THE IMPACT OF SELECTED PARAMETER CHANGES ON THE UNCERTAINTIES RELATED TO MATERIALS OF GEOLOGICAL REPOSITORY

Dorota Flamíková, Dana Barátová, Vladimír Nečas

Slovak University of Technology in Bratislava, Faculty of Electrical Engineering and Information Technology, Institute of Nuclear and Physical Engineering, Ilkovičova 3, 81219 Bratislava, Slovak Republic
e-mail: dorota.flamikova@stuba.sk, dana_baratova@stuba.sk, vladimir.necas@stuba.sk

Received 12 May 2017; accepted 25 May 2017

Abstract

This paper briefly describes results of the sensitivity analysis applied on the near field of geological disposal system for spent nuclear fuel located in crystalline rocks. The impact of chosen parameter changes on the radionuclide release rates has been investigated. The calculations of release rates were carried out for one disposal container using the simulation software GoldSim.
Based on the results, same radionuclides dominate the total release rate in all cases. When comparing the sensitivity analysis results with the reference case, the most significant difference in total release rate was recorded for the non-conservative values of bentonite buffer´s distribution coefficients.

1. Multi-barrier concept of the disposal system

The multi-barrier concept is based on a combination of engineering and natural barriers that provide sufficient protection for the population and the environment against undesirable radiation over a very long period of time (thousands to millions of years).

Waste form is spent nuclear fuel or highly radioactive waste, which has undergone the process of treatment and conditioning to form a solid state product with satisfactory chemical and physical parameters.

Processed and appropriately molded nuclear waste is placed in a disposal canister, which must be designed with respect to radiation and nuclear safety during handling, transporting and final disposal. The canister also has to be designed to provide sufficient protection for the environment against radiation for several hundreds to thousands of years.

Another barrier that is used in the deep repository system is the sealing or the filling material. The filling material in most of the foreign burial concepts consists of clayey rock - called bentonite. Bentonite is a mixture of clay minerals, a group of smectite, which intensifies its volume in contact with water [1]. Suitable geological formation in combination with suitable sealing material can isolate high-level nuclear waste or spent fuel from the biosphere for a long time even after the failure of the safety function of the disposal canister.

By selecting appropriate geological formation, protection of the storage system will be ensured in particular against adverse effects that could lead to the deterioration of the integrity of engineering barriers [2].

2. Description of the reference case of disposal system

In the Slovak Republic a final locality for the geological disposal facility has not been selected yet. The operation of the deep geological repository in Slovakia is planned to start approximately in year 2065. Therefore the assessment of long-term safety for a hypothetical geological repository located in crystalline rocks was performed.

As a reference case the normal groundwater scenario was chosen. The aim of this scenario is to assess a long-term safety of deep geological disposal for spent nuclear fuel. In the normal groundwater scenario safety of the disposal system is influenced only by normal evolution processes that lead to the degradation of individual repository components.

Host rock is modelled like a fractured zone where each transport pathway has a different transmissivity. Transport pathways in crystalline rocks are represented by individual fractures and faults.

Spent fuel, disposed in geological repository, is the heterogeneous system and therefore was within the model conceptually divided into the structural material, UO_2 matrix and instant release fraction (IRF) [3]. Instant release fraction is a fraction of inventory which is after water contact released rapidly, in the term of long-term safety instantaneously. Then the long-term release occurs congruently with the degradation of the fuel matrix and structural material.

Another important barrier is disposal container, where spent fuel assemblies (from VVER-440 reactors) are considered to be disposed in. The disposal capacity of a one container is 7 fuel assemblies. This container is made of stainless steel (inner part) and carbon steel (outer part) with an outer diameter of 650 mm and a length of 3670 mm. Fuel assemblies are inserted into the inner profiled pipes made of aluminium alloy [4].

Within this assessment the calculations were performed for the spent fuel with an initial average enrichment of 4.87 % of U-235 and burnup 60 MWd/kgU. The time of storage before the final disposal was set at 60 years. Disposal container is surrounded by a bentonite buffer with a wall thickness of 300 mm.

After the disposal canister fails due to normal evolution processes (1000 years) and water comes into contact with the source term (fuel and structural material), released radionuclides start to migrate through the bentonite buffer, excavation disturbed zone (EDZ) and crystalline host rock. Concentrations of radionuclides in the void volume of the disposal container and in the bentonite buffer are limited by the solubility of each chemical element.

According to the international safety assessments and certain assumptions as high solubility limits, relatively long half-lives, poor sorption on bentonite buffer, significant IRF values, 36 radionuclides were identified as relevant for the safety analysis.

Modelling was carried out by using the simulation software GoldSim, whose Radionuclide Transport module allows users to reliably model a mass transport within the complex system of engineering and natural barriers [5].

3. Important transport processes and parameters of the near field

3.1 Sorption

Sorption is the process, where dissolved contaminants partition from the ground-water and adhere to the particles of the bentonite matrix. Sorption of the dissolved radionuclides onto the matrix results in slowing (retardation) of contaminants relative to the average advective ground-water flow velocity and a reduction in dissolved concentrations in ground water.

The rate of sorption is described by distribution coefficient K_d, which is defined as the ratio of the sorbed contaminant concentration to the dissolved contaminant concentration. For systems described by a linear isotherm, K_d is a constant [6].

$$K_d = \frac{m_S/M_s}{m_f/V_f}, \tag{1}$$

where: K_d - distribution coefficient (m^3/kg),
m_s - weight of the nuclide sorbed on the solid material (kg),
M_s - weight of the solid material (kg),
m_f - weight of the nuclide dissolved in the liquid medium (kg),
V_f - volume of the liquid medium (m^3).

3.2 Solubility

The concentration of the contaminant can be limited by its solubility. The solubility of the element depends on several factors such as the groundwater composition, temperature, pressure, etc. If the amount of the element is greater than its solubility limit, precipitations are formed. A solubility limit refers to an individual element (not nuclides). This means that the solubility of the element is divided between its isotopes based on the isotopic ratio.
Before the precipitation occurs, the nuclide must reach a saturation concentration values in a transport medium.

3.3 Diffusion

Diffusion as a transport mechanism is based on the thermal movement of molecules that migrate from areas with a higher concentration to areas with lower concentration. The diffusion is determined by the diffusion flow J, which represents the amount of the substance, which in the presence of the concentration gradient, passes through a unit area perpendicular to the direction of diffusion over a certain time unit. In a simple liquid, diffusion is governed by Fick's first law [6]:

$$J_f = -D_f \nabla c_f, \tag{2}$$

where: J_f - diffusion flux of the substance in the free water (kg/m^2.s),
D_f - diffusion coefficient of the substance in the free water (m^2/s),
c_f - mass concentration of the substance in the free water (kg/m^3),
∇ - vector operator.

In a porous environment where diffusion is complicated by geometry, the diffusion flux equation takes the form [6]:

$$J_{por} = -\varepsilon \frac{\delta}{\tau^2} D_f \nabla c_f = -\varepsilon G D_f \nabla c_f = -D_e \nabla c_f, \tag{3}$$

where: J_{por} - diffusion flux in porous medium (kg/m^2.s),
ε - porosity (-),
δ - constrictivity (-),
τ - tortuosity (-),
G - factor representing the geometrical properties of pores (-),
D_e - effective diffusion coefficient (m^2/s).

4. Sensitivity analysis of the near field parameters

In this part of the paper the analysis of simulation results, which describes the behaviour of the near field model in dependence of selected chemical parameters is processed. These results are compared with the simulation results for the reference case, which is described above.

Within the sensitivity analysis, the changes in distribution coefficients K_d, diffusion coefficients D_e, and solubility limits were investigated.

The values of the selected parameters were determined by using the international research achievements (Nagra - Switzerland) with optimistic and pessimistic scenarios application [7]. These scenarios and corresponding values of selected parameters are shown in Tab.1.

Tab. 1. *Values of selected parameters for optimistic and pessimistic scenarios [7]*

PARAMETER	K_d [m^3/kg]			D_e [m^2/s]			Solubility [mol/m^3]		
CASE/SCENARIO	reference	optimistic	pessimistic	reference	optimistic	pessimistic	reference	optimistic	pessimistic
ELEMENT									
Ac	20	300	1	2.00E-10	2.00E-11	4.00E-10	1.00E-03	5.00E-05	3.00E-02
Am	20	300	1	2.00E-10	2.00E-11	4.00E-10	1.00E-03	5.00E-05	3.00E-02
C	0.00006	0	0	3.00E-12	2.00E-11	4.00E-10	3.00E+00	6.00E-01	7
Cl	0	0	0	3.00E-12	3.00E-13	6.00E-12	-1.00E+00	-1.00E+00	-1
Cm	10	300	1	2.00E-10	2.00E-11	4.00E-10	1.00E-03	5.00E-05	3.00E-02
Cs	0.1	0.3	0.03	2.00E-10	2.00E-11	4.00E-10	-1.00E+00	-1.00E+00	-1.00E+00
I	5.00E-04	5.00E-03	5.00E-05	3.00E-12	3.00E-13	6.00E-12	-1.00E+00	-1.00E+00	-1.00E+00
Nb	30	900	1	2.00E-10	2.00E-11	4.00E-10	3.00E-02	1.00E-05	1.00E-01
Np	60	600	6	2.00E-10	2.00E-11	4.00E-10	5.00E-06	3.00E-06	1.00E-05
Pa	5	100	0.2	2.00E-10	2.00E-11	4.00E-10	1.00E-05	1.00E-05	1.00E-02
Pb	7	100	0.5	2.00E-10	2.00E-11	4.00E-10	2.00E-03	2.00E-05	8.00E-02
Pd	5	100	0.2	2.00E-10	2.00E-11	4.00E-10	5.00E-05	1.00E-07	2.00E-04
Pu	20	300	1	2.00E-10	2.00E-11	4.00E-10	5.00E-05	3.00E-06	1.00E-03
Ra	0.002	0.01	3.00E-04	2.00E-10	2.00E-11	4.00E-10	2.00E-08	4.00E-09	5.00E-05
Se	0	0	0	3.00E-12	3.00E-13	6.00E-12	5.00E-06	2.00E-08	1.00E-02
Sm	4	100	0.1	2.00E-10	2.00E-11	4.00E-10	5.00E-04	3.00E-04	9.00E-04
Sn	800	10000	1	2.00E-10	2.00E-11	4.00E-10	1.00E-05	5.00E-06	1.00E-04
Tc	60	600	0.5	2.00E-10	2.00E-11	4.00E-10	4.00E-06	1.00E-06	1.00E-05
Th	60	200	10	2.00E-10	2.00E-11	4.00E-10	7.00E-04	2.00E-04	3.00E-03
U	40	400	2	2.00E-10	2.00E-11	4.00E-10	3.00E-06	3.00E-07	5.00E-04
Zr	80	4000	1	2.00E-10	2.00E-11	4.00E-10	2.00E-06	3.00E-08	2.00E-06

To determine the impact of analysed data uncertainties on repository performance, the total maximum release rate from the geosphere was quantified for each case of interest. The case, where the total maximum release rate was higher than the reference case is called a conservative case and a case, where the total maximum release rate was lower than in the reference case is called a non-conservative case.

The results of the sensitivity analysis are summarized in Figure 1. It represents the dependence of the total release rate (activity flux per year) of considered radionuclides located in the inventory of the deep geological repository and the time after disposal of spent nuclear fuel.

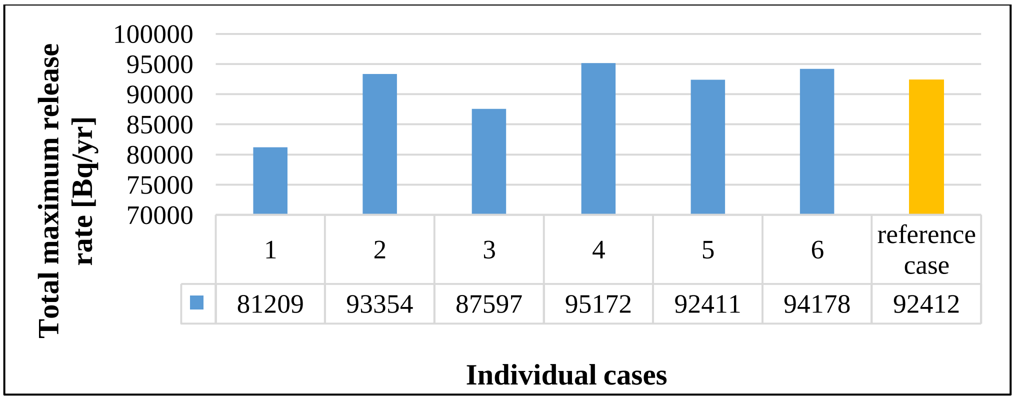

Fig. 1. *Total maximum release rate in individual cases*
Case 1-K_d parameter change: optimistic scenario, Case 2-K_d parameter change: pessimistic scenario, Case 3-D_e parameter change: optimistic scenario, Case 4-D_e parameter change: pessimistic scenario, Case 5-Solubility limit change: optimistic scenario, Case 6-Solubility limit change: pessimistic scenario, Reference case

The release rate of many nuclides is reduced due to the transport of nuclides through the host rock and the major water-conducting fault. Actinides are relatively strongly sorbed on the bentonite as well as on the host rock matrix and therefore their release rates are very low. In each case of interest, the largest contribution to total release rate from the geosphere represent activation and fission products C-14, Cl-36, I-129, Se-79 and Cs-135.
I-129 and Cl-36 are assumed to have very poor retentive properties and that's why these nuclides dominate the total release rate for a long period of time. In the early years of the analysis, the most significant nuclide dominating the total release rate is C-14. It is caused by its relatively high specific activity in comparison with long-lived radionuclides. It can be seen, that the release rates are strongly dependent on the retentive properties of individual nuclides.

We can generally say, that the most significant influence on the total maximum release rate in this analyses is caused by the change parameters related to sorption and diffusion, especially considering the optimistic scenarios. Changes in the total maximum release rates, between the conservative and non-conservative case for these parameters is about 15 % for distribution coefficient K_d, and almost 8 % for diffusion coefficient D_e.

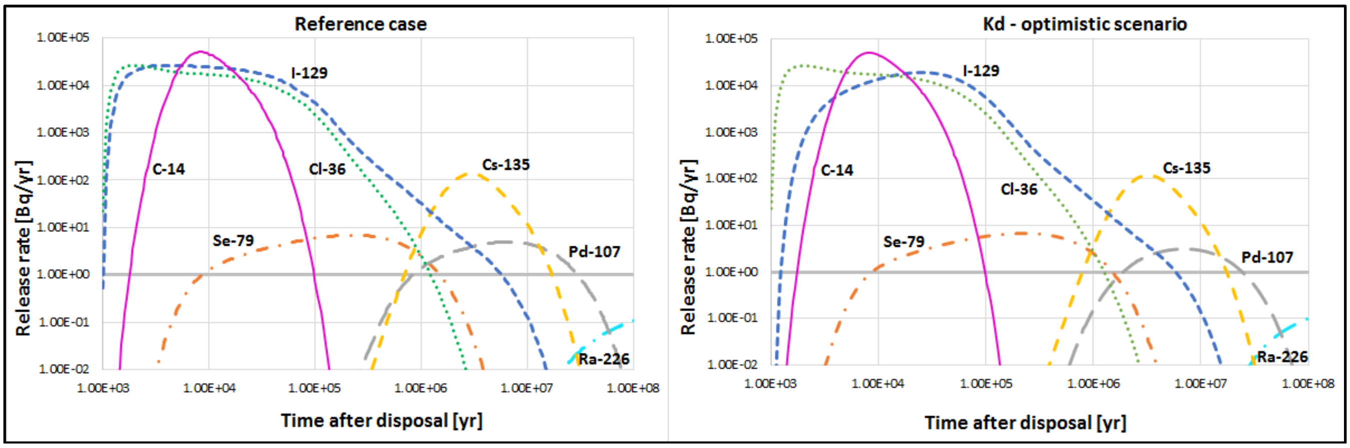

Fig. 2. *Detailed comparison of reference case and non-conservative case of K_d change*

The most significant difference in total maximum release rate compared to the reference case represents case 1, when change of distribution coefficient K_d in optimistic scenario was considered. The detailed comparison of these cases shows Figure 2. This difference is about 12 %, while in other cases is less than 5 %. This difference is caused by the change of diffusion coefficient of iodine about one order of magnitude.

Conclusion

To determine the behaviour of deep geological repository system by change of selected parameters related to transport processes, the sensitivity analyses was carried out. Based on the results of the analysis it can be said, that the value of total maximum release rate is influenced by the same radionuclides in all considered cases. The total maximum release rate was quantified for each case of interest and the differences between the conservative and non-conservative case were evaluated. Based on the results, it can be said, that performed parameter changes do not influence total release rates rapidly. The most significant influence on the total maximum release rate is caused by the parameters related to sorption and diffusion, especially considering the optimistic scenarios. The most visible difference in total maximum release rate compared to the reference case represents case, when change of distribution coefficient K_d in optimistic scenario was considered. This difference is about 12 %, while in other cases is less than 5 %. The difference is caused by the change of distribution coefficient of iodine about one order of magnitude.

Acknowledgement

This project has been supported by the Slovak Grant Agency for Science through grant VEGA 1/0863/17.

References

[1] Nuclear Decommissioning Authority. *Geological Disposal, Steps Towards Implementation*. Oxfordshire: NDA, (2010), 59 p. ISBN 978-1-84029-397-5.

[2] BARÁTOVÁ, D. Modelling of radionuclide migration from the geological repository using the simulation software GoldSim, diploma thesis, Bratislava, (2014).

[3] POINSSOT, C., FERRY, C., KELM, M., *et al.* (2004) Spent Fuel Stability under Repository Conditions - Final Report of the European Project. European Commission, (November 2001-October 2004), 104 p. CONTRACT No. FIKW-CT-2001-00192 SFS.

[4] Radioactive Waste Repository Authority (1999) Reference Project of Geological Repository: Technological Part. Czech Republic: RAWRA.

[5] GOLDSIM Technology Group LLC (2010) GoldSim Contaminant Transport Module, User`s Guide. Washington, USA.

[6] PULKKANEN, V., M., NORDMAN, H.: *Modelling of Near-Field Radionuclide Transport Phenomena in a KBS-3V Type of Repository for Nuclear Waste with GoldSim Code – and Verification Against Previous Methods.* Finland: Posiva Oy, (March 2010)

[7] PRVÁKOVÁ, S., NILSSON, K.F. (2006) Treatment of Data Uncertainty for the Modelling of Radionuclide Migration in Geological Repository. Office for Official Publications of the European Communities, Luxembourg.