MODELLING OF RADIONUCLIDE RELEASES FROM THE NEAR FIELD OF THE GEOLOGICAL REPOSITORY IN CRYSTALLINE ROCKS FOR RBMK-1500 SPENT NUCLEAR FUEL

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ABSTRACT

During 2002-2005 the assessment of possibilities for disposal of spent nuclear fuel (SNF) in Lithuania was performed with support of Swedish experts. Potential geological formations for disposal of SNF were selected, disposal concept was developed and preliminary generic safety assessment was performed. Performing safety assessment the analysis of radionuclides migration from the repository as well as their impact to human and environment were also very important issues.
In this paper results on the analysis of the radionuclide releases from the near field of the geological repository for RBMK-1500 SNF in crystalline rocks in Lithuania are presented. Radionuclide migration in the near field region was assessed using computer code COMPULINK7 (SKB, Sweden) The results of analysis show that most of safety relevant radionuclides of RBMK-1500 SNF are effectively retarded in the near field region. The release is dominated by ^{129}I and ^{59}Ni firstly while after app. 50 thousand years ^{226}Ra is dominating. The sensitivity analysis was also performed for the parameters that have direct influence on releases.

1. Introduction

During 2002-2005 the assessment of possibilities for disposal of the SNF in Lithuania was performed with the support of Swedish experts. Potential geological formations for disposal of the SNF were selected, disposal concept was developed and preliminary generic safety assessment was performed. Disposal concept for RBMK-1500 SNF in crystalline rocks in Lithuania is based on Swedish KBS-3 concept with SNF emplacement into the copper canister with cast iron insert. The bentonite and its mixture with crushed rock are also foreseen as buffer and backfill material. Detailed description of the disposal concept for RBMK-1500 SNF in crystalline rocks in Lithuania is presented in the paper presented to this conference [1].

The radionuclide transport analysis was performed for the RBMK-1500 SNF with initial enrichment of 2.8 % ^{235}U and 0.6 % Er_2O_3. The SNF burnup is app. 29 MWd/kgU, the radionuclide inventory was assessed by computer code SAS2H (computer code system SCALE 5) [2]. Radionuclide transport calculations were performed using computer code COMPULINK7 (Sweden) for the safety relevant radionuclides for RBMK-1500 SNF that were identified in [3]. For sensitivity analysis, each parameter value influencing the radionuclide release directly was varied by its variability factor and the relative change in the maximum release value indicates the sensitivity of releases to parameter uncertainties.

2. Processes in the near field

The evolution of a failed canister is complex and depends on a number of uncertain factors. Water is likely to intrude into the canister, causing corrosion of the cast iron insert with hydrogen gas generation. The build-up of gas pressure in the canister can be considerable and lead to the suppression of further water entry and also to gas release through the buffer. As corrosion proceeds, corrosion products will exert mechanical pressure on the copper canister, corrosion also causes a weakening of the cast iron insert and this could also lead to expansion of defects [4]. The evolution will also be influenced by external factors like the external mechanical load on the canister and by the thermal conditions. When the initial defect becomes larger water can also enter freely and a continuous water pathway is expected to form. The materials of fuel claddings are stable in water and corrode only very slowly, but microscopic cracks could develop eventually, forming passages through the tube walls. As there is some current discussion as to how much short term credit can be taken for the tubes as part of the multibarrier system, usually it is considered that the lifetime of the cladding is very short under anoxic repository conditions and the claddings do not provide a barrier for the transport of radionuclides released from SNF matrix. As the groundwater comes into the contact with SNF its alteration/dissolution processes and the release of radionuclides begin. Key parameters influencing SNF matrix alteration/dissolution are specific activity of SNF, primary radiolytic yields, temperature, specific surface of SNF, iron and H_2 concentration, concentration of carbonates, initial oxidation state of SNF matrix, etc. [5]. Depending on the heterogeneous distribution of radionuclides in the structure of SNF their release to groundwater is classically described by the contribution of two terms: an instantaneous and a slow long-term release of the radionuclides. As SNF matrix degrades, the radionuclides embedded in it are congruently released into the volume inside the canister. Solubility limits for dissolved radionuclides are applied and in case of radionuclide concentration exceeds solubility limit they start to precipitate. The release of dissolved radionuclides from the waste package by diffusion and/or advection depends on the conditions to be expected in the repository as well as on the properties of engineering barriers. Buffers based on the clay have very low permeability to water flow. The advective transport of the dissolved radionuclides is generally considerably smaller than that due to molecular diffusion. Dissolved radionuclides will diffuse in the water existing in the clay pores and could interact with the pore surface. Sorption term is general and includes contributions from all heterogeneous reactions of dissolved contaminants with solid surfaces: chemisorption and physisorption, precipitation, as well as ion exchange and isomorphic substitution.

3. Modelling of radionuclide transport

The radionuclide transport calculations were performed for the canister defect scenario. Taking into consideration the similarities in the repository environment and repository concept, the selection of scenario is based on experience from the safety assessment performed in Sweden [6]. For Lithuanian case it is assumed that one canister out of total 1400 will pass through quality inspection with penetrating defect of 1 mm^2 in size. For this preliminary study only the main processes determining the repository safety in the case of SNF disposal in the crystalline rocks are taken into account. Specific features and processes related to KBS-3H design indicated in [7] (e. g. a loss or redistribution of buffer mass, eventual gas bubble transport and transport of volatile radionuclides, the accumulation of gas along the top of the drift and its effects on groundwater transport, the eventual chemical alteration of bentonite, effect on rhelogical properties, swelling and hydraulic conductivity of bentonite due to steel components) have not been addressed in this preliminary study.

Due to very low permeability of the bentonite buffer, the transport of released from SNF matrix and dissolved radionuclides in the near field is going to be diffusion dominated. For radionuclide transport assessment the methodology presented in [8] was used. According to this methodology the barrier system is discretized into compartments. The material balance over the compartment connected to some other compartments for n radionuclide is expressed as follows:

$$V_i \varepsilon_i R_i^n \frac{dC_i^n}{dt} = V_i \cdot S^n + V_i \varepsilon_i R_i^{n-1} C_i^{n-1} \lambda^{n-1} - V_i \varepsilon_i R_i^n C_i^n \lambda^n - \sum_j \left(\frac{AD_e}{d} \Delta C \right)_{i,j}^n \quad (1)$$

where V - compartment volume (m^3), C - radionuclide concentration (mol/m^3), ε - material porosity (m^3/m^3), R - retardation factor (-), S - general source term, λ - decay constant (1/yr), A – compartment cross section area (m^2), D_e - diffusivity in material (m^2/yr), d - diffusion length (m). Mass transfer of dissolved species from the stagnant porewater in the bentonite into the groundwater flowing in a fracture intersecting the deposition tunnel is limited by the boundary layer resistance. The assessment of this transfer is handled through the fictitious equivalent flow rate Q_{eq} [9]:

$$Q_{eq} = 2\pi \cdot r_2 \cdot 2b_v \sqrt{\frac{4D_w u}{\pi^2 \cdot r_2}}, \quad (2)$$

where $2b_v$ is the volume aperture of the fracture (m), r_2 is the radius of the deposition tunnel (m), D_w is the diffusivity in water (m^2/yr), *u* is the velocity of water in the fracture (m/yr).

As there are no yet an experimental data on RBMK-1500 SNF dissolution under reducing conditions as well as data on instant release fraction for some nuclides, the data compiled for other UO_2 type SNF were used instead. Based on the data available in [4, 10, 11] the rate of SNF alteration/dissolution rate of 10^{-7}/yr is assumed as reference value. The structural parts of SNF assembly are assumed to be fully corroded and radiouclides only present in structural parts of SNF assembly are assumed to be fully available. It is also assumed that after the defect becomes larger the entire void in the canister at closure, approximately 0.5 m^3, will be filled with water and radionuclides released instantaneously or during SNF alteration/dissolution dissolve in the water inside in the canister. If the concentration of nuclides exceeds its solubility limit, they start precipitate. Sorption on the internal parts of the canister is neglected. Thus the solubility of radionuclides limits the concentration in the canister and the release rate of the species escaping from the canister. The solubility depends on the composition of water entering the canister which is influenced by chemical processes in the buffer material and inside the canister as well. Solubility limits may also be altered by possible changes in the redox potential in the repository by radiolysis of the water entering the canister. Since the solubility limits also depends on the conditions in the repository environment and they have not been determined for Lithuanian repository yet, the radionuclide solubility values were used from [6]. Because of the conditional nature of diffusivity, sorption coefficients, porosity, particularly of K_d, they have to be determined for the conditions expected to be relevant for the repository environment. Since these site specific parameters have not been determined yet it was assumed that the conditions of the repository environment is similar to that for other granitic rocks. Data on diffusivity, sorption that were derived based on systematic datasets available in the literature and/or on thermodynamic models and presented in [12] were used in the calculations. In the present analysis the hydraulic gradient around the deposition tunnel is assumed to be 1 % after the closure of the repository and a reference fracture intersecting tunnel is assumed to have a transmisivity of 10^{-8} m^2/s and a volume aperture of 250 μm based on [9].

4. Results

As could be seen in Fig. 1, most of identified safety relevant radionuclides for RBMK-1500 SNF disposal are effectively retarded in the near field region. The release to the far field is dominated by ^{129}I and ^{59}Ni firstly while after app. 50 thousand years ^{226}Ra is dominating. ^{226}Ra is formed in the decay chain of ^{238}U and it is more mobile than its predecessors. Detailed study of the output shows that the concentrations of the following nuclides reach their solubility limits in the canister: ^{242}Pu, ^{239}Pu, ^{238}U, ^{236}U, ^{235}U, ^{234}U, ^{233}U, ^{232}Th, ^{230}Th, ^{229}Th, ^{237}Np, ^{231}Pa, ^{79}Se, ^{93}Zr, ^{99}Tc, ^{107}Pd, ^{126}Sn.

The sensitivity analysis was also performed for the parameters that have direct influence on releases (SNF matrix dissolution rate, solubility, diffusivity, sorption coefficient). For the parameter sensitivity analysis the same methodology as presented in [13] is applied. Firstly the radionuclide release from the near field region is assessed using the reasonable values of parameters. Secondly, by varying each parameter individually by a factor according to its variability, the relative change in the performance of the reference case is calculated as the ratio of maximum release rates. For the sensitivity analysis few radionuclides have been chosen, considering the half-life, the capacity of sorption and diffusion in the backfill material (^{135}Cs, ^{129}I, ^{239}Pu, ^{238}U, ^{59}Ni, ^{226}Ra).

The lower and upper values of sorption and diffusivity coefficients reported in [12] indicate a large range for some radionuclides (e. g., actinides). A variability factor of about 5 is selected to test the parameter sensitivity. Since the range of solubility variation is of some orders of magnitude, a rough estimation of the uncertainty in the solubility data may result in the variability factor about 10 [13]. The equivalent flow rate is assumed to vary by factor 5 that could correspond to the increase of the fracture transmissivity by one order of magnitude.

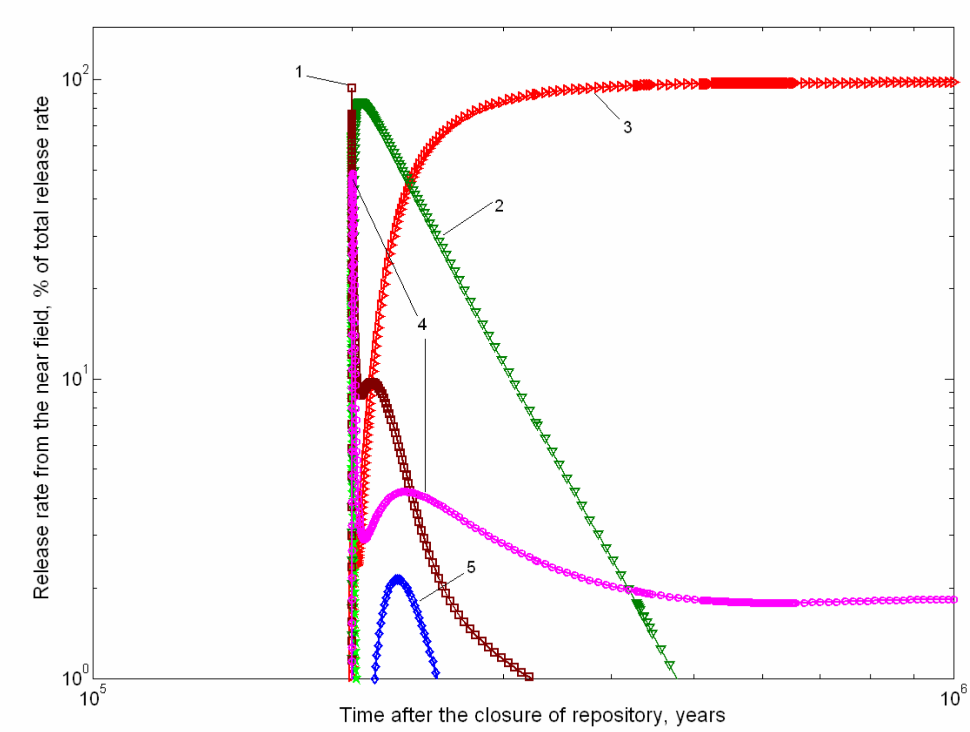

Fig. 1. Release rate from the near field (1 - ^{129}I, 2 - ^{59}Ni, 3 - ^{226}Ra, 4 - ^{135}Cs, 5 - ^{94}Nb)

Uncertainties in the fuel matrix dissolution have influence on the release of radionuclides whose concentration in the canister is not controlled by solubility limit (maximum release rate increased 4 times for ^{226}Ra). For radionuclides with instant release fraction the significance of increased fuel matrix dissolution depends on what (instant release fraction or congruent release from SNF matrix) governs the maximum release from the near field. In this reference case, increased fuel dissolution rate by factor 10 results in increased maximum release from the near field for cesium (about 4 times), while for iodine the release caused by instant release fraction (IRF=2%) becomes only comparable with part congruently released from the SNF (the maximum release increased only app. 24 %). For radionuclides whose release from the canister is controlled by solubility, the uncertainties in solubility limit have significant influence on the release (maximum release increased about 10 times for ^{238}U and about 7 times for ^{239}Pu).

The influence of uncertainties in the radionuclide diffusion coefficient is more significant when the concentration of dissolved radionuclides in the canister remains constant (e. g. limited by solubility). In this case decreased buffer resistance and increased concentration gradient results in higher release from the near field (5 times for ^{238}U, and app. 250 times for ^{239}Pu in this study). In other case increased diffusivity could result in increased release to the far field until it is controlled by buffer resistances and not by the concentration of radionuclides available to migrate (as for Ra). As diffusivity for iodine is low compared with the diffusivity for cesium, increased iodine diffusivity had an influence on increased its release from the near field. But this increase is less significant (maximum release of iodine increased 3 times) than for solubility limited radionuclides, as more rapid release from the canister results in decreased concentration and smaller concentration gradient.

Uncertainties in the sorption coefficient have a strong influence on the release of more sorbing radionuclides. Due to increased sorption sorbing radionuclides remain longer in the near field region and they have more time to decay to insignificant concentrations. Sorption capacity of the bentonite surrounding the canister and the hydraulic properties of fractured (through equivalent flow rate) rock nearest to the deposited canister have to be accurately determined in order to minimize uncertainties in the release to the far field. In case of increased sorption the maximum release of radionuclides decreased 10^1-10^4 times, in case of increased fracture transmisivity (subsequently Q_{eq}) the maximum release of the radionuclides increased app. 3-4 times. On the other hand, for nonsorbing long-lived iodine, with low diffusion coefficient uncertainties in the hydraulic properties have a very small effect on the release.

5. Summary

Preliminary analysis of radionuclide transport from possible repository for disposal of SNF in Lithuania was performed. The main features and processes important for radionuclide migration from the repository in case of SNF disposal in crystalline rocks were analyzed and available RBMK-1500 specific data were used. The possible impact of the specific features and processes related to KBS-3H real design were not taken into account.

The results of radionuclide transport analysis show that the most of identified safety relevant radionuclides for RBMK-1500 SNF disposal are effectively retarded in the near field region. The release to the far field is dominated by ^{129}I and ^{59}Ni firstly while after app. 50 thousand years ^{226}Ra is dominating. The sensitivity analysis of the results obtained was also performed.

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