MOBILITY MISSION REPORT

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KLIKNĚTE NEBO KLEPNĚTE SEM A ZADEJTE TEXT.

MISSION TITLE

Training Course: Geochemical and Reactive Transport Modelling for Geological Disposal

DESCRIPTION

Concerned organisations

- Research entities
- Technical support organisations
- Waste management organisations

Concerned infrastructures or facilities

- High-performance computing
- University

Concerned phases

- Phase 1: Site evaluation and site selection
- Phase 2: Site characterisation

Themes and topics

Theme 3: Engineered barrier system (EBS) properties, function and long-term performance

- Clay-based backfills, plugs and seals
- Cementitious-based backfills, plugs and seals
- EBS system understanding

Keywords

Reactive transport codes; geochemical modelling.

EXECUTIVE SUMMARY

Geological disposal systems for radioactive waste and spent fuel are based on multi-barrier principle in which different engineered and natural materials act complementary to ensure the repository safety. The interaction between the different materials in the near field of a disposal system (both the engineered barrier system and the host rock) will induce geochemical changes of these materials as them are not in geochemical equilibrium.

The geochemical evolution as a consequence of physical and chemical perturbations needs to be part of the safety and performance analyses of the repository as it will influence (i) the durability of the different materials, and (ii) the speciation and mobility of radionuclides.

Given the complexity of systems and time scales involved (ten thousand to hundred thousand years), assessing the evolution can only be done by numerical modelling in which geochemistry is linked to transport using reactive transport codes. Geochemical models calculate geochemical state variables based on thermodynamic equilibrium and kinetic processes accounting for processes as aqueous speciation reactions, dissolution/precipitation based on saturation state, sorption based on mechanistic sorption models (exchange reactions, surface complexation) and possible kinetic processes.

The theoretical basis is enhanced by lectures on (i) principles of geochemical and reactive transport modelling, (ii) their applications for processes and evolution of materials in a geological repository (cementitious materials, glass, steel, clay, granite) (iii) speciation and migration of radionuclides, and (iv) advanced topics related to uncertainty and machine learning.

Practical skills are improved by computer sessions in which participants will use available software to implement and analyze models for calculating properties and evolution of materials and speciation of radionuclides.

1. MISSION BACKGROUND

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1.1. R&D background

I would first like to shortly describe my experience in using the geochemical software. Therefore, I use PHREEQC geochemical software for modelling the hydration process of cement-based material, based on PHREEQC geochemical software capacity to perform thermodynamic equilibrium calculations by solving the law of mass action (LMA) and GEMS software capacity to minimizing the Gibbs free energy of the system and also using an appropriate thermodynamic database, hydration products formation and precipitation were thermodynamically predicted for a commercial cement type using its oxide composition in powder form. To build the model which simulates the hydration kinetic phase and assemblage over time phase, the method developed by Parrott and Killoh was used and integrated in PHREEQC.

We know that the most important properties of cementitious materials such as workability, setting behaviour, strength development, but also durability are related to the cement hydration process, especially early hydration. But to understand, describe and predict the long-term performance and service life of cement-based materials, it is needed to perform the simulation and modelling of some processes, such as corrosion, dissolution, degradation and anothers.

As the training course covered lectures on different topics related to geochemical and reactive transport modelling, including the simulation and modelling of such degradation processes, my participation in this course gave me the opportunity to grow and develop my knowledge in field of geochemical and coupled reactive transport modelling in the framework of disposal of radioactive waste, which is a very useful tool for demonstrating the safety function associated with cementitious engineered barriers in a radioactive waste repository.

1.2. Mission objectives

As the geochemical evolution of the engineering barrier system of a radioactive waste repository is important in view of the safety and performance analyses of the repository and given the time scales involved (ten thousand to hundred thousand years), it is clear that assessing the evolution can only be done with numerical models in which geochemistry is linked to transport, thus with reactive transport codes.

The fact that the training course covered lectures related to geochemical and reactive transport modelling, such as: modelling geochemical systems, modelling slow processes (corrosion, dissolution, degradation), modelling properties and geochemistry of cementitious systems, geochemistry of the host rock and natural barrier materials, speciation of radionuclides, sorption of radionuclides, reactive transport modelling, will allow me have a better understanding of the principles of geochemical thermodynamic and also enlarge the knowledge and expertise in kinetic modelling and reactive transport modelling.

As I already had some minimal knowledge of operating the PHREEQC code, my attention during the course focused on the modelling with the help of this software of the processes related to cementitious materials (ordinary Portland cement): hardening,

geochemistry and other properties, chemical degradation/alteration, leaching or carbonation as an important aspect in the framework of disposal of radioactive waste and also in understanding and predicting the long-term evolution and performance of repository engineering barrier system materials.

1.3. Mission request

Financial support from EURAD Project for my attending this training course

1.4. Mission composition

Host organisation

Paul Scherrer Institut (PSI) with support of EURAD WP13 and University of Bern

Host facility

Institute of Geological Sciences- University of Bern, Switzerland

Mission dates

6 – 10 February 2023

2. MAJOR PRACTICES, TECHNIQUES, METHODS, TOOLS OR SYSTEMS OPERATED OR STUDIED

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2.1. Practice, technique, method, tool or system operated or studied during the mission

Carbonation in air of a simple hydrated cement system

Description

The practical exercise consisted in the calculation of the phase changes in 150g HPC by adding 50g of CO2 in 200 steps. It was assumed that the process takes place in the air.

Usage

The model was created following:

1. Create Single System – SysEq: - Create a New Project;
   - Select Data Bases relevant for your system;
   - Choose the elements you will have in your system;
   - Set the system parameters (pressure, temperature);
   - Enter the recipe;
   - Check the system and calculate equilibrium.

2. Make Process: - Create a new process file;
   - Select the single file you want to build your process file on;
   - Make a new process: - Name your process file;
     - Make sequential changes in the composition;
     - Select the items to sample/plot;
     - Establish the number of steps we want to take in the process file and amount of output;
   - Calculate and plot.

Benefits

Calculation of the phase changes generated by the carbonation.

Limitations

We cannot obtain a kinetic of the cement phases, as the code does not allow to keep constant the amount of CO2 for a period of time.

Applicability

Being able to model degradation processes such as carbonation will allow me to understand and predict the evolution of cementitious materials as part of the engineered barrier system of a future radioactive waste repository and at the same time to bring evidences to demonstrate the safety function associated with cementitious materials in support of implementation the programs for LILW-LL and CANDU spent fuel.

2.2. Practice, technique, method, tool or system operated or studied during the mission

Leaching of simplified cement paste in water at 25 degree Celsium

Description

The practical exercise consisted of calculating the mass changes of the phases in 150g of HPC during 100 g of H2O percolating the HPC in 100 steps.

Usage

The model was created following:

1. Clone a new record from Single System – SysRq previously created

2. Make Process: - Create a new process file;
   - Select the single file you want to build your process file on;
   - Make a new process: - Name your process file;
     - Make sequential changes in the composition;
     - Select the items to sample/plot;
     - Establish the number of steps we want to take in the process file and amount of output;
   - Calculate and plot.

Benefits

Estimation of the mass changes of the phases in the cement paste generated by the penetration of water into the system.

Limitations

We cannot obtain a kinetic evolution of the cement phases since the code does not allow to keep a constant amount of water for a period of time.

Applicability

Being able to model degradation processes such as carbonation will allow me to understand and predict the evolution of cementitious materials as part of the engineered barrier system of a future radioactive waste repository and at the same time to bring evidences to demonstrate the safety function associated with cementitious materials in support of implementation the programs for LILW-LL and CANDU spent fuel.

2.3. Practice, technique, method, tool or system operated or studied during the mission

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Description

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Usage

Klikněte nebo klepněte sem a zadejte text.

Benefits

Klikněte nebo klepněte sem a zadejte text.

Limitations

Klikněte nebo klepněte sem a zadejte text.

Applicability

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2.4. Practice, technique, method, tool or system operated or studied during the mission

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Description

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Usage

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Benefits

Klikněte nebo klepněte sem a zadejte text.

Limitations

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Applicability

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3. MISSION FINDINGS AND CONCLUSIONS

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3.1. Lessons learned and conclusions

Even if I initially thought that I would use the PHREEQC code, I came to the conclusion that for modelling the processes related to cement chemistry the most suitable code would be GEMS, which is why I followed the practical sessions dedicated to it.

Participating in this training course allowed me to gain the necessary knowledge that is the basis of thermodynamic modelling of geochemical processes, as well as of reactive transport, in the context of radioactive waste disposal systems. At the same time it allowed me to become acquainted and to develop and deepen my knowledge in the operation and running of some geochemical modelling codes through the hands on section. This consisted in useful practical exercises such as geochemical modelling of cementitious systems (modelling of slow processes such as chemical degradation (carbonation, leaching), modelling of properties and geochemistry of cement systems, geochemistry of the host rock and constituent materials of natural barriers, radionuclide speciation and uranium sorption on clay materials, reactive transport modelling.

This experience will allow me to understand the evolution of cementitious materials and at the same time to asses and predict their durability and to demonstrate the safety function associated with the materials (mortars and concretes) used in the construction of the engineering barrier system of the Romanian near-surface repository (under licensing process). Also, the knowleage accumulated during this training course will be applied in the support of ANDR (National Agency for Radioactive Waste) in the development process of the Romanian geological repository for LILW-LL and CANDU SF.

3.2. Relevant findings and conclusions for home organisation

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3.3. Relevant findings and conclusions for host organisation

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3.4. Relevant findings and conclusions for other organisations

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4. POTENTIALS FOR IMPROVEMENT OR DEVELOPMENT

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4.1. Generic potentials

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4.2. Potentials for home organisation

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4.3. Potentials for host organisation

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APPENDICES

Mission journal

Day 1
Mr. D. Jacques (SCK CEN Belgium), S. Churakov (PSI, Switzerland) welcomed us and officially opened the course, which began with a briefly presentation of the participants. The lecture series was opened by Mr. D. Kulik (PSI) who introduced us to geochemical modeling – equilibrium, thermodynamics and the evolution of chemical reactions. Three codes used by participants during the whole week were presented: GEMS (D. Kulik), ORCHESTRA (H. Meeussen), HPx/PHREEQC (D. Jacques). The second lecture of the day was given by Mrs. B. Lothenbach (EMPA, Switzerland) who spoke about thermodynamic modeling of cement-based systems. The day ended with the first hands-on session where the participants worked with one of the three codes, depending on their option.

Day 2
The day started with the presentation of Mr. E.C. Gaucher (University of Bern, Switzerland) related to host rock geochemistry in the context of geological disposal. This presentation was followed by a new practical session in which the participants simulated and carried out a series of geochemical modelling (cement carbonation, leaching). The next theoretical presentation of the iCP code was given by Ms. E. Coene (Amphos 21, Spain), followed by another practical session and a lecture presentation by E. Laloy (SCK CEN) about reactive transport. The day ended with the presentation of Crunch geochemical modeling code, focused on reactive transport modeling.

Day 3
This day day started with a presentation on radionuclide speciation given by Mr. Ulrich Mayer (UBC, Canada), followed by the practical session. The next presentation was given by Mr. Sergey Churakov regarding molecular aspect and thermodynamic modelling of sorption phenomena. After another practical session, the Min3P calculation code was introduced by Mr. Ulrich Mayer.

Day 4
This day was opened by Mr. Laurent de Windt (Mines Paris, France) with a presentation on the modeling of kinetically controlled processes in radioactive waste repositories. Mr. De Windt also introduced Hytec, a computational code for reactive transport modeling. A hands-on session in which participants performed advanced geochemical and reactive transport modeling took place also in this day. After this session, Mr. J. Samper (CICA, Spain) presented Core2 computing code used for the transient flow, heat transport and contaminant transport both in local chemical equilibrium conditions and in kinetic conditions, in heterogeneous and anisotropic porous and fracturated media. The day ended with a practical session.

Day 5
The day started with a practical session followed by Mr. Javier Samper' presentation on sensitivity analyzes and uncertainties. The last presentation on the concept of machine learning for accelerating reactive transport model simulations and analysis was given by Mr. Nikoloas Prasianakis (PSI). The last practical session was dedicated to clarifing specific issues, after which a short closing ceremony of this course took place.

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MISSION BENEFICIARY

Ionuț Cosmin FLOREA
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Radioactive Waste Treatment and Conditioning Department
Institute for Nuclear Research Pitești (RATEN ICN), Romania

PARTNER EXPERTS CONTRIBUTING TO THE MISSION

Host organisation experts

- Diederik Jacques, Head of Unit Engineered and Geosystem Analysis at SCK CEN, Belgium;
- Sergey V. Churakov, Head of Laboratory of Waste Management in the Department of Nuclear Energy and Safety at the PSI and Full Professor in the Institute of Geological Sciences at the University of Bern, Switzerland;
- Dmitrii Kulik, Senior Scientist at LES PSI, Switzerland
- Hans Meeussen, Nuclear Research Group Petten & Wageningen University, The Netherlands.

Home organisation experts

- Crina Maria Bucur, Head of RATEN ICN R&D Programme on RW and SF Management, Institute for Nuclear Research Pitești (RATEN ICN), Romania.

Other organisations experts

- Barbara Lothenbach, group leader of the Cement Chemistry and Thermodynamics Group of the Concrete & Asphalt Laboratory at Empa, the Swiss Federal Institute for Materials Science & Technology ;
- Emilie Coene, Senior Consultant of the Materials Engineering team at Amphos 21 Consulting S.L., Spain ;
- Eric Laloy, works with the Engineered and Geosystem Analysis unit at SCK CEN ;
- Javier Samper, University of A Coruña, Spain ;
- Laurent De Windt, senior researcher in applied geochemistry in the Geosciences Department at Mines Paris (France) ;
- Ulrich Mayer, professor of hydrogeology at the University of British Columbia (UBC) in the Department of Earth, Ocean and Atmospheric Sciences ;
- Eric C. Gaucher is an expert geochemist working at the University of Bern.

REPORT APPROVAL

Date	Beneficiary	Home mentor/supervisor	Host mentor/supervisor
Date of last signee	Ionuț Cosmin Florea	Crina Maria Bucur	Diederik Jacques
			Visa