirical data), and the radionuclide to total element ratio is assumed to be identical in the plant and in the soil, when equilibrium has been reached after contamination with the radionuclide.

These three models were used to simulate different assumed concentrations of contaminating radionuclides. Dependence on the total element concentration (stable + radioactive nuclides) was included in the simulations. The modelled soil radionuclide concentrations were the maximum concentrations that could end up into the soil according to worst scenarios, and the total element concentrations in soil were assumed to vary within the values observed in our boreal forest data.

In the experimental part of the project, mesocosms were put up at the research garden of the university, with two different types of soils differing with respect to the concentrations of uranium and several other elements. Apart from the soil, the ecosystem of the mesocosms consisted of three plant species (downy birch, narrow buckler fern, Scandinavian small-reed) and two animal species (earthworm, copse snail). In addition, uptake into snails was studied in microcosms including either only snail food (birch leaves) or both food and soil; this experiment was done to study the role of direct soil contact in transfer of elements into snails. Total element concentrations in soil, plant and animal samples were analysed by ICP-MS, i.e., this study (like our previous studies) used total element concentration as a model for the transfer of radionuclides in ecosystems.

6.25 International Socio-Technical and Safety Challenges for Implementing Geological Disposal of Spent Nuclear Fuel – Finland and EU – FInSOTEC -2012-2014 (Project 39)

Tapio Litmanen, University of Jyväskylä

In the first phase of the FInSOTEC project the main socio-technical and safety challenges for implementing geological disposal were identified and safety and risk dialogue between experts and expert organizations were also studied. The results of the earlier phases of the project are presented in a country report (Nurmi et al. 2012) and several other publications (Litmanen et al. 2013a; Litmanen et al. 2013b; Litmanen et al. 2012; Litmanen et al. 2014a; Litmanen et al. 2014b).

In the article entitled “Socio-technical megaproject comes to its waypoint: The challenges and uncertainties of the final disposal of spent nuclear fuel as future management” (Litmanen et al. 2013a; Litmanen et al. 2013b) conclusions on the first phase of the FInSOTEC project are drawn. There the researchers underline

the importance of understanding the current phase of nuclear waste management in the context of long socio-technical transformation. After thirty years, the Big Science project conducted by the nuclear waste company Posiva has arrived at its evaluation waypoint. This socio-technical conglomeration can be characterized as a unique outcome of jurisprudence, economic, political, cultural, scientific and technological efforts. The project has integrated other parts of society into itself, it has required institutional changes, absorbed research and supervision resources, ignited a huge societal debate on its social legitimacy, and impacted on societal development in terms of issues such as questions about energy. The megaproject indicates the complexity of future management. Based on the research group's studies, several different socio-technical challenges and uncertainties still exist. Some challenges are linked very closely to the project, whereas others are related to the socio-technical order. The project also continues to face scientific-technical, epistemological-methodological, socio-political, administrative-jurisdictional, environmental and project management challenges. The handling of a construction licence for a repository for the disposal of spent nuclear fuel can be seen as societal evaluation in which many technical and political questions are interconnected. If the advancement of the project is permitted, it will face a variety of management challenges in different phases.

In a later phase of the FInSOTEC project many resources were directed to the study of the scientific controversy, possibly even threatening the implementation of the final disposal of spent nuclear fuel in Finland and Sweden (Litmanen et al. 2014a). The risk of corrosion in oxygen-free water had become a safety critical issue also affecting the political decision-making on the advancement of the geological disposal of spent nuclear fuel. In Sweden there has been extensive discussion about the issue since 2007, but only recently has this debate increased in Finland, although a similar disposal concept (KBS-3) is applied in both countries. The team's report (Litmanen et al. 2014a) analysed how the implementer, Posiva (a Finnish nuclear waste company), and the regulator, STUK (the Finnish Radiation and Nuclear Safety Authority), have been engaged in a dialogue on the risk of copper corrosion. For over thirty years the implementer and regulator have been engaged in a series of negotiations on the advancement of research, planning and technical design related to SNF disposal. The aim here was to determine 1) how the implementer, Posiva Oy, has presented the issue of copper corrosion and copper corrosion related research, 2) how the regulator, STUK, has assessed and reacted to what Posiva has presented and 3) what the long-standing risk dialogue tells about the transformation of the Finnish regulatory culture and the relationship of the parties. Moreover, the study discusses the importance of risk dialogue at different stages of the risk governance processes and how the risk dialogue transforms the roles of the parties. The insight into the copper corrosion risk dialogue between Posiva and STUK was gained by examining core documents regarding this interaction, namely the Research, Development and Technical Design (RTD) review process and the construction licence application

(CLA) review process. From Posiva's side RTD programmes published in 2003, 2006 and 2009 and their successor, the Nuclear Waste Management (NWM) programme published in 2012, were studied. When examining STUK, the statements by the regulator to the Ministry of Employment and the Economy (MEE) on the basis of Posiva's reports were analysed. Posiva's pre-licence construction application (pre-CLA) in autumn 2009 and STUK's review of it were also included in the analysis. As STUK's review of Posiva's construction licence application, submitted in 2012, was at the time of the study still under way, only Posiva's application was examined.

Finnish nuclear waste risk governance is characterized by a strong role of central actors, STUK and Posiva. The Finnish regulatory culture is deemed flexible, development oriented and, as such, oriented towards gradual learning and refinement. The results of the risk dialogue study suggest that Posiva's reporting evolved from merely presenting a situation to more focused and extensive discussions. The investigation of the dialogue showed that STUK exercised its right to demand further information, while the implementer, Posiva, was compelled to comply with the requirements. Nevertheless, the organizations appeared to operate consensually, meaning that they both pursue the successful development of a safe repository for SNF, indicating rapprochement and transformation of clear-cut roles. However, the results show that under the normal steady flow of interaction, the risk governance process is oriented towards mutual learning and improvement, but at the time of crucial decision-making extra tensions came into the relationship. In the ideal cases the roles of implementer and regulator should be clear-cut, but the study of long-standing interaction indicates that the engagement in dialogue has transformed STUK's role in the direction of development orientation. Thus shifting STUK's input to the advancement of the project gives it a sort of consultative role, while at the time of crucial decision-making the role of regulator is strengthened.

At the final phase of the FInSOTEC project two important tasks were to study (1) risk regulation regimes in Finland and Sweden and (2) the public debate over the construction licensing procedure in the Finnish and Swedish print media.

The study of risk regulation regimes in Finland and Sweden was conducted in cooperation with the Department of Social Sciences at Michigan Technological University, USA. It has produced an article manuscript (Litmanen et al. 2015), which will be submitted to an international journal after the finalizing touches. The cases to be studied are the Finnish and Swedish plans, which are deemed the most advanced in terms of nuclear waste management, to dispose of spent nuclear fuel in geological repositories for thousands of years. In Finland, the nuclear waste company Posiva submitted its construction licence application at the end of 2012. For the Swedish case, in March 2011 the Swedish Nuclear Fuel and Waste Management Company (SKB) submitted license applications for a general license to construct, possess and operate a spent nuclear fuel repository at one municipality, and an encapsulation plant in another municipality. The companies have assured the public that their plans will hold up to critical assessment and that the disposal of spent fuel will

not harm the environment and the health of future generations. However, critics doubt that safety can be guaranteed. The paper focuses on: 1) institutional waste management frameworks in both countries, paying special attention to the role of civil society and transnational risk regulation; 2) how laypeople and civil society organizations (CSOs) have been able to participate in and contribute to recent spent nuclear fuel decision-making; and 3) the nature of nuclear waste risk debates in both countries. The research data includes official documents of the nuclear waste companies and nuclear safety authorities, but also information from CSOs and laypeople. The theoretical framework applied is a risk regulation regime approach, which is oriented towards institutional issues. However, it is argued that regulation by civil society has to be better integrated into these regimes. CSOs can press corporations to deliver improvements in their research, development, and technical design and similarly they can follow the work of regulators and propose potentially superior ideas for improving waste management.

Fewer resources were allocated to the study of the public debate over construction the licensing procedure in the Finnish and Swedish print media. The rationale for this was the idea of only collecting this data at the end of the KYT2014 research programme to be able to possibly continue the study of the media debate during the new KYT2018 research programme period.

FInSOTEC 2012-2014 has been a joint project of the Department of Social Sciences and Philosophy, University of Jyväskylä and the School of Management, University of Tampere. The KYT2014 research programme has provided national funding for the research team in order to participate comprehensively in a larger EU-project called "(International) Socio-Technical Challenges for implementing geological disposal" (InSOTEC). This InSOTEC project was funded by the 7th Euratom Research and Training Framework Programme on Nuclear Energy for the European Commission (Theme: Fission-2010-1.1.2; Research activities in support of the implementation of geological disposal; Grant agreement no: 269906).

Annex 1

KYT2014 research projects 2011-2014

	Time	Project	Organisation
		New and alternative technologies in nuclear waste management	
1	2011-2014	Advanced nuclear fuel cycles – new separation technologies	HYRL
2	2011-2014	Advanced Fuel Cycles – Computational Fuel Cycle Analysis	VTT
3	2011-2014	Transmutation of nuclear waste in an ADS reactor (FLUTRA)	Aalto
		Safety case	
4	2011-2013	Safety case for final disposal (LS-TUPER)	VTT
5	2011-2012	Safety case for final disposal (LS-TUPER) – Complementary considerations	HYRL
6	2011-2012	Complementary considerations of the safety case TUPER/ GTK	GTK
7	2011-2012	Safety case for final disposal (LS-TUPER); computational model development	Ludus Mundi
		Buffer and backfill materials	
8	2011-2014	Assessment of bentonite characteristics (BOA)	VTT
9	2011-2014	Assessment of bentonite characteristics (BOA), Subproject	Numerola
10	2011-2014	Assessment of bentonite characteristics (BOA), Subproject	HYFL
11	2011	Assessment of bentonite characteristics (BOA); subproject THM model	Ludus Mundi
12	2011-2014	Assessment of bentonite characteristics, subproject mineralogical studies at GTK (BOA/GTK)	GTK
13	2011-2014	Phenomenological THM modelling of bentonite (subproject of BOA consortium)	JYFL
14	2012-2014	The mechanical behaviour of bentonite and backfill block surfaces in shear stress	Aalto
15	2011-2014	Subproject The effect of colloids on radionuclide migration	HYRL
		Canister	
16	2011-2014	Corrosion of copper by water under oxygen free conditions	Aalto
17	2011-2014	Mechanical strenght of copper canister waste	Aalto
18	2011-2014	Material Integrity of Welded Copper Overpack	VTT
19	2011-2013	Sulphide-induced embrittlement of CuOFP	VTT
20	2012	Corrosion monitoring under disposal vault conditions	VTT
21	2013-2014	Effect of microbial action on different corrosion processes of the copper canister (MICCU)	VTT
		Other safety related studies	
22	2011-2014	Durability of engineered concrete barriers under final disposal conditions (subproject 1)	Aalto
23	2011-2014	Durability of engineered concrete barriers under final disposal conditions (subproject 2)	VTT
24	2011	Rock quality: Visualization and modeling of rock quality parameters in 1-3 dimensions in crystalline bedrock; confidence assessment of the models*	Aalto
25	2014	KARMO – Mechanical Properties of Rock Joints	Aalto
26	2011-2014	Release of 14C (Carbon-14)	VTT
27	2011-2014	In situ long term diffusion experiments in Grimsel	HYRL
28	2013-2014	Chemical forms and sorption of radiocarbon in the geosphere	HYRL

29	2011	Combining the heterogeneous properties of rock matrix into matrix diffusion modelling	JYFL
30	2012	Including heterogeneous rock structure in the modeling of matrix diffusion	JYFL
31	2013-2014	Pore structure characterization of rock with nanotomography combined with matrix diffusion modeling	JYFL
32	2011-2012	Sorption of trivalent actinides onto clay and (hydr)oxide minerals	HYRL
33	2011-2014	Microbiological characterization of deep subsurface groundwaters (Geomikro)	VTT
34	2011-2014	Saline fluids, gases and microbes in crystalline bedrock (SALAMI)	GTK
35	2011-2014	Deep bedrock bioinformatics (GEOBIOINFO)	HY (2011-2012), Aalto (2013-2014)
36	2011-2014	Risks of microbiologically influenced corrosion in the Finnish nuclear waste repository	VTT
37	2013-2014	Microbial communities in the Gas Generation experiment	VTT
38	2011-2014	Use of empirical data to improve radioecological modeling applied to risk assessment of radioactive waste	ISY
		Social science studies related to nuclear waste management	
39	2012-2014	International Socio-Technical and Safety Challenges for Implementing Geological Disposal of Spent Nuclear Fuel - Finland and EU - FInSOTEC-2012-2014	JY

* no abstract in Chapter 6.

Annex 2

KYT2014 publications and theses 2011–2014

Articles in peer reviewed journals[9]

Aaltonen, P., Yagodzinskyy, Y., Saukkonen, T., Kilpeläinen, S., Tuomisto, F. & Hänninen, H. 2014. Role of excessive vacancies in TGSCC of pure copper. Presented in Environmental Damage under Static and Cyclic Loads in Structural Metallic Materials at Ambient Temperatures. Bergamo, Italy, 15 - 20 June 2014. Accepted for publication in Corrosion Reviews.

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Ganchenkova, M. G., Yagodzinskyy, Y. N., Borodin, V. A. & Hänninen, H. E. 2014. Effects of hydrogen and impurities on void nucleation in copper: simulation point of view. Philosophical Magazine, 94 (2014), 31, pp. 3522-3548.

Harjupatana, T., Alaraudanjoki, J. & Kataja, M. 2015. An X-ray tomographic method for measuring three-dimensional deformations and liquid transport in swelling clays. Accepted to be published in Applied Clay Science.

Huittinen, N., Rabung, Th., Schnurr, A., Hakanen, M., Lehto, J. & Geckeis, H. 2012. New insight into Cm(III) interaction with kaolinite - influence of mineral dissolution, Geochimica et Cosmochimica Acta 99 (2012) 100-109.

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KYT2014 Kansallinen ydinjätehuollon tutkimusohjelma 2011-2014 - Loppuraportti

Tiivistelmä | Referat | Abstract

Kansallinen ydinjätehuollon tutkimusohjelma KYT2014 käynnistyi vuonna 2011. Tutkimuskausi päättyi vuoden 2014 lopussa.

Tutkimusohjelman keskeiset aihepiirit ovat ydinjätehuollon uudet ja vaihtoehtoiset teknologiat, ydinjätehuollon turvallisuuden tutkimus ja ydinjätehuoltoon liittyvä yhteiskuntatieteellinen tutkimus. Ydinjätteiden loppusijoituksen turvallisuuden arvioinnissa on neljä osa-aluetta, jotka ovat turvallisuusperustelu, puskuri- ja täyteaineiden toimintakyky, kapselin pitkäaikaiskestävyys ja muut turvallisuustutkimukset.

Tutkimuskaudella on ollut käynnissä noin 40 tutkimushanketta, jotka ovat ensisijaisesti liittyneet ydinjätehuollon turvallisuuden arviointiin. Valtion ydinjätehuoltorahasto ohjasi rahaa tutkimushankkeisiin yhteensä noin 7 miljoonaa euroa.

KYT2014-tutkimusohjelmassa ovat olleet mukana seuraavat tutkimusorganisaatiot: VTT, Aalto-yliopiston teknillinen korkeakoulu, Helsingin yliopiston Radiokemian laboratorio, Geologian tutkimuskeskus, Jyväskylän yliopisto, Itä-Suomen yliopiston Kuopion kampus, Ludus Mundi Oy, Numerola Oy ja Tmi Pawel Simbierowicz.

Tutkimusohjelman työskentely on perustunut tutkimusohjelman johtoryhmän, kolmen tukiryhmän, koordinaattorin ja tutkimushankkeiden keskinäiseen yhteistyöhön ja työnjakoon. Tutkimusohjelman johtoryhmässä ovat olleet edustettuina sosiaali- ja terveysministeriö, Säteilyturvakeskus, työ- ja elinkeinoministeriö, ympäristöministeriö, Fennovoima Oy, Fortum Power and Heat Oy, Posiva Oy ja Teollisuuden Voima Oyj.

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KYT2014 Nationellt forskningsprogram om kärnavfallshantering 2011-2014 - Slutrapport

Tiivistelmä | Referat | Abstract

Det nationella forskningsprogrammet om kärnavfallshantering (KYT2014) startade 2011. Forskningsperioden slutade vid utgången av 2014.

Forskningsprogrammets centrala temaområden är ny och alternativ teknik för kärnavfallshanteringen, forskning i kärnavfallshanteringens säkerhet samt den samhällsvetenskapliga forskningen kring kärnavfallshanteringen. I bedömningen av säkerheten vid slutförvaring av kärnavfall ingår fyra delar vilka är funktionsförmågan hos buffertar och återfyllnadsmaterial, kapselns hållbarhet på sikt, säkerhetsbevisning och andra forskning i säkerhet.

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 KYT2014: VTT, Aalto-universitetets tekniska högskola, Helsingfors universitets radiokemiska laboratorium, Geologiska forskningscentralen, Jyväskylä universitet, Östra Finlands universitets campus i Kuopio, Ludus Mundi Oy, Numerola Oy och Tmi Pawel Simbierowicz.

Arbetet inom forskningsprogrammet har grundat sig på det ömsesidiga samarbetet och arbetsfördelningen mellan forskningsprogrammets ledningsgrupp, tre stödgrupper, samordnare och olika forskningsprojekt. I forskningsprogrammets ledningsgrupp har ingått företrädare för social- och hälsovårdsministeriet, Strålsäkerhetscentralen, arbets- och näringsministeriet, miljöministeriet, Fennovoima Oy, Fortum Power and Heat Oy, Posiva Oy och Teollisuuden Voima Oyj.

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KYT2014 Finnish Research Programme on Nuclear Waste Management 2011–2014 – Final Report

The Finnish Research Programme on Nuclear Waste Management, KYT2014, took place between 2011 and 2014. Key themes of the research programme include new and alternative nuclear waste management technologies, research into the safety of nuclear waste management and sociological research related to nuclear waste management. The assessment of the safety of final nuclear waste disposal comprises four sectors: safety case, capacity of buffer and filler materials, long-term integrity of the final disposal canister, other research on safety.

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 KYT2014 research programme presents the programme's objectives, organisation and research projects.

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