

# Analysis of radionuclide release through EBS of conceptual repository for Lithuanian RBMK spent nuclear fuel disposal – case of canister with initial defect

Povilas Poskas,  
Asta Narkuniene,  
Dalia Grigaliuniene,  
Raimondas Kilda

**Abstract.** This paper presents research on radionuclide transport from generic geological repository for the RBMK-1500 SNF of 2.8  $^{235}\text{U}$  initial enrichment (with Er absorber) and average burn-up of  $\sim 29$  MWd/kgU. Radionuclide transport analysis was focused on the engineered barrier system (EBS) and performed taking into account possible differences in the data on the initial size of a canister defect, defect enlargement time and radionuclide release start time. For the numerical simulations, computer code AMBER (UK) was used. The analysis of radionuclide transport regularities demonstrates that the release from the EBS is the most intensive after the defect enlargement. Most relevant radionuclides were identified based on the mass transfer analysis complemented by the analysis of radiotoxicity flux. The results showed that, depending on the differences of the initial defect size, defect enlargement time and release start time, the peak flux from the EBS may vary by a factor of 2 (for  $^{129}\text{I}$ ) and 1.5 (for  $^{226}\text{Ra}$ ) for RBMK-1500 SNF.

**Key words:** geological repository • near field • radionuclide release • radiotoxicity • RBMK reactor • spent nuclear fuel

## Introduction

At the Ignalina nuclear power plant (NPP), app. 22 thousands of spent nuclear fuel (SNF) assemblies from the RBMK-1500 type reactor have been accumulated [15]. During the operation of Ignalina NPP, the assemblies of SNF were stored in water pools and in storage containers, located in the interim storage facilities. According to revised Lithuanian Strategy for Management of Radioactive Waste [20] approved by the Lithuanian Government, it is planned to store the assemblies with SNF in the interim storage facilities for about 50–100 years until the decision on its final disposal option is made.

There is a broad international consensus that the interim storage of spent nuclear fuel is not acceptable as a sole solution, but it is only a part of integrated and long term solution for management of this type of radioactive waste. As the time-frame for the decrease of the SNF radiotoxicity comprises hundreds of thousand years, its management solution has to be projected far in the future. Disposal in geological formations has been identified as the most proper option available for SNF and other high level waste today. As the technology development for transmutation/partitioning may not be available in the near future, it is necessary to proceed with the research regarding the geological disposal. As a member of the European Union, Lithuania must develop a comprehensive and integrated programme for the investigation and implementation of national

P. Poskas<sup>✉</sup>, A. Narkuniene, D. Grigaliuniene, R. Kilda  
Nuclear Engineering Laboratory,  
Lithuanian Energy Institute,  
3 Breslaujos Str., LT-44403 Kaunas, Lithuania,  
Tel.: +370 37 401 886, Fax: +370 37 351 271,  
E-mail: poskas@mail.lei.lt

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repository for SNF as defined by the European Directive [14].

Consistently with the worldwide tendency towards SNF disposal, research on SNF disposal in Lithuania was also initiated. Subsequently, with the assistance of Swedish experts from the Swedish Nuclear Fuel and Waste Management Company (SKB), the Lithuanian Energy Institute (LEI) proposed concepts of deep geological repository in clay and in crystalline rock formations [17]. These formations are available in Lithuania and potentially could be used for SNF disposal.

The role of natural barriers in safe disposal of SNF is played by stable geological formations of large extent where the disposal facility could be constructed. A high level of consensus exists on geosphere retention and transport processes, their definition and their generic importance [5]. However, there are contradictions that due to tectonics and long-term changes in rock structure, rock can serve only as a “mechanical support to the chemical apparatus” and that effective containment of hazardous elements can only be managed by properly designed and manufactured containers [19]. Regardless of the consideration of the role of natural barrier, the engineered barrier system (EBS) is very important for the functionality and safety of the underground facility. Besides, there are certain advantages in achieving a high confidence and reliability in isolation of SNF by the EBS – EBS components can be designed and constructed with well-described uniform properties, the evaluation of the performance of individual and assembled EBS components can be tested over a wide range of controlled conditions [1]. During the SNF disposal, the possibility of the emplacement of a canister with a small initial defect (pinhole) in the wall cannot be excluded. Even the probability of such event is low and is based on the efficiency of non-destructive control methods, the subsequent evolution of the disposal facility and radionuclides release has to be evaluated. For example, in Safety assessment SR-97 of repository for Swedish SNF [22] this situation was analyzed in the so-called “canister defect scenario”.

Up to now, the research done on radionuclide release and transport analysis from the geological repositories for RBMK type SNF is of limited extent. Preliminary studies on the radionuclide transport through the engineered and natural barriers of conceptual repository for RBMK-1500 SNF were performed and presented in [2, 16]. The analysis was performed for the canister whose initial defect increases and the continuous groundwater pathway forms after 200 thousand years after the repository's closure. The radionuclides  $^{59}\text{Ni}$ ,  $^{129}\text{I}$ ,  $^{135}\text{Cs}$ ,  $^{226}\text{Ra}$  were identified as dominating the radioactivity of the near field flux. Limited amount of scientific research on radionuclide transport from the geological repository for RBMK type SNF forces the continuation of research in this area. The present study is concentrated on the analysis of radionuclide transport peculiarities, the definition of dominant radionuclides in the flux from engineered barriers of conceptual RBMK-1500 SNF repository in the crystalline rocks at the Ignalina NPP region in the case of a canister with an initial small defect. Based on the differences in defect enlargement, initial size and radionuclide release start time, the modelling was performed for five cases.

## Methodology

There is a general IAEA methodology [10] which is being widely applied for the assessment of repository safety and transport of radionuclides. The key components of the methodology for the radiological impact assessment for the period after the repository's closure are: specification of the assessment context, description of the waste disposal system, development and justification of scenarios, formulation and implementation of models, performance of simulations and analysis of results, including sensitivity and uncertainty analyses, comparison with safety criteria, review and modification of the assessment, if necessary (i.e. iteration). Following the main steps of this methodology, the analysis of radionuclides transport and repository safety could be performed in a clear and systematic way.

## Repository concept

During operation of the Ignalina NPP, the nuclear fuel of a slightly different initial enrichment was used. Initially, the uranium dioxide with an initial enrichment of 2%  $^{235}\text{U}$  was used, while later the enrichment was increased up to 2.4%  $^{235}\text{U}$  with 0.41% of neutron absorber erbium. In 2001, 2.6%  $^{235}\text{U}$  with 0.5% Er uranium-erbium nuclear fuel was introduced and later, the fuel with 2.8%  $^{235}\text{U}$  and 0.6% burnable absorber erbium was used. Nuclear fuel with higher initial enrichment has the same geometry and mass as the initial type of fuel (2%  $^{235}\text{U}$  enrichment), however, due to higher initial enrichment its burn-up might be higher. Subsequently, the activity and residual decay heat would be higher. Therefore, the analysis of radionuclide transport from the repository was performed for SNF with an initial uranium enrichment of 2.8%. The radionuclide inventory to be used in this study was assessed by numerical code ORIGEN-S under the assumption of initial uranium enrichment of 2.8%, average burn-up of  $\sim 29$  MWd/kgU [6]. The initial concentration of impurities in the RBMK nuclear fuel and the structural parts of fuel assemblies were set as the maximum values.

The proposed generic repository concept for RBMK-1500 SNF disposal in the crystalline rocks was developed in [17] taking into consideration the international experience and was based on KBS-3 concept (Sweden). Taking into account the results of the criticality, dose rate assessment and thermal calculations as well as the existing experience in the canister shifting and emplacement technology, it was proposed to load 32 half-assemblies of RBMK-1500 SNF in a disposal canister. Based on preliminary assessment, the reference copper canister with cast iron insert would be of 1050 mm in diameter and 4070 mm in length. The amount of uranium within the fuel assembly is app. 111 kg, thus the total amount of uranium within SNF assemblies to be disposed of in the repository is app. 2436 tones. As it has already been mentioned, by the end of 2009 app. 22 thousands of SNF assemblies were accumulated in the Ignalina NPP. Thus, for Lithuania's SNF disposal purposes app. 1400 canisters should be used. The SNF canisters would be disposed of at 1.2 m distance from each other in horizontally bored

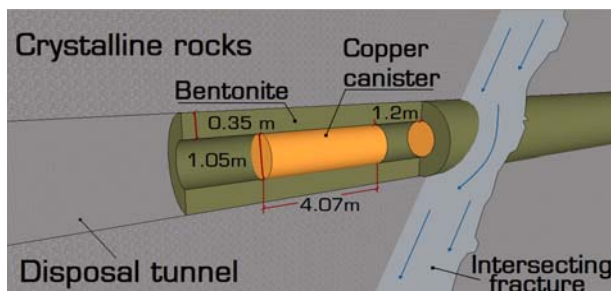


Fig. 1. Geometry of the repository near field.

emplacement tunnels (Fig. 1). The length of 250 m of SNF emplacement tunnels is accepted at this stage of investigations. The diameter of a SNF emplacement tunnel is proposed to be 1.85 m like in Sweden. The centre to centre distance between the emplacement tunnels is about 40 m. The length of shafts is dependent on the repository’s layout and the selected site. Compacted bentonite was proposed as a buffer material and a bentonite/crushed rock mixture would be employed as a backfilling material. The clay barrier offers a very good isolation of the canisters because of its favourable properties (very low hydraulic conductivity, high swelling potential, favourable rheological properties, colloid filtering capacity, etc.) [18]. In case of the site at the Ignalina NPP region, several clay formations (at different depth) and crystalline basement rocks (granite) have a potential to be a host rock of the geological repository. The Ignalina NPP is located in the north eastern part of Lithuania, close to the state border with the Republic of Belarus and Latvia. In this region, the crystalline basement rocks are overlaid by the system of sedimentary rocks (Quaternary and Prequaternary rocks). The crystalline basement occurs at a depth of app. 700–800 m.

Radionuclide transport scenario

Analysis of the radionuclide transport through the engineered and natural barriers of the repository into the biosphere is a complex task which incorporates a large number of disciplines (material science, chemistry, mechanics, hydraulics, geology) on different aspects related to a particular part of the repository. The extent of research and investigation in this field in each country depends on radioactive waste management strategy, available resources and facilities. The main processes related to the radionuclide transport through the engineered and natural barriers are their release mechanisms from the waste matrix, transport mechanisms,

interaction with barrier materials, transport and accumulation in the ecosystems, food chains, radioactive decay/ingrowth. In certain cases, radioactive ingrowth could be disregarded if the parent radionuclides are not distributed within the natural barrier or ecosystem, for example, due to their significant retardation in the engineered barriers.

The radionuclide release from the SNF disposal canister emplaced in a deep geological environment can occur if the waste package loses its tightness in the course of time. The causes for this event could be naturally occurring processes such as container corrosion, earthquake, glaciations or human related processes (such as inadvertent human intrusion). In the context of safety evaluation of this type of nuclear facilities, the course of various processes and events is being developed for the evaluation of possible impact on the environment under assumed conditions (scenario development). In this study, the radionuclide transport analysis was performed to model the situation of possible emplacement of a canister with an initial defect.

In case of a canister with an initial small defect in the wall, such defect is expected to be related to the welding of the canister’s lid to the cylinder or similar places. For the transport of the radionuclide from the canister, the outer shell and the inner insert has to be untight (have some discontinuities) in order to provide a way for groundwater to come into contact with SNF. The course of events involved in the formation of the continuous water pathway is complex and depends on the coupled physical processes (mechanical, chemical, hydraulic, etc.). Due to large uncertainties, the evolution of the initial defect and the beginning of radionuclide transport out of the disposal canister is treated in a simplified way. Based on the mechanical, chemical, and hydraulic analysis of the disposal canister, the potential time-frame for the defect growth (enlargement) and continuous water pathway development is usually proposed. Different time of defect enlargement and radionuclide release start as well as different initial defect size was analyzed performing the safety assessments of Swedish, Finish SNF repositories [23–25]. In this study, the investigation of radionuclide transport in the near field (engineered barriers) of the SNF repository was performed considering different assumptions regarding the canister’s defect evolution as summarized in Table 1.

The laboratory investigations and modelling of SNF behaviour after its disposal had been carried out over 25 years and lead to a common understanding that some radionuclides are released in two parts – a fast release of radionuclides which are not contained in the

Table 1. Summary of different assumptions regarding the canister’s defect evolution and radionuclides release time based on Refs. [23–25]

Cases	Size of canister defect before enlargement $A_{\text{defect}} \text{ (m}^2\text{)}$	Time of canister defect enlargement $T_{\text{large}} \text{ (y)}$	Radionuclide release start time $T_{\text{start}} \text{ (y)}$
Case A-1 (basic case for comparison)	$1 \times 10^{-6}$	200 000	200 000
Case A-2	$1.3 \times 10^{-5}$	10 000	1 000
Case A-3	$1 \times 10^{-6}$	20 000	300
Case A-4	$5 \times 10^{-6}$	–	0
Case A-5	–	10 000	10 000

matrix and a progressive and relatively slow release of radionuclides [8, 9, 11]. The instant release fraction (IRF) depends on the radionuclide distribution within the fuel pellet (matrix), linear power and burn-up [11]. There are some experimental data on IRF for PWR, BWR type of SNF, but the IRF values for iodine, chlorine, carbon, selenium are still largely unknown and are represented by bounding values that remain to be confirmed [9]. However, the data for RBMK type fuel has not been reported yet. In this study, it was assumed that data on instant release fraction for the radionuclides was the same as reported in [26]. The values of the highest probability were accepted (from probability distribution functions being suggested).

The congruent release from SNF matrix depends on its dissolution under particular chemical conditions and is related to relatively slow uranium dioxide dissolution (low uranium solubility under reducing conditions). Based on [4, 26] key parameters influencing SNF matrix alteration/dissolution are: specific activity of SNF, primary radiolytic yields, temperature, specific surface of SNF, iron and H<sub>2</sub> concentration, concentration of carbonates, initial oxidation state of SNF matrix, etc. Taking into consideration the inhibiting effect of hydrogen generation within the canister, SNF matrix dissolution rate ranges from 10<sup>-6</sup> to 10<sup>-8</sup> 1/y (based on the experimental results reported) as stated in [26]. However, as it is mentioned in [9] the beneficial effect on H<sub>2</sub> in reducing the potentially oxidizing effect alpha radiolysis (related to radiolytic SNF matrix dissolution) is known only for relatively simple solutions that are not representative for the groundwater in the repository's environment. As there is no information on RBMK type SNF dissolution (neither experimental nor modelling data) published, the fractional dissolution rate of 10<sup>-7</sup> 1/y was selected for the current study. This is the most expected value from the suggested probability distribution function in [26].

As radionuclides are released from the SNF matrix (gradually or instantly), the interaction with the groundwater takes place within the canister. Depending on the prevailing chemical conditions (pH, Eh, groundwater composition) and the concentration of the radionuclides, a certain amount of radioactive particles could start to precipitate and remain within the environment of the SNF. Solubility is usually used as an upper limit of the radionuclide concentration in given chemical conditions and depends on the groundwater composition, which is determined by a number of processes in other EBS materials (dissolution of impurities in the bentonite buffer, conversion of montmorillonite to non swelling minerals, etc.). Site specific data on the radionuclides solubility are not available for Lithuania's crystalline rocks, therefore this data was based on the literature survey. Data reported for the crystalline rocks in Sweden (Beberg Site) [23] were assumed to be used in this analysis. During the selection of the data base for solubility limit values, the source with most comprehensive list of radionuclides was preferred. Conservatively no shared solubility was assumed.

Radionuclide transport through the engineered barriers occurs by advective and diffusive transport mechanisms which are described by mass transport laws. The dominating mechanism of the radionuclide transport in

the geological environment depends on the nature of the geologic formation. The compacted bentonite buffer serves as a low permeability barrier, thus radionuclides migration through it is controlled by diffusion, driven by concentration gradients whose upper value is limited by the radionuclide solubility. In addition, they are retarded by sorption onto the mineral constituents of the buffer material in the course of migration. According to the proposed repository concept, the disposal canisters would be surrounded by a MX-80 bentonite layer of very low hydraulic conductivity (< 10<sup>-13</sup> m/s), thus the dominant transport mechanism is expected to be diffusive transport. While being transported towards the surrounding geologic media, the radionuclides interact with the material of EBS in the form of chemical reactions (ion exchange, surface complexation, absorption), physical adsorption and could be retained in EBS. The mentioned interaction mechanisms depend on the pore water composition and are site-specific and commonly expressed as "sorption" term described by the distribution (sorption) coefficient. For this study, data on diffusion and sorption in the MX-80 type bentonite was taken from [13].

In the hard rock (such as crystalline rocks), the advective transport through the intersecting fractures forms the main pathways of radionuclide transport towards the biosphere. After being transported to the boundary of the surrounding bentonite layer, the radionuclides are expected to be transferred to the surrounding host rock. Mechanisms of radionuclide transfer from the EBS to the rock will depend on the hydraulic conditions around the emplacement tunnel, the excavations disturbed zone (EDZ) extent and prevailing groundwater flow conditions in it. In this study, it is assumed that the emplacement tunnel will be excavated with a tunnel boring machine which causes the EDZ of a very limited extent. The bentonite is expected to expand and close the minor fractures due to its swelling properties. Nevertheless, the radionuclides could be transported to a larger fracture, which could intersect the emplacement tunnel and provide a preferential pathway of groundwater movements around the emplacement area. Data for the evaluation of equivalent flow rate carrying the radionuclides into intersecting fracture based on [12] was assumed as follows: fracture aperture 250 μm, fracture transmissivity 1 × 10<sup>-8</sup> m/s, head gradient of 1%.

### Conceptual model

In this study, the radionuclide transport analysis was performed for a horizontally emplaced disposal canister. In order to evaluate the effect of defect related parameters on radionuclide flux, five cases were distinguished with data on the canister's defect enlargement as summarized in Table 1.

For the assessment of the most dangerous radionuclides leaving the EBS, the radionuclide release was not analyzed in terms of mass transport only. The radiological characteristics of radionuclides (activity and radiotoxicity) transferred to the natural barrier were also assessed.

The main processes determining the radionuclide release from the near field of the repository in case of

SNF disposal in the crystalline rocks were modelled and included SNF matrix dissolution, instant release from SNF matrix, dissolution in the groundwater, diffusion, sorption, decay/ingrowth and release to the groundwater flowing around the disposed canister. The main assumptions made during the computational study were as follows:

- The space inside the disposal canister (app. 0.5 m<sup>3</sup>) will be filled by the groundwater after the defect enlargement.
- Structural parts of the SNF assemblies (spacer grids, fuel rods, etc.) do not limit the release of the radionuclides embedded in them.
- Certain radionuclides are released instantly and congruently with matrix dissolution (<sup>14</sup>C, <sup>79</sup>Se, <sup>129</sup>I, <sup>135</sup>Cs and some others).
- Sorption on the inner parts of the canister, structural parts of fuel bundle and concentration gradient is disregarded due to large uncertainties.
- Bentonite buffer is fully saturated at the beginning of radionuclide release.
- Dissolved radionuclides are transported out of the canister by diffusion.
- Radionuclides distributed within the bentonite buffer diffuse into the groundwater flowing in the rock fracture intersecting the emplacement tunnel.

In this study, the radionuclide release through the EBS was performed for the safety relevant radionuclides for RBMK-1500 SNF [3]. Safety relevant radionuclides were selected on the basis of comparative study. The analysis was carried out using different methodologies, proposed by SKB (Sweden), NAGRA (Switzerland) and JNC (Japan). Nineteen fission and activation products and 27 actinides and their daughter nuclides were identified as relevant (potentially important) radionuclides for the safety of RBMK-1500 SNF geological repository (Table 2) and used in the present study.

### Mathematical model

As it has already been mentioned, different phenomena, determining the radionuclide release from the near field of the repository (SNF dissolution, instant release, solubility limitation, diffusion, sorption, decay/ingrowth), were taken into account in the simulations. Due to a very low permeability of the bentonite buffer, the transport of radionuclides released from SNF matrix and dissolved in water through the near field is diffusion dominated. The disposal system was discretized into compartments and the integrated finite difference method was used. In this study, the analytical solutions were embedded to speed up the calculations based on [21] instead of fine discretization at sensitive zones, for example, at the exit point of the canister hole (defect) and at the entrance to fractures. The material balance

over the compartment, connected to some other compartments, for  $n$  radionuclide is expressed as follows

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

where  $V$  – compartment volume (m<sup>3</sup>),  $C$  – radionuclide concentration (mol/m<sup>3</sup>),  $\varepsilon$  – material porosity (m<sup>3</sup>/m<sup>3</sup>),  $R$  – retardation factor (-),  $S$  – general source term (mol/y),  $\lambda$  – decay constant (1/y),  $A$  – compartment cross-section area (m<sup>2</sup>),  $D_e$  – diffusivity in material (m<sup>2</sup>/y),  $d$  – diffusion length (m).

Retardation factor ( $R$ ) is defined as follows

$$(2) \quad R^n = 1 + \frac{1-\varepsilon}{\varepsilon} K_d \rho_s$$

where  $\varepsilon$  – porosity (m<sup>3</sup>/m<sup>3</sup>),  $K_d$  – sorption coefficient of  $n$  radionuclide (m<sup>3</sup>/kg),  $\rho_s$  – material density (kg/m<sup>3</sup>).

The ratio  $d/AD_e$  in Eq. (1) could be expressed as the transport resistance with analogy with electrical resistance. For the representation of additional transport resistance, concentrated near the canister's defect where the diffusing species spread out spherically, it is expressed as a plug resistance [21]

$$(3) \quad R_p = \frac{d}{AD_e} = \frac{1}{\pi r_{\text{hole}} D_e \sqrt{2}}$$

where  $d$  – diffusion length of the plug (m),  $A$  – diffusion area set equal to the area of the hole ( $A_{\text{defect}}$ ) (m<sup>2</sup>),  $r_{\text{hole}}$  – radius of the hole (m).

Mass transfer of dissolved species from the stagnant pore water 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  $Q_{eq}$  [12]

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

where  $2b_v$  – aperture of the fracture (m),  $r_2$  – radius of the deposition tunnel (m),  $D_w$  – diffusivity in water (m<sup>2</sup>/y),  $u$  – velocity of water in the fracture (m/y).

Radiotoxicity (RT) of a given amount of radioactive material is expressed in terms of hypothetical dose resulting from ingestion of the activity  $A_i$  at a time  $t$

$$(5) \quad RT(t) = \sum A_i(t) \cdot DC_{i,ing}$$

where  $DC_{i,ing}$  – dose coefficient for the ingestion of  $i$  radionuclide.

In this study, the modelling was performed using computer program AMBER (UK) [7] which is a flexible software tool for building of the dynamic compartmental models to represent the migration and fate of the

**Table 2.** Safety relevant radionuclides for RBMK-1500 SNF disposal [3]

Actinides and their daughter nuclides	Fission and activation products
<sup>244</sup> Cm, <sup>240</sup> Pu, <sup>236</sup> U, <sup>232</sup> Th, <sup>228</sup> Ra, <sup>228</sup> Th (4N)	<sup>3</sup> H, <sup>10</sup> Be, <sup>14</sup> C, <sup>59</sup> Ni, <sup>63</sup> Ni, <sup>79</sup> Se, <sup>90</sup> Sr, <sup>93</sup> Zr, <sup>93m</sup> Nb,
<sup>245</sup> Cm, <sup>241</sup> Pu, <sup>241</sup> Am, <sup>237</sup> Np, <sup>233</sup> U, <sup>229</sup> Th (4N+1)	<sup>94</sup> Nb, <sup>99</sup> Tc, <sup>107</sup> Pd, <sup>126</sup> Sn, <sup>129</sup> I, <sup>135</sup> Cs, <sup>137</sup> Cs, <sup>147</sup> Sm,
<sup>246</sup> Cm/ <sup>242m</sup> Am, <sup>242</sup> Pu, <sup>238</sup> U/ <sup>238</sup> Pu, <sup>234</sup> U, <sup>230</sup> Th, <sup>226</sup> Ra, <sup>210</sup> Pb, <sup>210</sup> Po (4N+2)	<sup>151</sup> Sm, <sup>166m</sup> Ho
<sup>243</sup> Am, <sup>239</sup> Pu, <sup>235</sup> U, <sup>231</sup> Pa, <sup>227</sup> Ac (4N+3)	

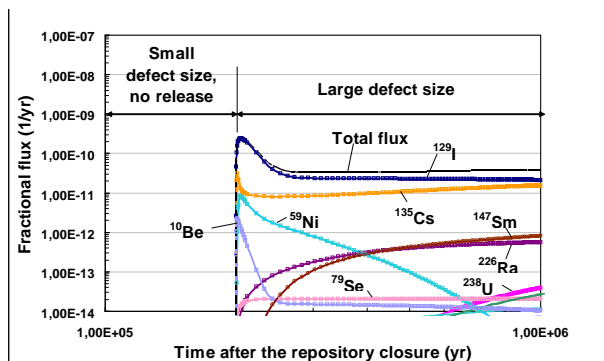
contaminants in the system. System discretization is similar to that of reported in [23].

## Results and discussion

As mentioned earlier, preliminary studies of the radionuclide transfer through the engineered and natural barriers of conceptual repository for RBMK-1500 SNF were performed and presented in [2, 16]. These results were based on the radionuclide release analysis with the same defect related assumptions as in the basic case of this study, i.e. the defect becomes large and the continuous groundwater pathway to the SNF matrix is established at 200 thousand years after repository closure. The radionuclides  $^{59}\text{Ni}$ ,  $^{129}\text{I}$ ,  $^{226}\text{Ra}$  were identified as the dominating ones in the near field flux for this case. The first two radionuclides dominate the total activity soon after the defect enlargement and later  $^{226}\text{Ra}$  is dominating. However, the dominant radionuclides were identified based on the analysis of the flux activity. As outlined in the introduction, the current study was concentrated on the analysis of radionuclide mass transport peculiarities, the definition of dominant radionuclides in the flux from engineered barriers of conceptual RBMK-1500 SNF repository in the crystalline rocks at the Ignalina NPP region in case of a canister with an initial small defect. The analysis was based on the comparative study of five cases covering the transport analysis through a small and enlarged defect with different defect related parameters. The most important radionuclides were identified based on mass transport analysis complemented with the analysis of the radiological characteristics. Part of the analysis was dedicated to analyze the impact of the canister defect related parameters on radionuclide peak flux.

### Mass transport through the EBS

Modelling of the radionuclide release through the EBS was performed for different cases under different assumptions, regarding the canister's defect size evolution and radionuclide release start time. The analysis of radionuclides flux from the canister and transport through the surrounding bentonite barrier showed that the majority of safety relevant radionuclides would be effectively retarded in the near field region of the

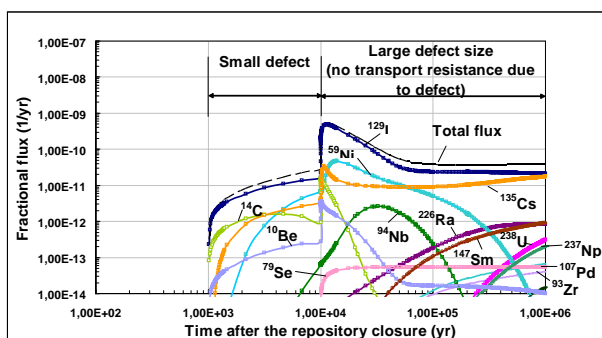


**Fig. 2.** Fractional flux of radionuclide diffused through the engineered barriers to the geosphere in the case of canister with initial defect (case A-1 -  $T_{\text{large}} = 2 \times 10^5$  y,  $T_{\text{start}} = 2 \times 10^5$  y).

repository. Figure 2 illustrates the modelling results in terms of fractional flux of radionuclides diffused out of the near field region (EBS) for case A-1 (the radionuclide flux (mass release rate) per 1 mole in the canister). It is likely that for radionuclides  $^{129}\text{I}$ ,  $^{135}\text{Cs}$  the flux profile soon after the defect enlargement and at the beginning of radionuclide release is determined by the inventory released instantly from SNF matrix (IRF) and the rest part corresponds to the slower release by SNF matrix dissolution. In case of radionuclides with 100% availability (conservative assumption on instant release) (e.g.  $^{59}\text{Ni}$  from structural parts of SNF assemblies), peak flux (maximum mass release rate) and its time are determined by the concentration gradient and interaction with material. Decreasing profile of flux corresponds to decreasing concentration gradient and the inventory decreasing due to radioactive decay. It should be pointed out that the potential flux of  $^{10}\text{Be}$  might be possibly overestimated. This was caused by conservative initial assumptions for this radionuclide (no sorption in MX-80 bentonite and no solubility limit).

Figure 3 illustrates the results of radionuclide mass transport for case A-2. The difference between this case and the basic case (case A-1) is that in this case the radionuclide transport starts earlier than the enlargement of the defect occurs and a part of the inventory is being transported while the defect still remains small. As can be seen in Fig. 3, the flux profiles before and after the defect enlargement are noticeably different. The results demonstrate that at the beginning, the release from the canister is limited, while the canister's defect is small. The flux of most radionuclides increases gradually with time of the defect enlargement (with the exception of  $^{14}\text{C}$ ). When the defect becomes large, the flux increases sharply and the peak flux of dominant radionuclides is then observed.

The comparative analysis of the results of all cases showed that the dominant radionuclides in the flux from the bentonite barrier were the non-sorbing long-lived radionuclide  $^{129}\text{I}$  and long-lived radionuclides  $^{135}\text{Cs}$ ,  $^{59}\text{Ni}$ . The total flux is determined by iodine in all cases being analyzed and its impact is decreasing with time, while the contribution of  $^{135}\text{Cs}$  is increasing in the long term. As the half-lives of  $^{14}\text{C}$ ,  $^{94}\text{Nb}$  are shorter than that of  $^{129}\text{I}$  or  $^{135}\text{Cs}$ , their contribution to the total flux is more dependent on the defect enlargement time. As the defect enlargement occurs after the longest period of time after the repository's closure (case A-1,  $T_{\text{large}} = 200$  thousand years,  $T_{\text{star}} = 200$  thousand years), their impact



**Fig. 3.** Fractional flux of radionuclides diffused from the engineered barriers to the geosphere in the case of canister with initial defect (case A-2 -  $T_{\text{large}} = 10^4$  y,  $T_{\text{start}} = 10^3$  y).



is significantly decreased due to radioactive decay. No short-lived radionuclides would be released in this case (case A-1), since they would have decayed by the time the groundwater contacted the SNF matrix.

Assessed flux profiles do represent the rate of radionuclide transport from the near field and its change with time. In order to evaluate the total mass of the radioactive material transported away to the fracture, mass flux was integrated over the analyzed time period ( $\int_{t=0}^{t_{end}} Q_i(t) = CDF$ ). Estimated total amount of radioactive materials to the natural environment was compared to radionuclide's initial amount in the disposal canister ( $CDF/M_{i0}$  (mol/mol)). The results of such analysis for case A-1 revealed that the total amount of  $^{129}\text{I}$  released to the natural barrier corresponds to app. 9% of its initial amount, for  $^{10}\text{Be}$  – app. 14.7%,  $^{59}\text{Ni}$  – app. 0.63%,  $^{135}\text{Cs}$  – app. 1.8% (Fig. 4). This indicates the considerably small amount of mass being transported away from the repository during such a long time period. For caesium isotope this amount is close to that part of the initial inventory that is released instantly (2% of IRF), for  $^{10}\text{Be}$  and  $^{129}\text{I}$  it is slightly higher than IRF (10 and 2%, respectively). In case of nickel isotope, which is produced in the structural parts of SNF assemblies under the neutron flux in the reactor core ( $^{59}\text{Ni}$ ), this is significantly lower than the amount instantly released (100%) from the structural parts. This confirms that even for the long-lived non sorbing or weakly sorbing radionuclides the engineered barriers act as an important system component leading to the decreased flux from the repository when the disposal canister does not act as a containment or a release controlling barrier, i.e. when there is a continuous groundwater pathway and a large defect.

However, for certain actinides (such as  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ,  $^{229}\text{Th}$ ) the total amount of radioactive material released to the natural barrier during the analyzed period could be significantly higher (up to 3–5 orders of magnitude) than its initial amount in the disposal canister. This is a result of the radioactive ingrowth from the parent radionuclides during a long time after the repository's closure and a small initial amount in the disposal canister. However, the comparison to the radionuclides dominating in the mass flux from the EBS showed that the flux of  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ,  $^{229}\text{Th}$  and other actinides is lower at least by one order of magnitude. Mass flux of Ra isotopes is more significant only due to its relatively high mobility in the EBS in comparison to the other actinides.

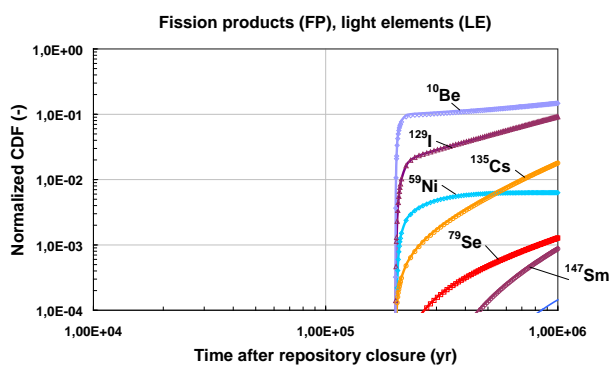


Fig. 4. Normalized cumulative functions of the radionuclides dominating the mass flux from the EBS (case A-1 –  $T_{large} = 2 \times 10^5$  y,  $T_{start} = 2 \times 10^5$  y).

### Impact of defect related parameters on radionuclide peak flux

As the peak flux (maximum mass release rate) is usually analyzed for the assessment of repositories safety, a part of the analysis was focused on the evaluation of the effect of defect related assumptions on the peak flux. Defect related parameters include initial defect size, defect enlargement time and radionuclide release start time. Comparison of the modelling results of the analyzed cases was based on the ratio of peak flux for particular case and case A-1 (chosen as a basic case). The results showed that the peak flux of the radionuclides depends on these assumptions differently (Fig. 5). In case of  $^{129}\text{I}$  and  $^{226}\text{Ra}$ , the assumption about the defect enlargement vs. no enlargement (case A-4 – no enlargement,  $T_{start} = 0$  y) has the highest importance. The peak flux of  $^{129}\text{I}$  and  $^{226}\text{Ra}$  for case A-4 (without defect enlargement) is lower than in the basic case app. 11 and 33 times, respectively. In case of radionuclides  $^{94}\text{Nb}$  and  $^{14}\text{C}$  (not presented in figure), the assumption on the defect enlargement time plays the greatest role. For  $^{14}\text{C}$ , the observed peak flux ( $Q_{14\text{C},\text{bentonite}}^{\text{max}}$ ) is decreasing as follows: case A-2 ( $T_{large} = 10^4$  y,  $T_{start} = 10^3$  y)  $\rightarrow$  case A-5 (no small defect,  $T_{large} = 10^4$  y,  $T_{start} = 10^4$  y)  $\rightarrow$  case A-3 ( $T_{large} = 2 \times 10^4$  y,  $T_{start} = 300$  y)  $\rightarrow$  case A-4 (no enlargement,  $T_{start} = 0$  y)  $\rightarrow$  case A-1 ( $T_{large} = 2 \times 10^5$  y,  $T_{start} = 2 \times 10^5$  y) (not significant). The minimal changes of the peak flux in case of different assumptions were obtained for radionuclide  $^{135}\text{Cs}$ . Due to the differences in the initial defect size, enlargement time and release start time, the peak fractional flux may vary by a factor of 2 for  $^{129}\text{I}$  and 1.5 for  $^{226}\text{Ra}$  (Fig. 5). The results suggest that the impact of the variations in defect related parameters might cause a variation of peak flux of limited extent for dominating radionuclides. The significance of peak flux variations should be evaluated considering the uncertainty in other transport related parameters.

The results of cases under consideration indicated that the maximal total flux from the EBS ranges between  $(3.7 \times 10^{-11} - 5.5 \times 10^{-10}) \times M_{\text{total}}$  (mol/y) for RBMK-1500 SNF. The variation is within one order of magnitude despite the significant differences in the defect enlargement time, release start time and the initial defect size. This suggests that the engineered barrier system works effectively as such a small part of the initial amount of radioactive material in the canister is

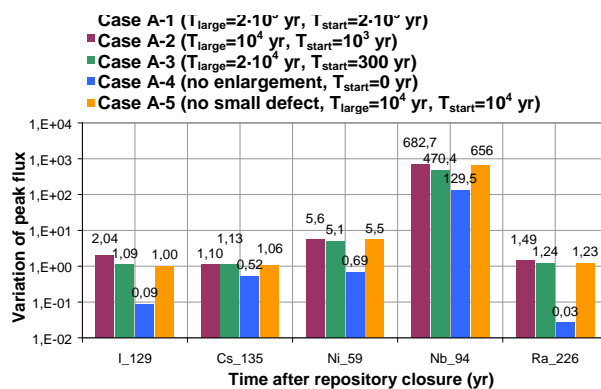
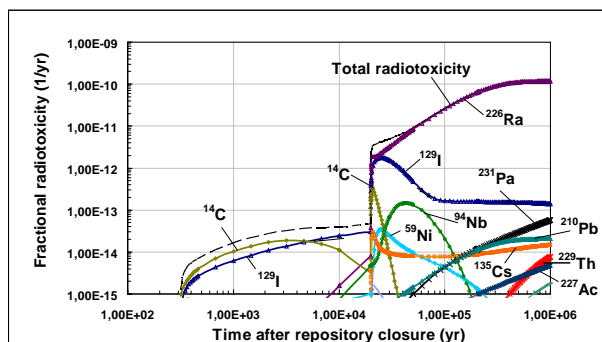


Fig. 5. Comparison of peak fluxes from EBS observed with different assumptions on initial canister defect size, enlargement time and radionuclide release start time (compared to basic case).

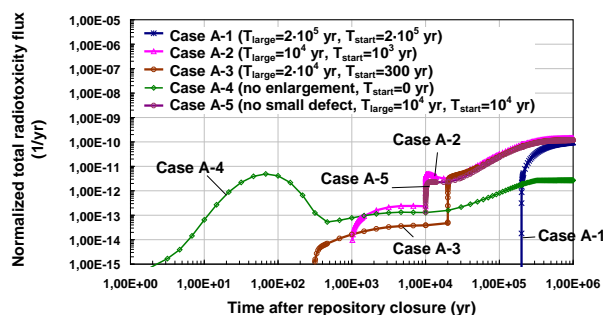
being transported away to the environment even if the canister does not function as release limiting barrier (canister with a small defect).

### Radiotoxicity flux

The largest amount of the particle being transported away from the SNF emplaced in the canister does not necessarily correspond to the highest radiological hazard to the human or environment, thus in case of radioactive particles, the radiological characteristics have to be determined and analyzed. Taking into consideration the radionuclide activity and radiotoxicity, the most dangerous radionuclides to human health were identified. Comparison of the results of cases being analyzed was performed. It was observed that based on radiotoxicity, the radionuclides  $^{14}\text{C}$  and  $^{129}\text{I}$  were the most dangerous radionuclides in the flux leaving the bentonite barrier if the canister defect is small. The situation is different after the defect enlargement. With the large defect the dominating radionuclides are  $^{129}\text{I}$  and  $^{226}\text{Ra}$  dominate the total radiotoxicity. This difference is well illustrated in Fig. 6 with results for case A-3. Results are presented as fractional radiotoxicity (radionuclide's radiotoxicity flux per total initial radiotoxicity of all radionuclides in the canister  $\{(RT_{i,\text{bentonite}}) / RT_{\text{total}}\}$ ,  $\text{(Sv/y)/Sv}$ ). Apart from these radionuclides, according to relative radiotoxicity ( $RT_{i,\text{pr}}(t) > 0.001 \sum RT_{i,\text{pr}}(t)$ ) [3] the radionuclides  $^{59}\text{Ni}$ ,  $^{94}\text{Nb}$  and  $^{135}\text{Cs}$  are relevant as well. Based on the modelling results,  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  become important ones during 0–300 years after the repository closure period too, if the release from the disposal canister starts within this period.



**Fig. 6.** The contribution of radionuclides to the total radiotoxicity flux, transported to the natural barriers (case A-3 –  $T_{\text{large}} = 2 \times 10^4$  y,  $T_{\text{start}} = 300$  y).



**Fig. 7.** Radiotoxicity flux observed with different assumptions on the canister defect size, its enlargement time and radionuclide release start time.

Profiles of total radiotoxicity flux from EBS under different assumptions on initial canister defect size, enlargement time and radionuclide release start time are presented in Fig. 7. According to the results presented in Fig. 7, the radiological impact of radionuclides flux leaving the near field is different for different cases. Radiotoxicity flux is non-monotonic and time dependent. It can be seen from Fig. 7 that the maximum of total radiotoxicity flux in case A-4 (no defect enlargement) is observed earlier in comparison to the cases where the defect enlargement is considered. For other cases with defect enlargement, the peak of total radiotoxicity flux is observed by the end of one million years. Thorough analysis of the radionuclide's specific radiotoxicity revealed that the radiotoxicity of  $^{226}\text{Ra}$  plays an important role when the canister's defect enlargement is considered. Based on the modelling results it could be concluded that the maximal total radiotoxicity of the all radionuclides leaving the EBS varies in app. two order of magnitude and ranges in  $(4.9 \times 10^{-12} - 1.4 \times 10^{-10}) \times RT_{\text{total}}$  (Sv/y). As in case of mass release rate, a fraction of initial radiotoxicity contained in the canister, which could be transported to the environment, is considerably small due to the efficiency of the EBS containing the most of actinides in sorbed form and of limited concentration.

### Conclusions

The complex and systematic analysis of the potential radionuclide transport from the conceptual repository for RBMK-1500 SNF disposal in crystalline rocks in case of the canister defect scenario was carried out. The analysis was performed for five cases with different defect enlargement time, radionuclide release start time and initial defect size. Based on the modelling results, the regularities of radionuclide transport were estimated, the impact of the defect related parameters on the peak radionuclides flux was assessed, and the following conclusions were made:

1. Radionuclides  $^{129}\text{I}$ ,  $^{14}\text{C}$ ,  $^{135}\text{Cs}$  and  $^{59}\text{Ni}$  dominate in the mass flux from EBS before the defect enlargement and  $^{129}\text{I}$ ,  $^{135}\text{Cs}$  and  $^{59}\text{Ni}$  dominate after the defect enlargement.
2. The maximum of total flux (mass release rate) from the EBS ranges between  $(3.7 \times 10^{-11} - 5.5 \times 10^{-10}) \times M_{\text{total}}$  (mol/y) and corresponds to only small fraction of initial inventory in the disposal canister.
3. Peak flux value for  $^{94}\text{Nb}$  and  $^{14}\text{C}$  depends on defect enlargement time, but for  $^{129}\text{I}$  and  $^{226}\text{Ra}$  the defect enlargement is the most important factor.
4. Within the analyzed ranges of the enlargement time, initial defect size and radionuclide release start time, the maximum of radionuclide flux from the engineered barriers could vary by a factor of 2 for  $^{129}\text{I}$  and 1.5 for  $^{226}\text{Ra}$ .
5. Taking into account toxicity of radionuclides  $^{14}\text{C}$  and  $^{129}\text{I}$  are the most dangerous radionuclides diffused through a small defect and engineered barriers. In case of defect enlargement, radionuclides  $^{129}\text{I}$  and  $^{226}\text{Ra}$  (in the long-term) are of the greatest importance; however,  $^{59}\text{Ni}$ ,  $^{94}\text{Nb}$ ,  $^{135}\text{Cs}$  cannot be neglected. In case of the early beginning of the release



(0–300 years after the repository closure), the short-lived radionuclides  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  are also important.

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