Deliverable 15.4: Updated Training materials

Work Package ConCorD

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Disclaimer

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Executive Summary

This training course is based on the work inside the EURAD project CONtainer CORrosion under Disposal conditions (ConCorD). It aims to provide an overview of the evolution of the environmental conditions focused on the near-field and their impact on corrosion behaviour with a focus on irradiated-accelerated corrosion, microbial activity and degradation during nearfield transients. Furthermore, a detailed overview of novel technological concepts for container materials is provided. The final part of the course gives a theoretical and practical overview of prediction tools for assessment of long-time barrier integrity and the integration of corrosion phenomena in performance assessments. This part is separated into a specific part about corrosion modelling of carbon steel embedded in bentonite with a reactive transport model: coupling or not an electrochemical model with bentonite-iron geochemistry and a more general part discussing prediction tools for assessment of long-time barrier integrity.

The outline and learning outcomes for each of the topics is given in this report. In general, the target audience for all lectures are people that already have a basic background in corrosion but want to learn more details about a topic outside their field. For example, beginning PhD students/scientists in the field. This enables people to gain a broad basic background in studying corrosion processes related to nuclear waste disposal. In principle, the slides can be used on a stand-alone basis to learn the main principles; however, more subtle information will be gained if the slides are taught by a lecturer. The slides are attached as annexes.

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1. Introduction

Even though the feasibility and safety of established container solutions has been demonstrated, recent progress in the understanding of materials and processes has shown that optimisation of container manufacturing and performance is possible. Widespread interest in repository optimisation exists and relevant projects are ongoing, e.g., WP HITEC and the HotBENT experiment in Grimsel. The systematic exploration of novel materials, while placing existing solutions in a broader context, will provide a solid state-of-the-art for the pursuit of container optimisation according to available geology, disposal concept and regulatory requirements. The testing of alternative container materials, their mechanical structural response and corrosion resistance will also bring new insights into novel technical solutions for container design.

To further increase the knowledge base and reduce remaining uncertainties, WP ConCorD aims to:

- Explore the potential of novel/advanced container materials and processes for optimisation of container performance within the engineered barrier system.
- Increase/extend the understanding of complex/coupled interfacial processes influencing container performance under repository relevant conditions, with a focus on irradiation-accelerated corrosion, microbial activity and degradation during nearfield transients, at varying scales.
- Mechanistic process understanding and development of predictive models, which will incorporate system variability and will lead to improved performance assessments addressing identified safety needs

During and after the end of ConCorD various national programs have been able to evaluate cooperatively the long-term behaviour of container materials to such an extent as to be able to ensure confidence in the performance of engineered barriers. Such progress will particularly benefit early-stage programmes and those with a small inventory. An Expert Review Group has provided guidance on the scientific work performed. Emphasis was given to issues relevant to end user needs and repository implementation, while ensuring that the generation of information is implementable and useful for performance assessment.

This document contains the content and learning outcomes of training materials developed based on the SotA (deliverable 15.1) and the results obtained within the ConCorD project. Training materials are in the form of lectures divided over five topics related to the ConCorD project. In general, the target audience for all lectures are people that already have a basic background in corrosion but want to learn more details about a topic outside their field. For example, beginning PhD students/scientist in the field. This enables people to gain a broad basic background in studying corrosion processes related to nuclear waste disposal. In principle, the slides can be used on a stand-alone basis to learn the main principles; however, more subtle information will be gained if the slides are taught by a lecturer.

1.1 Environmental factors altering the corrosion behaviour

1.1.1 Content of the presentation

The corrosion of container materials has been extensively studied under constant conditions. However, the environment is not constant but the evolution of the chemistry of the repository environment in the period after its closure will depend mainly on two factors: the engineered barriers and the composition of groundwater. After facility closure, the thermal phase of the development of the repository is important, with a typical example being the saturation rate of the bentonite barrier leading to swelling and the development of mechanical stresses on the container. Such transients can be further influenced by attempts at repository footprint optimisation (e.g., increased container heat production). The corrosion of container materials has been usually studied under constant conditions and the translation of the experimental result to the evolving chemical, mechanical and redox conditions of the early post-closure period needs to be verified and complemented. The different transients and their impact on the corrosion behaviour are discussed in the lectures.

1.1.2 Learning outcomes

Following this presentation, people are able to:

- Understand the function of the waste container in the disposal of HLW/SF
- Sum up the different container concepts & expected exposure conditions
- Identify the different transient process in the near field environment
- Comprehend the effect of transient processes on the corrosion behavior of waste containers

1.1.3 Further reading

- https://www.ejp-eurad.eu/publications/eurad-d151-concord-initial-sota
- Fernández A.M., Alonso U., Nieto P., Mingarro M, Missana T., Muñoz A., Finck N., Müller N., Schild D., Singh A.R., Šachlová S., Kašpar V., Dobrev D., Gõtz D., Kolomá K., Vecerník P., Zuna M., Matulová M. (2024): Final version as of 29.04.2024 of deliverable D15.12 of the HORIZON 2020 project EURAD. EC Grant agreement no: 847593.
- Dixon, D. (2019). Review of the THMC Properties of MX-80 Bentonite, NWMO-TR-2019-07
- King, F. (2017). 13 - Nuclear waste canister materials: Corrosion behavior and long-term performance in geological repository systems. Geological Repository Systems for Safe Disposal of Spent Nuclear Fuels and Radioactive Waste (Second Edition). M. J. Apted and J. Ahn, Woodhead Publishing**:** 365-408
- Fraser King, 2017 - 13 - Nuclear waste canister materials: Corrosion behavior and long-term performance in geological repository systems - doi.org/10.1016/B978-0-08-100642-9.00013-X
- Landolt, D., A. W. Davenport, J. H. Payer and D. W. Shoesmith (2011). A Review of Materials and Corrosion Issues Regarding Canisters for Disposal of Spent Fuel and High-Level Waste in Opalinus Clay. *ChemInform* 42

1.2 Radiation-induced corrosion

1.21 Content of the presentation

An essential inquiry arising in predicting the lifespan of copper and carbon steel containers within a GDF concerns the influence of radiation on corrosion behaviour. Currently, there is no consensus on the extent of an additional margin required to accommodate an increase in corrosion rate after the buffer reaches saturation due to radiation emitted by the decaying waste package. It remains unclear whether dose rate or total dose predominates as the crucial factor for consideration. This presentation provides an overview of anticipated radiation dose rates during geological disposal, the impact of radiation on copper and carbon steel, and the experiments conducted within ConCorD.

1.2.1 Learning outcomes

Following this presentation, people are able to:

- Understand the effect of radiation on copper and carbon steel
- Sum up the factors that play a role impact on corrosion behavior under irradiation
- Comprehend the challenges related to radiation studies

1.1.1 Further reading

- https://www.ejp-eurad.eu/publications/eurad-d151-concord-initial-sota
- Clayton Bevas (Jacobs), James Hesketh (Jacobs), Andy Rance (Jacobs), Leigh-Anne Stevenson (Jacobs), Shorubhi Uthayakumaran (Jacobs) Ben Pateman (Jacobs), Cristiano Padovani (Jacobs), Šárka Šachlová (UJV), Vlastislav Kašpar (UJV), David Dobrev (UJV), Daniel Götz (UJV), Kateřina Kolomá (UJV), Petr Večerník (UJV). (2024): Elucidation of the effect of radiation on the corrosion of canister materials. Final version as of 02/02/2024 of deliverable D15.7 of the HORIZON 2020 project EURAD. EC Grant agreement no: 847593
- Allard, T., E. Balan, G. Calas, C. Fourdrin, E. Morichon and S. Sorieul (2012). Radiation-induced defects in clay minerals: A review. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* 277: 112-120
- King, F. and M. Behazin (2021). A Review of the Effect of Irradiation on the Corrosion of Copper-Coated Used Fuel Containers. *Corrosion and Materials Degradation* 2: 678-707

1.3 Microbial activity and degradation during nearfield transients

1.21 Content of the presentation

Microbially influenced corrosion can accelerate anoxic corrosion due to microbial metabolism. MIC can also play a role in the high-level waste and spent fuel repository environment, even if biofilm formation on the surface of canisters is not expected (owing to the high initial thermal load and radiation field), nor yet observed in relevant experiments. However, it is well established that H_2 will be produced via anoxic corrosion of some container materials. The H_2 produced could drive microbial processes such as sulphate reduction and methanogenesis. The associated sulphide production in the former could affect the corrosion rate, particularly for copper but potentially also for steel containers. It has already been demonstrated that sulphate-reducing bacteria are present in the host rock at most repository locations considered, as well as in potential backfill materials. Thus, the question is not whether sulphate reduction can occur in the repository, but whether it can be inhibited or controlled in a way that minimizes the impact on the corrosion rate of the container materials in the repository. The bentonite buffer is expected to play a key role in precluding microbial activity to limit any negative microbial impact on corrosion rates. Thus, it is critical to optimally select and design the bentonite buffer to inhibit or control microbial activity. In this presentation, the current status on research related to microbially influenced corrosion is discussed.

1.3.1 Learning outcomes

Following this presentation, people are able to:

- Understand the process of microbially influenced corrosion
- Comprehend factors that alter microbial process
- Get acquainted with novel experimental strategies to study microbial processes in bentonite

1.1.2 Further reading

- https://www.ejp-eurad.eu/publications/eurad-d151-concord-initial-sota
- Mar Morales Hidalgo, Cristina Povedano Priego, Marcos F. Martinez Moreno, Fadwa Jroundi, Mohamed L. Merroun, Ursula Alonso, Ana María Fernández, Miguel García-Gutiérrez, Manuel Mingarro, Tiziana Missana Jesus Morejón, Paula Nieto, Pedro P. Valdivieso, Deepa Shree

Bartak, Jakub Ríha, Katerina Cerná, Šárka Šachlová, Vlastislav Kašpar, David Dobrev, Petr Vecerník, Natalia Jakus, Eliot Jermann, Pierre Bena, Christina Zarkali, Rizlan Bernier-Latmani, Ting-Shyang Wei, Vladyslav Sushko, Andrea Cherkouk, Carla Smolders, Bruno Kursten, Kristel Mijnendonckx (2024): Integration of the findings on the impact of irradiation, dry density and particle size on the microbial community. Final version as of 04/04/2024 of deliverable D15.9 of the HORIZON 2020 project EURAD. EC Grant agreement no: 847593.

- Jakus, N., Vivek, P., Mischler, S., Bernier-Latmani, R. (2024): Deliverable D15.10: Impact of bentonite-associated oxygen on microbial activity and viability. Final version as of 07/05/2024 of deliverable D15.10 of the HORIZON 2020 project EURAD. EC Grant agreement no: 847593.
- Enning, D. and J. Garrelfs (2014). Corrosion of iron by sulfate-reducing bacteria: new views of an old problem. *Appl Environ Microbiol* 80: 1226-1236

1.2 Novel material solutions for nuclear waste disposal container concepts

1.2.1 Content of the presentation

The understanding of degradation mechanisms and resulting container durability estimates is mature and has been demonstrated for existing disposal concepts envisaging the use of copper (e.g. Sweden, Finland, Canada) and carbon steel in a clay environment (e.g. France, Switzerland, Japan, Czech Republic). These materials are called “traditional materials” but are also the ones on which “corrosion allowance” designs are based, i.e. materials that corrode slowly and in a uniform and predictable manner. The second type of materials under consideration are the “novel materials”, which are typically subject to very low corrosion rates. For the most promising novel materials, the planned experimental studies contributed to the understanding of corrosion/degradation processes. The work was organised according to the two main types of material classes: ceramic and metallic materials. In the case of ceramic materials, the experimental work was focused on studying materials and processes (deposition, sealing) for innovative solutions of ceramic containers and ceramic coatings on steel. For containers made of bulk ceramics, the key properties for long-term performance are fracture toughness and leaching/alteration. For thin coatings, a primary concern would be the presence of through-porosity or flaws. For metallic materials, the main aim of the experimental work was to study and consider the implementation of innovative solutions for metallic anti-corrosion coatings or optimised bulk metallic options. Studies focused on material development (alloy composition, coating technology) and initial corrosion tests to identify the benefit of these new container solutions. The lecture provides an overview of different “novel materials” under consideration and corrosion processes are discussed in more detail.

1.2.2 Learning outcomes

Following this presentation, people are able to:

- Understand the advantages & disadvantages of the use of ceramic/metallic materials in nuclear waste disposal
- Identify the current knowledge gaps of the use of ceramic/metallic materials in nuclear waste disposal
- Give an historical overview of the use of ceramic materials in nuclear waste disposal
- Provide an overview of the contribution of ConCorD to the knowledge on novel material solutions for nuclear waste containers

1.2.3 Further reading

- https://www.ejp-eurad.eu/publications/eurad-d151-concord-initial-sota
- Gaggiano, R., Debelle, A., Rossignol F., Cornu, I., Bosch, C., Ganster, P., Dias, R. De C. C. D., Sayenko, S., Kuprin, A., Trentin, A., Vaari J., Pakarinen, J. (2024): Deliverable 15.6: Novel materials and processes for the optimisation of long-term container performance. Final version as of 06.05.2024 of deliverable D15.6 of the HORIZON 2020 project EURAD. EC Grant agreement no: 847593.
- Holdsworth, S.R., 2013. Ceramic Material Solutions for Nuclear Waste Disposal Canisters. NAGRA report NAB12-45.
- Holdsworth, S.R. 2018. Alternative coating Materials as Corrosion Barriers for SF and HLW Disposal Canisters. NAGRA report NAB18-19.
- INTERNATIONAL ATOMIC ENERGY AGENCY (2006). Development of Specifications for Radioactive Waste Packages. IAEA-TECDOC-1515. IAEA, Vienna

1.3 Prediction tools for assessment of long-time barrier integrity

1.3.1 Content of the presentation

The disposal of high-level radioactive waste (HLW) and spent nuclear fuel (SF) poses a unique challenge in predicting the long-term performance of corroding structures. This challenge stems from the expectation that HLW/SF containers may need to endure for one million years or more in certain scenarios. Over the past 45 years, a range of empirical and deterministic models has been developed to forecast long-term corrosion behavior. These models encompass predictions for uniform and localized corrosion, environmentally assisted cracking, microbiologically influenced corrosion, and radiation-induced corrosion. More recently, fracture mechanics-based approaches have emerged to address joint mechanical-corrosion degradation modes. Regardless of the choice between empirical or deterministic models, it remains imperative to demonstrate a comprehensive mechanistic understanding of the corrosion processes involved. Alongside process models that target specific corrosion mechanisms, there exists a demand for performance-assessment models as integral components of demonstrating the safety of a deep geological repository. In the first set of slides, an overview is given of the nature of a disposal environment and its evolution followed by different process models categorized by corrosion processes. Finally, performance assessment models are discussed categorized by material and the concept of confidence building is explained. The second set of slides presents a more detailed overview of corrosion modelling of carbon steel embedded in bentonite with a reactive transport model; coupling or not an electrochemical model with bentonite-iron geochemistry.

1.3.2 Learning outcomes

Following this presentation, people are able to:

- Identify principles of different modelling approaches for several corrosion processes
- Understand advantages & disadvantages of different modelling tools
- Compare processes relevant for copper & steel containers
- Understand how corrosion processes can be integrated in performance assessments (PA)
- Get acquainted with basic modelling tools
- Comprehend the concept of confidence building

1.3.3 Further reading

- https://www.ejp-eurad.eu/publications/eurad-d151-concord-initial-sota
- King, F. (2014). "Predicting the Lifetimes of Nuclear Waste Containers." JOM 66(3).
- King, F., M. Kolář, S. Briggs, M. Behazin, P. Keech, N. Diomidis (2024). "Review of the modelling of corrosion processes and lifetime prediction for HLW/SF containers – Part 1: Process models." Corros. Mater. Degrad. 5, 124-199.
- King, F., M. Kolář, S. Briggs, M. Behazin, P. Keech, N. Diomidis (2024). "Review of the modelling of corrosion processes and lifetime prediction for HLW/SF containers – Part 2: Performance assessment models." Corros. Mater. Degrad., submitted.
- Nagra. (2002). Demonstration of disposal feasibility for spent fuel, vitrified high-level waste and long-lived intermediate-level waste. Nagra Report NTB 02-05.
- Performance Assessment for the Proposed High-Level Radioactive Waste Repository at Yucca Mountain, Nevada https://www.sciencedirect.com/journal/reliability-engineering-and-system-safety/vol/122/suppl/C
- Svensk Kärnbränslehantering, A. (2010). Corrosion calculations report for the safety assessment SR-Site. SKB TR-10-66

Appendix A. Environmental factors altering the corrosion behaviour

Appendix B. Radiation-induced corrosion

Appendix C. Microbial activity and degradation during near field transients

Appendix D. Novel material solutions for nuclear waste disposal container concepts

Appendix E. Prediction tools for assessment of long-time barrier integrity

Appendix F. Corrosion modeling of carbon steel in bentonite with iCP: coupling of an electrochemical model in COMSOL with the bentonite-iron chemical system in PhreeqC