Modelling of radionuclide release from a near-surface repository taking into account sorption changes due to concrete barriers degradation

Dalia Grigaliuniene[1]; Povilas Balcius[1]
Lithuanian Energy Institute, Kaunas, Lithuania

INTRODUCTION

Near-surface repositories (NSR) in many countries are considered as an appropriate disposal option for short-lived low and intermediate level radioactive waste (RAW). The safety of such repositories is based on a multi-barrier system. Cement-based materials play a significant role in the NSR barrier system. However, the engineered barriers undergo degradation due to negative environmental impact. This can lead to changes in the barrier ability to retard radionuclide (RN) migration. When performing a safety assessment of the RAW disposal facility, these changes have to be considered. A common approach is to model radionuclide sorption in cement based on a ratio between the concentration of sorbed contaminant and the concentration in solution (Kd concept). Therefore, the chemical changes in concrete can be introduced by selecting the appropriate Kd value for each concrete degradation stage and for each radionuclide under consideration.

The aim of this work is to investigate radionuclide release from a NSR, considering sorption changes due to concrete barrier degradation, applying different conceptual models, and to compare the results.

SYSTEM DESCRIPTION

The near-surface repository consists of concrete vaults filled with cemented RAW and covered with concrete slab and natural barriers of clay, sand, gravel and vegetation on the top (see Chart 1). This work considers only concrete barriers (top and bottom slabs of the vault and the waste zone) as indicated by the blue rectangle in Chart 1. Selected radionuclides: I-129, Ni-59 and Pu-239.

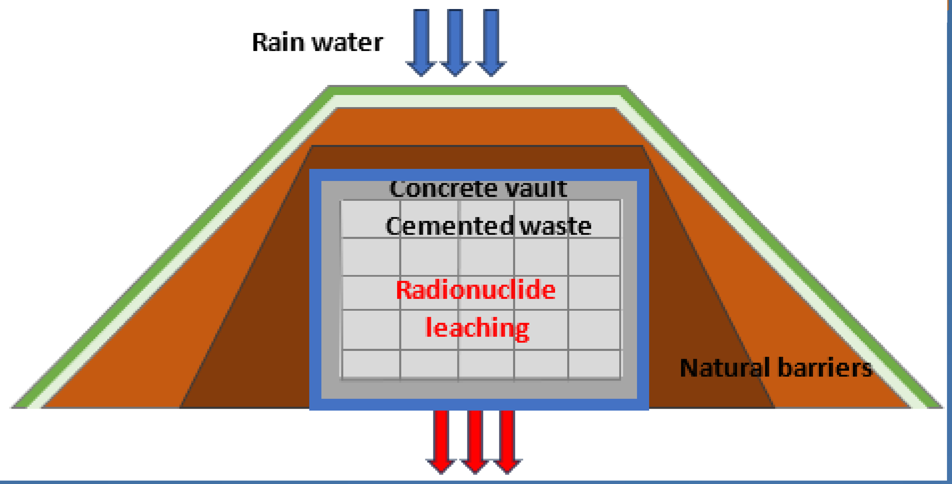

Chart 1. Conceptual model of the near-surface repository (blue rectangle indicates the region considered in this work)

RN MIGRATION SCENARIO

After the repository closure, rainwater infiltrates through the top barriers and causes degradation of concrete and leaching of radionuclides from the waste. The leached radionuclides migrate with the infiltrated water down through the repository and through the bottom slab into the environment.

MODELLING APPROACH

Assessment of the radionuclide leaching was performed in two steps. At first, concrete degradation was modelled to identify concrete degradation stages based on pH changes in the concrete. The link between pH and degradation stage is presented in Table 1. In the next step, for each degradation stage and each radionuclide under consideration, appropriate Kd values were selected (based on [1], see Table 1) and introduced in the radionuclide migration model.

Table 1. Link between concrete degradation stage and pH; and selected Kd values (based on [1, 2])

pH		>12.5	~12.5	12.5-10.5	<10.5
Concrete degradation stage		Stage 1	Stage 2	Stage 3	Stage 4
Kd (l/kg)	Ni	40	40	40 - 4	4
	I	3	3	3 - 0.3	0.3
	Pu	5000	5000	5000 - 1000	1000

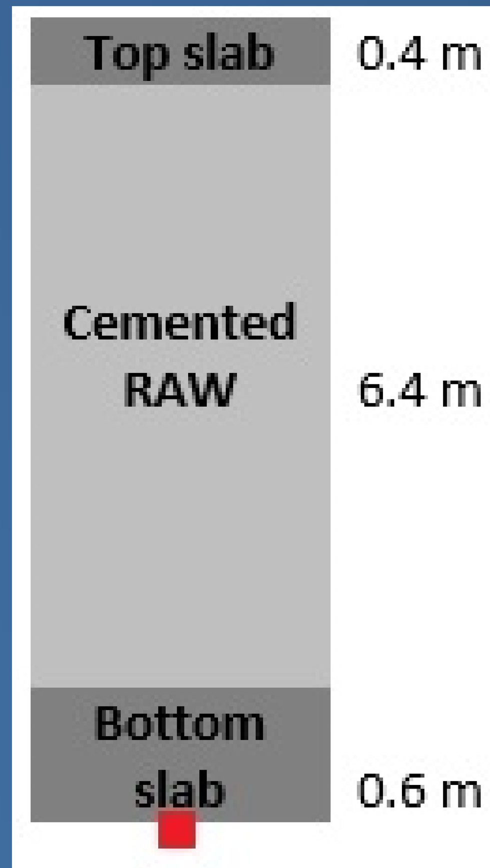

Chart 2. Conceptual model of the analysed domain with the identified location for the output of the results (red square)

The changes of Kd values can be modelled in different ways. Two conceptual models were developed for this purpose in this study:

- **Case 1**. Kd values change at the beginning of a new stage of concrete degradation (one-step change).
- **Case 2**. Kd values change from one stage to the next in multiple-steps to follow a more accurate representation of the changes in pH.

Note: Concrete degradation stage for the whole bottom slab is assigned based on the upper 1 cm zone of the slab (i.e. as soon as 1 cm of the slab enters the next stage, this stage is assigned to the whole slab). For the waste zone, due to larger dimensions, it was divided into two equal subregions, and the concrete degradation stage of each subregion was assumed based on its top 1 cm layer.

For comparison of the Case 1 and Case 2 models, relative (scaled to initial inventory) radionuclide flux into the environment is compared.

The domain of interest was modelled in 1D geometry with computer software PHAST [3] (see Chart 2). Sorption was modelled as radionuclide reaction with a surface, as described in [3].

RESULTS AND DISCUSSION

Modelling results of Step 1 are presented below in Chart 3. As an example, changes in pH over time at the top of the waste zone are provided. (Similar changes in pH were also obtained in the lower subregion of the waste zone and in the concrete bottom slab.) In addition, Chart 3 demonstrates the differences in Kd changes between Cases 1 and 2 for Ni-59.

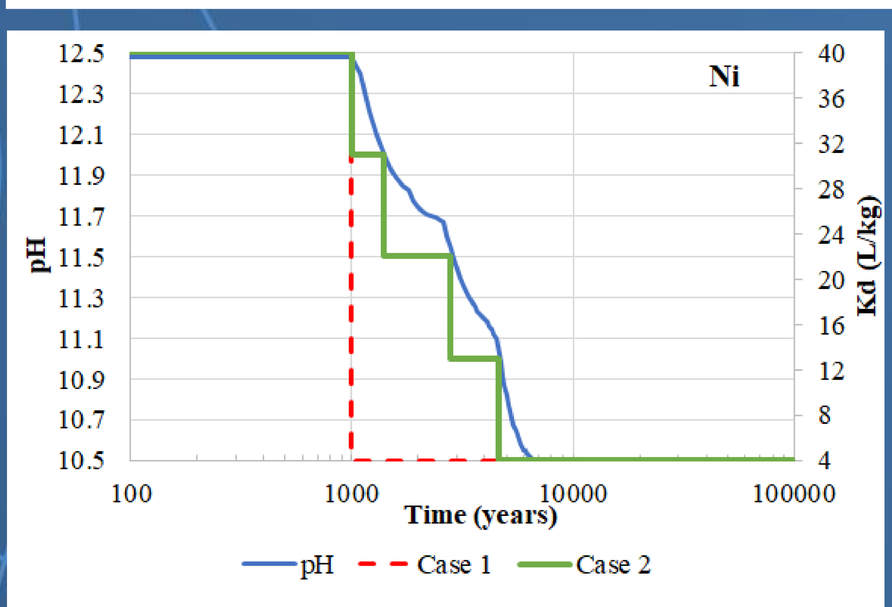

Chart 3. Evaluated pH changes over time at the top of the waste zone and difference between Cases 1 and 2 regarding Kd changes – Ni example

Chart 3 indicates that the degradation of the upper subregion of the waste zone could reach stage III in about 1000 years, and this is the time for Kd change in Case 1. During stage III, pH drops to the value of 10.5 in about 3600 years. These changes are represented in 4 steps in Case 2.

Radionuclide fluxes into the environment (scaled to initial inventory) obtained applying different approaches for Kd change are presented in Chart 4. It can be seen, that there is no difference between Cases 1 and 2 for weakly sorbed radionuclide I-129 as it is transported out of the system before concrete degradation. The limited effect of the selected approach has been identified on the flux of the medium sorbed radionuclide Ni-59. The peak of Ni-59 flux differs by about 30 % between the cases and, in Case 1, more rapid transport of Ni-59 out of the system is observed. The highest difference between Cases 1 and 2 is for the strongly sorbed radionuclide Pu-239. The peak of Pu-239 flux from the repository in Case 1 is about 3 times higher than in Case 2. However, there are periods in the timeline where the difference between the cases can be higher than one order of magnitude.

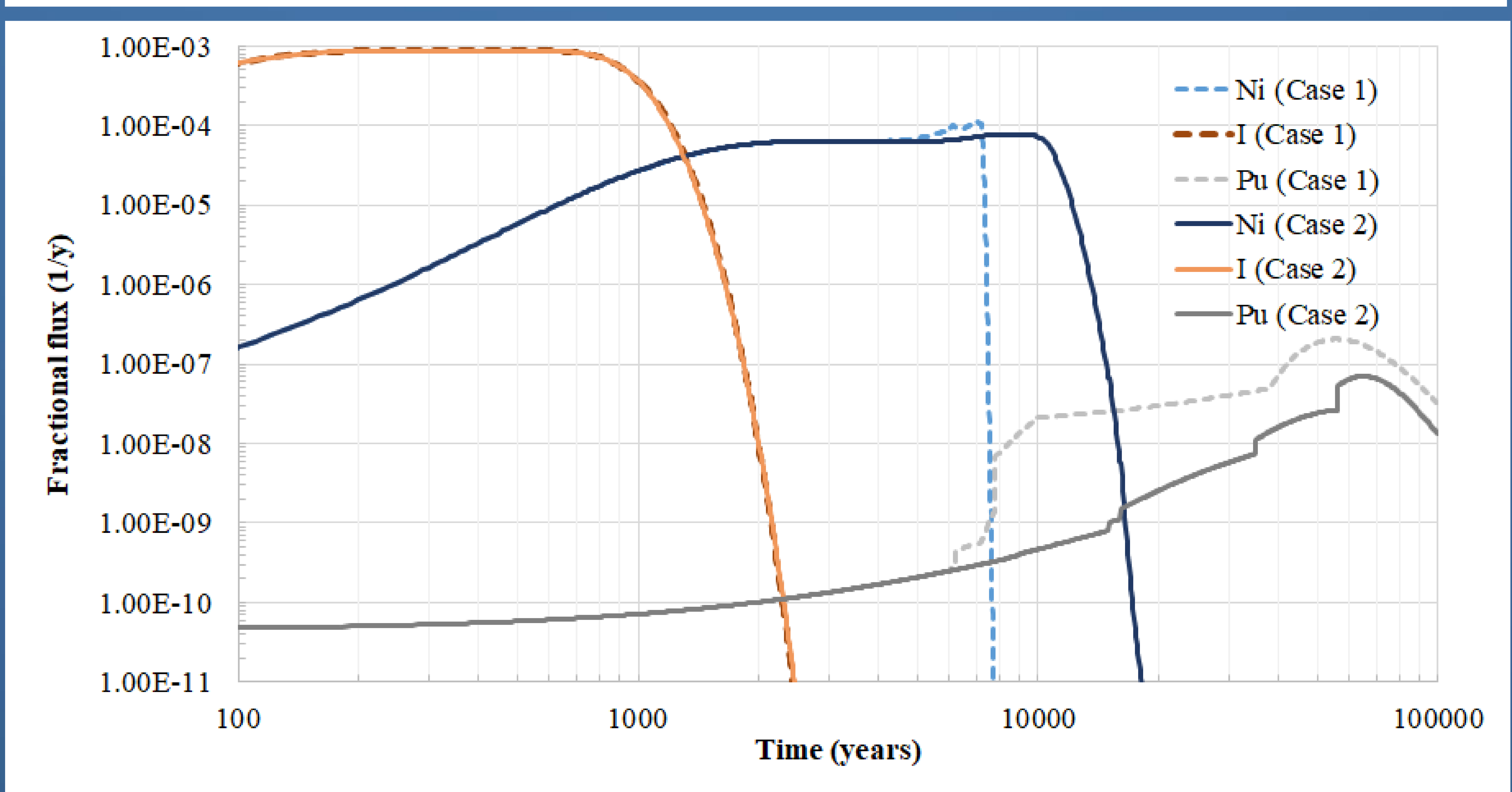

Chart 4. Radionuclide fractional flux into environment

CONCLUSIONS

1. Consideration of concrete degradation in radionuclide migration modelling as a single step change of Kd value results in a higher radionuclide flux and is conservative in comparison with the more gradual Kd change approach for medium and strongly sorbed radionuclides Ni-59 and Pu-239.
2. Consideration of concrete degradation has no significant impact on transport of the weakly sorbed radionuclide I-129.
3. The gradual Kd change approach can be used in the safety assessment of the near surface repositories to reduce conservatism in the assessment.

REFERENCES

1. Safety assessment methodologies for near surface disposal facilities: results of a co-ordinated research project. Volume 1 Review and enhancement of safety assessment approaches and tools. Vienna: IAEA, 2004. ISBN 9201040040.
2. D. Jacques et al. Modelling chemical degradation of concrete during leaching with rain and soil water types. Cement and Concrete Research. doi:10.1016/j.cemconres.2010.02.008
3. PHAST Version 2—A Program for Simulating Groundwater Flow, Solute Transport, and Multicomponent Geochemical Reactions [online]. Denver: U.S. Geological Survey, 2010. Available from: https://pubs.usgs.gov/tm/06A35/pdf/TM6-A35.pdf.

CONTACT

Dalia Grigaliuniene
Lithuanian Energy Institute
Email: dalia.grigaliuniene@lei.lt
Website: www.lei.lt

RAD CONFERENCE
THIRTEENTH INTERNATIONAL CONFERENCE ON RADIATION, NATURAL SCIENCES, MEDICINE, ENGINEERING, TECHNOLOGY AND ECOLOGY
JUNE 16-20, 2025
HUNGUEST HOTEL SUN RESORT, HERCEG NOVI, MONTENEGRO
LITHUANIAN ENERGY INSTITUTE