

Seminar 2 – Waste management, decommissioning and disposal – Session 2

Chaired by F. Sentuc (GRS) / E. Balibas (LEI)

13:45 - 14:30 | No. 205

Sorption parameter uncertainty propagation in reactive transport modeling using the example of cesium diffusion in clay

W. Pfingsten (PSI), A. Ayoub (ETHZ), L. Podofillini (PSI) and G. Sansavini (ETHZ)

The quantification of parameter uncertainty is of major interest for modelling radionuclide migration from nuclear waste repositories in performance assessment studies. Of special interest is the quantification of uncertainties of sorption parameters and its influence on the prediction of radionuclides' concentration levels, arrival times and migration distances in clays. Up to now, however, the parameter uncertainty for sorbing radionuclides was limited to simple k_d sorption approaches, with focus on the radionuclides' k_d values and diffusion coefficients (see e.g. [1, 2]). Depending on the uncertainty of these two parameters, a range of arrival times are calculated. Here, we use a complex three-site sorption model for Cs described in [3] within a multi-species reactive transport model (MCOTAC, [4]) in combination with a two-step sensitivity analysis, the Morris method [5] followed by an extensive Sobol sensitivity analysis [6]. The Morris analysis quantifies the average effect of a parameter on the output of interest, whereas the Sobol analysis quantifies the relative contribution of a parameter to the variance of the output, which yields a ranking of uncertain parameters' importance. The implementation of reactive transport calculations into a probabilistic framework allows the quantification and ranking of the sensitivity to sorption parameters of MCOTAC for Cs diffusion in clay using reference illite sorption parameters. These uncertain parameters can be divided into two groups of different origin. One group refers to the uncertainty of sorption model parameters for the specific equilibrium cation exchange reactions (constant in time); the second group refers to the uncertainty of major cations concentrations involved in the cation exchange reactions, whereas this porewater composition might be time dependent, when the geochemical system is evolving, especially for long-term radionuclide migration. Multiple realizations (>20000) of Cs breakthrough curves calculations are evaluated showing a variation of Cs arrival times for different diffusion distances due to sorption parameters and cation concentration uncertainties. From initially 12 assumed uncertain parameters, the analysis yields major influence of equilibrium sorption reaction constants of Cs on Type2 and FES sites, in addition to the concentrations of respective cations involved in these reactions (Na^+ and K^+) on the diffusion of Cs through the clay.

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Moreover, these results enable quantitative statements and measures about the -up to now performed- qualitatively described importance of the considered sorption parameters. With these multiple realisations of Cs breakthrough curves, a classification tree is constructed, showing the combination of parameters values leading to early Cs arrival times and maximum Cs concentrations at specific locations. In addition, the multiple realisations of the Cs breakthrough curves, yield ratios of values for Cs sorbed and Cs in solution, i.e. a “complex Cs sorption model isotherm”, which includes all the uncertainties of the considered sorption model parameters and major cations concentrations. These isotherms are state-of-the-art, more accurate, look-up table for modelling Cs sorption-diffusion in reference illite and can be applied for simple k_d approach calculations in performance assessment studies. Finally, the new approach presented here identifies a roadmap for how and what has to be done to quantify the importance of each uncertain parameter in reactive transport modelling. The developed knowledge can be used in other complex reactive transport problems with respect to parameter uncertainty and related influence on migration fronts of contaminants, precipitation or dissolution fronts etc. To this aim, if the computed breakthrough curves are affected by large uncertainty or variability, parameter uncertainty analysis can help in designing laboratory experiments with respect to geometrical (e.g. sample thickness), and geochemical set-up (major cation concentrations chosen) in order to produce informative data and reduce uncertainty.

[1] Hedin, A., *Reliability Engineering & System Safety*, 79(2): 195-204 (2003).

[2] Trincherio, P. et al., *Computers & Geosciences*, 86, 55-63 (2016).

[3] Bradbury, M.H., Baeyens, B., *J. Cont. Hyd.*, 42(2): 141-163 (2000).

[4] Pfingsten, W., Paul Scherrer Institut, Villigen, Switzerland, PSI-Report 94-15 (1994).

[5] Morris, M.D., *Technometrics*, 33(2): 161-174 (1991).

[6] Sobol, I.M., *Mathematical modelling and computational experiments*, 1(4): 407-414 (1993)

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14:30 - 15:00 | No. 206

Reduction of the activation of an accelerator used for radio-isotopes production: all benefits for the dismantling

F. Schmitz (Bel V) and C. Kennes (Bel V)

In Belgium, the regulator, the Federal Agency for Nuclear Control, may delegate some of its tasks to its subsidiary Bel V. These tasks include, inspections of class I and class IIa facilities (class IIa is a subcategory of class II). Class IIa are mainly consisting of cyclotrons and irradiators. In Belgium there are almost twenty cyclotrons. Among them some were shut down more than 30 years ago, arising the question of their dismantling and about the preventive measures that can be taken on still operating machines or under installation to decrease the level of activation. This document lists the preventive measures that can be adopted and makes explicit some of the characteristics of the accelerator's operations that should be taken into account when considering its dismantling.

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15:00 - 15:30 | No. 207

Research and development support to nuclear decommissioning and environmental remediation at JRC

P. Hubert (JRC), F. Raiola (JRC), K. Abbas (JRC) and P. Peerani (JRC)

Nowadays only about 20 nuclear facilities worldwide have undergone the full decommissioning process and were released from regulatory control, while more than 125 nuclear power reactors worldwide have been already shut down and this number is expected to go up drastically in the near future. A challenge is that a large number of nuclear facilities reach the end of their useful life in the coming decades therefore the activity sector of nuclear decommissioning and waste management (ND&WM) will arise. In this context, the European Parliament required in 2012 that the Joint Research Center (JRC) builds upon its experience with the decommissioning of JRC nuclear facilities and further reinforces its research to support safe decommissioning in Europe.

Decommissioning and Environmental Remediation (D&ER) activities is the most visible contribution of what can R&D bring to the field of nuclear decommissioning. R&D activities provide scientific and technical support to ND&WM field as a whole, which is one of the EU priorities to improve standardization for a safe decommissioning and waste management, and to prepare for the vast emerging decommissioning market.

Along with Horizon 2020 EU R&D work-programme, using competences of its different sites (Ispra (Italy), Karlsruhe (Germany), Geel (Belgium) and Petten (The Netherlands)), JRC contributes in its reaching this objective through multi-annual research projects on several topics such as testing, development and validation of nuclear material characterization, standardisation of methods and materials, imaging techniques, mixed-realities and other innovative IT approaches (Innovative Technology Integration Sector of Decommissioning Unit). Under Horizon 2020, JRC has also organised and contributed to numerous education and training events in ND&WM.

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As an illustration of an example of a JRC research project in this field is ITSP that stands for Innovative Techniques/technologies and Standardisation of Practices in decommissioning. ITSP includes six research workpackages that cover most of research needs in ND&WM or D&ER such as NDA characterisation techniques, imaging techniques (gamma, laser, muons), and standardization. NDADecom (Non-destructive analysis for Decommissioning) and MetroDecom-II (In-situ metrology for decommissioning nuclear facilities) are two workpackages fully dedicated to enhance assay and imaging techniques such as tomography for waste. We will describe one of the largest automated non-destructive assay facility in JRC Ispra, which is designed for analysis and characterization of low and intermediate nuclear waste. This waste characterisation facility was commissioned at the end of 2007 by the A.N Technology Ltd. (UK), i.e. ANTECH. It comprises two measurement stations for waste drums based on gamma spectrometry and neutrons. The gamma station operates two characterisation protocols, the segmented and the tomographic scanning while the neutron station is based on passive and active neutron interrogations using a D-T neutron generator.

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Explosion safety of organic water-soluble compounds and nitric acid solutions evaporation during SNF and RW treatment

A. Rodin (SEC NRS)

SNF and RW treatment consists of extraction, sorption, and precipitation methods. The increase in the depth of fuel burnout, reduction of SNF holding time should be taken into account during deep separation of SNF and RW valuable components. It results into the radiation load and into the cocomplexity of technological processes of spent nuclear fuel cycle facilities. This also will increase the range of using compounds such as different types of extractants and diluents, reducing and complexing agents which have not been previously used.

The issue of explosion and fire hazard is an important component of ensuring nuclear and radiation safety is the solution of. The experience of radiochemical plants shows that explosions and fires lead to significant environmental impact and are the initial events of radiation accidents. The main hazards for the plants include explosions and fires during evaporation when nitric acid solutions may contain water-soluble organic compounds. The presence of reducing agents, complexing agents, and other organic compounds in nitric acid solutions creates a risk of uncontrolled heat and gas release.

Such organic compounds include reducing agents and complexing agents, such as hydrazine nitrate, carbonylhydrazide, and acetohydroxamic acid. During the extraction solutions of nitric acid and these substances are practically not dangerous, since the solutions contain low concentrations of both oxidizing and reducing agents, and they are used at low temperatures. However, when such solutions are evaporated, it is possible to increase their concentrations, resulting in the transformation from exothermic processes to the thermal explosion mode. The danger of evaporation process is evidenced by a number of incidents in production [1].

Oxidation processes were studied using thermal analysis methods. Experiments showed that acetohydroxamic acid and carbonylhydrazide can be concentrated during evaporation. As a result, exothermic reactions can potentially arise to thermal explosion accompanying by significant amount of gaseous products and heat.

1. Nazin E.R., Zachinyaev G.M., Fire and explosion safety of radiochemical production processes. M.: SEC NRS, 2009. 189 p.