European Joint Programme on Radioactive Waste Management

MOBILITY MISSION REPORT

This work has been partially supported by the EURAD project that has received funding from H2020-EURATOM 1.2 under grant agreement ID 847593.

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MISSION TITLE

Participitation in Geochemical and Reactive Transport Modelling for Geological Disposal course.

DESCRIPTION

Concerned organisations

UJV Rez, a.s., Czech Republic (home institution of participitant)
University of Bern, Bern, Switzerland (provided infrastructure for course (S. Churakov))
WP13 EURAD (organizer)
SKC CEN Belgium, Mol, Belgium (organizer, course lead by D. Jacques)

Concerned infrastructures or facilities

Student Hall, Instute of Geology, University of Bern, Baltzerstrasse 3, Bern, Switzerland

Concerned phases

Themes and topics

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

  - Spent Fuel and high-level waste disposal canisters
  - Containers for long-lived intermediate and low level wastes
  - Clay-based backfills, plugs and seals
  - Cementitious-based backfills, plugs and seals
  - Salt backfills
  - EBS system understanding
- Theme 4: Geoscience to understand rock properties, radionuclide transport and long-term geological evolution
  - Long-term stability (uplift, erosion and tectonics)
  - Perturbations (gas, temperature and chemistry)
  - Aqueous pathways and radionuclide migration

Keywords

geochemical transport modelling; reactive transport modelling; geological disposal, training course, EURAD

EXECUTIVE SUMMARY

As a junior researcher at UJV Rez in the Czech Republic, I have been actively participating in experiments connected to EURAD FuTURE. As a crucial part of the lab work, I consider not only obtaining relevant data but also being able to use the correct models to interpret obtained data and to understand the occurring processes fully. That is the reason for attending the Geochemical and Reactive Transport Modelling course organized by EURAD, to get a better understanding of processes behind geochemical modelling and to get better at using the PHREEQC code. Successful attendance in an intensive course focusing mainly on PHREEQC modelling of the different scenarios regarding the geological disposal processes helped me see how to implement PHREEQC knowledge in my experimental work.

1. MISSION BACKGROUND

1.1. R&D background

UJV Řež is involved in Task 2.2. FUTURE WP, where sorption of selected elements (Ni, Cs and others) has been observed on fissure infill and host rock as a part of a natural barrier. As a junior researcher, I am part of the team which has to acknowledge know-how about the issues concerning the WP FuTURE. However, there have been issues which required not only empirical approaches but also implementing modelling of the processes occurring during the experiments, as a part of a group involved in the sorption programme for FUTURE WP that resulted in my master's diploma work Migration of nickel in barrier materials. After that, I continued to do sorption experiments with Ni and Cs on calcite and clay fissure infill. Evaluating results a more thorough education and training in geochemical modelling might help to evaluate the results, distinguishing between sorption and precipitation in the case of calcite or understanding the process of sorption on different sites of selected clay fissure infill for Cs. I have a basic knowledge of PHREEQC, and intensive training would be beneficiary to get a more profound understanding and an overview of the modelling. Moreover, it would also be helpful in the last phase of FUTURE WP in evaluating sorption results for the final reporting. As the training is a whole week event, there was no planned funding for such a case in the UJV budget.

1.2. Mission objectives

The attendance on Geochemical and Reactive transport modelling course organized by EURAD to deeply understand PHREEQC code modelling, especially of more complex problems, and to convert new gain knowledge and skills to the modelling of practical systems in active participation in experiments done in WP FuTURE.

1.3. Mission request

To attend the Geochemical and Reactive transport modelling course organized by EURAD, I requested a EURAD Mobility grant to cover travelling expenses, as attendance at this training was not planned within the WP FuTURE budget in UJV.

1.4. Mission composition

Host organisation

EURAD WP13

Host facility

Institute of Geology, University of Bern, Baltzerstrasse 3, Bern, Switzerland

Mission dates

06 February 2023 – 10 February 2023

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

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

HPxGeochemistry (PHREEQC) modelling.

Description

HPxGeochemistry environment and his advatages for geochemical modelling.

Usage

HPxGeochemistry has been used as the interface between geochemical calculation code and reactive transport. It referred to coupling with PHREEQC to the HYDRUS, and understanding the user interface and its possibilities is crucial to obtain reasonable results from our model geochemical problems.

Benefits

HPxGeochemistry could help me, to simplify the basic input code for PHREEQC and also to fully understand the meanings of the different keywords, syntax of origin PHREEQC is, by most users, used in the programming language C++, so for beginners without the basics in computer coding, It could be a problem to enhance the potential of PHREEQC for geochemical issues fully. HPxGeochemistry has a much more friendly user interface. As an interface between geochemical calculation and reactive transport, calculations done in HPxGeochemistry could eventually be used for reactive transport in HYDRUS, because of HPxGeochemistry is coupled with It.

Limitations

Nevertheless, HPxGeochemistry is free to use, in order to be able to use it, You have to download the HYDRUS which has free trail for limited period of time but after that It is paid application.

Applicability

HPxGeochemistry could help me, to simplify the basic input code for PHREEQC and also to fully understand the meanings of the different keywords, syntax of original PHREEQC.

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

Cement Hydration – Primitive and Simple system – Ca-Si system modelling

Description

During the course at the hand-on session number 2 and 3, I tried to calculate the pore water composition, solid phase composition and some other properties (e.g. solid, gel volume etc.)

Usage

Hydration of cement (compositions of pore water, solid phase) is one the crucial modelling problems to overcome during the overall prediction of radionuclides transport in deep geological repositories.

Benefits

We started with primitive system containing only Ca and Si, which helped us to understand the simpler model of cement (containing only portlandite) and then to get on the more complex model of cement hydratation with more solid solutions and solid phases.

Limitations

It's only simplified model of cement so It can be used only in some very special occasions and not fully represent the actuall composition of cement material which will be presented in the actual repository.

Applicability

At our department at UJV Řež, we are focusing on all of the transport properties in barrier system of deep geological repository, including the cement barrier, so gain information about the calculation of pore water and solid phase composition could be tranferred to modelling similar outputs for our tested cement materials.

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

Uranium speciation modelling

Description

As an element with very complex aqueous chemistry, Uranium could exist in various complexes and compounds, dependent on pH and the amount of carbonate present, which could enhance/reduce its retention. So to fully understand data for sorption experiments, U speciation should be well-known from geochemical calculations done by PHREEQC.

Usage

Calculating the speciation of any elements, essential in assessing deep geological repositories, is crucial to understanding the processes during their transport.

Benefits

Undestanding the calculation of a Uranium speciation could, with some changes, be transferable to other elements, systems etc. Uranium speciation under different pH could be graphically interpreted for better visualization.

Limitations

Some stability constants for the elements' possible compounds or complexes are not listed in available thermodynamic databases, or their values are only robust approximations.

Applicability

Speciation calculation could be beneficial in our department, where we work with various radionuclides (e.g. 63Ni, 134Cs, 133Ba, etc.) in different systems where we need to know the exact elements' speciations to understand the values from the diffusion and sorption experiments.

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

Uranium sorption on clay minerals modelling

Description

We have done sorption of Uranium on clay mineral (montmorillonite), where we observed distribution coefficients under different pH (sorption edges) or various initial concentrations (isotherms) and also tried to differentiate the impact of presented sites on Uranium sorption.

Usage

Sorption is one of the processes which could affect the transport of leaking radionuclides in the deep geological repository by one of the barriers. Clay or bentonite barrier is projected because of its properties as swelling or high sorption capacities to retent leaking elements. As a part of the models used in prediction for real in situ environments, one crucial parameter is distribution coefficients from sorption experiments. Not only that but also, for a deeper understanding of the sorption processes, the fundamental knowledge of sorption is needed. One of the possibilities is to use modelling to explain experimental data from sorption experiments successfully.

Benefits

Using PHREEQC to model sorption distribution coefficient data for different initial uranium concentrations could explain the sorption capacities for various sorption sites on clay minerals for uranium species.

Limitations

Information about the uranium and aqueous phase is needed, and it is also required to know the properties of the mineral phase, like cation exchange capacity, amount of different sorption sites, etc.

Applicability

At our department, we systematically have been doing sorption experiments with various elements and materials to be able to model the experimental data and to have better insight into which minerals or phases could have much higher sorption capacity for chosen elements.

3. MISSION FINDINGS AND CONCLUSIONS

3.1. Lessons learned and conclusions

The geochemical and reactive transport modelling course provides deep insight into PHREEQC codes, different scenarios of issues connected to real waste repositories and how to solve them, e.g. hydration of cement, U speciation, and U sorption of Its transport through clay mineral. Sessions in the form of lectures teach the basics needed for the theoretical understanding of modelling, and that was continuously followed up, in my case, on to the PHREEQC session governed by D.Jacques as a crucial part of the course showed the versatility of possible modelling problems and, with practical examples, showing its applicability for issues connected to the assessment of geological disposal as mentioned earlier. I learned to write code in the HPxGeochemistry interface, Its connection to HYDRUS and how it used PHREEQC code for modelling. Also, I learned about the meaning of every step in our calculations, which leads to successful output containing essential variables (e.g. concentrations etc.). It was well explained, which helped me fully understand the processes during the modelling step-by-step to the successful end in the form of a graph or text output. Overall, I would like to build up on the lessons learned during the course, especially during the hands-on sessions and use known codes to implement some of their features in a way to the models for my experimental work in UJV Rez.

3.2. Relevant findings and conclusions for home organisation

3.3. Relevant findings and conclusions for host organisation

3.4. Relevant findings and conclusions for other organisations

4. POTENTIALS FOR IMPROVEMENT OR DEVELOPMENT

4.1. Generic potentials

The geochemical and reactive transport modelling course organized by EURAD was very well structured and organized, and the lessons were very educational and informative and hands-on sessions. The potential overall of the course could be to expand the number of issues, not concentrating on the cement and clay minerals but also adding the problems of other types of rock surroundings. Another could be to focus on a kind of code, in my case, on the PHREEQC and expand the lessons and hands-on sessions.

4.2. Potentials for home organisation

After the course, I would like to implement some of the modellings in my usual experimental work in UJV to understand the whole occurring processes better.

4.3. Potentials for host organisation

APPENDICES

Mission journal

The course started at 9 am at Monday in Student Hall of University of Bern.

Monday (6.2.2023) –

start – 9:00 – Welcome invitation presentation – Round Table by D.Jacques, S.Churakov

9:30 – Lecture 1 Thermodynamics – general information on basic understanding of equilibrium, Thermodynamics, Reaction Progress (database)

10:30 – 12:00 – Explanation and some examples of codes GEMS by D. Kulik, Orchestra by H. Meeussen and PHREEQC by D.Jacques

14:00 – 15:00 – Lecture 2 Thermodynamic modelling of cementitious systems and their evolution by B.Lothenbach (PSI)

15:00 – 17:30 – Hands on session 1 – Introduction of HPx and HPGeochemistry for geochemical modelling using PHREEQC by D.Jacques

Tuesday (7.2.2023) –

9:00 – 10:00 – Lecture 3 Geochemistry of the host rock and natural barrier materialby Eric C. Gaucher

10:00 – 12:30 – Hands on session 2 – Modelling of Cement Hydration – Primitive system – Ca-Si system leading by D. Jacques

14 :00 – 16 :30 – Hands on session 3 - – Modelling of Cement Hydration – Primitive system – Ca-Si system leading by D. Jacques

16 :30 – 17 :30 – Lecture 4 Reactive transport

Wednesday (8.2.2023) –

9:00 – 10:00 – Lecture 5 Radionulide speciation modelling

10:00 – 12:30 – Hands on session 4 - Modelling of Cement system – Simple system – Hydration leading by D. Jacques

13 :30 – 14 :30 – Lecture 6 - Molecular aspect and thermodynamic modelling of sorption phemonena by S.Churakov

14:30 – 17:00 - Hands on session 5 - Modelling of Cement system – Simple system – Hydration leading by D. Jacques

Thursday (9.2.2023) –

9:00 – 10:00 – Lecture 7 Modelling of kinetically controlled processes in radioactive waste disposal, from radiolytic corrision to microbial activity by L. De Windt

10:00 – 12:30 – Hands on session 6 – Modelling of U speciation by D. Jacques

14:30 – 17:00 – Hands on session 7 - Modelling of U speciation and U sorption on clay minerals by D. Jacques

Friday (10.2.2023) –

9:00 – 10:00 – Hands on session 8 – Reactive transport model for U in clay in HYDRUS and PHREEQC by D. Jacques

11:30 – 12:30 – Lecture 8 Integration of processes at larger scale – sensitivity (uncertainty) analyses by J. Samper

14:30 – 15:30 – Lecture 9 Machine learning for accelerating reactive transport model simulations and analysis by N. Prasianakis

15:30 – 17:00 – Hands on session 9 - Reactive transport model for U in clay in HYDRUS and PHREEQC by D. Jacques

17:00 – 17:30 – Closing cermony

Mission bibliography

Leal A.M.M., Kulik D.A., Smith W.R., Saar M.O. (2017): An overview of computational methods for chemical equilibrium and kinetics calculations for geochemical and reactive transport modeling. Pure and Applied Chemistry 89, 597-643.

Jacques, D., et al., *The HPx software for multicomponent reactive transport during variably-saturated flow: Recent developments and applications.* JOURNAL OF HYDROLOGY AND HYDROMECHANICS, 2018. **66**(2): p. 211-226.

Parkhurst, D.L. and L. Wissmeier, *PhreeqcRM: A reaction module for transport simulators based on the geochemical model PHREEQC.* Advances in Water Resources, 2015. **83**(0): p. 176-189.

Lothenbach, B., *Thermodynamic equilibrium calculations in cementitious systems.* Materials and Structures, 2010: p. DOI 10.1617/s11527-010-9592-x.

MISSION BENEFICIARY

Karol Kočan
Junior researcher
Disposal Processes and Safety Department
UJV Rez, a.s., Czech Republic

PARTNER EXPERTS CONTRIBUTING TO THE MISSION

Host organisation experts

Didierik Jacques, Head of Unit Engineered and Geosystem Analysis Belgian Nuclear Research Centre (SCK-CEN)

Dmitrii Kulik, Paul Scherrer institute (PSI), Switzerland

Hans Meeussen, Nuclear Research Group Petten and Wageningen University, Netherlands

Sergey Churakov, Head of Laboratory for Waste Management in PSI, Switzerland

Home organisation experts

Václava Havlová, head of the Disposal Processes and Safety Department until 31.12.2022, UJV Rez, a.s., Czech Republic

Petr Večerník, head od the Disposal Processes and Safety Department from 1.1.2023, UJV Rez, a.s., Czech Republic

Filip Jankovský, senior researcher, Disposal Processes and Safety Department, UJV Rez, a. s., Czech Republic

Milan Zuna, senior researcher, Disposal Processes and Safety Department, UJV Rez, a. s., Czech Republic

Other organisations experts

REPORT APPROVAL

Date	Beneficiary	Home mentor/supervisor	Host mentor/supervisor
Date of last signee	Karol Kočan	Petr Večerník	Didierik Jacques
	Kočan	Ing. Petr Večerník, Ph.D. Digitálně podepsal Ing. Petr Večerník, Ph.D. Datum: 2023.02.22 11:30:08 +01'00'	Diederik Jacques Digitally signed by Diederik Jacques Date: 2023.02.24 15:57:03 +01'00'

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