ACCOUNTING FOR ENGINEERED BARRIER SATURATION, STEEL CONTAINER CORROSION, HYDROGEN PRODUCTION AND TRANSFER WITHIN FRACTURED ROCK IN THE ASSESSMENTS OF LOW SORBING RADIONUCLIDES RELEASE FROM GEOLOGICAL DISPOSAL FACILITIES

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The paper discusses the influence of bentonite saturation, package failure and water conductive fracture network on the transport of long-lived low sorbing radionuclides in geological disposal facilities (DGR). Based on the example of ^{129}I, the study evaluates the influence of space and time dispersion of radionuclide transport and processes mentioned above on the long-term DGR safety. It demonstrates that these effects can cause significant radionuclide retention, nevertheless having minor impact on the long-term DGR safety as regards low sorbing nuclides with their half-lives amounting to over hundreds of thousand years. The paper also considers the opportunities for enhancing the GDF safety considering such radionuclides.

Keywords: *radioactive waste, deep geological disposal, bentonite saturation, low sorbing long-lived radionuclides, water conductive fracture network.*

Introduction

Safe disposal of radioactive waste (RW) is seen as a crucial challenge faced by the nuclear industry especially when it comes to waste containing long-lived radionuclides and requiring deep geological disposal (RW Class 1 and 2 according to the Russian RW classification system [1], RW Class 1 hereinafter referred to as RW). In addition to some obvious problems, i. e., high radiological hazards and residual heat release from such waste requiring some complex and expensive engineering solutions to be developed, certain methodological questions with no universal answers yet provided still remain to be addressed.

A case in point is the numerical assessment of a time period during which a deep disposal facility is required to provide the containment of radionuclides contained in the RW emplaced into it. Russian standard [2] requires the DGR to "…provide reliable isolation[1] *ensuring the radiation safety of human and the environment for the entire period while the radioactive waste potentially remains hazardous.*" However, it has now been established that potential DGR hazard is mainly associated with possible

[1] In the IAEA safety glossary [6], "isolation" is referred to as the physical separation of radioactive waste away from the environment in order to prevent people from having physical contact with the waste, and to reduce negative environmental impacts on the engineered barriers and the host rocks. Prevention or control of releases of radioactive material to the environment is referred to as confinement.

environmental releases of long-lived practically non-absorbable forms of radionuclides, including ^{36}Cl, ^{79}Se, ^{99}Tc, ^{129}I with half-lives of $3.01 \cdot 10^5$, $3.27 \cdot 10^5$, $2.13 \cdot 10^5$ and $1.57 \cdot 10^7$ years, respectively (see, for example, [3], [4] providing the safety assessments of DGRs planned to be sited abroad in crystalline rocks are similar to the Russian disposal facility proposed to be constructed at the Yeniseiskiy site [5]).

It's hardly possible to demonstrate such "reliable isolation" if the considered radionuclides tend to remain radiologically hazardous for over hundreds of thousands and millions of years specifically when speaking of their zero release into the environment. Two approaches were proposed to address this problem.

Russian safety guideline [7] recommends to limit the calculation period so that the forecasts covering such a timeframe would show that the radiation exposure of the population from a DGR reaches its maximum level and cannot grow any further. This approach implies that radionuclide transport beyond the safety barriers can be predicted for a time period for which such assumptions can be considered reliable.

If maximum radionuclide release is not expected to occur within a timeframe during which the calculations can be considered reliable, other approaches are applied. A case in point is the safety case developed based on the requirements of [9] for the Finnish DGR in Olkiluoto [8] due to the copper container designs proposed under the Scandinavian concepts: theoretically, these containers can provide confinement for at least hundreds of thousands of years with quantitative safety[1] assessments covering a period of only 10,000 years. It is believed that this is the longest period for which somewhat reliable predictions can be made regarding global climate changes, geological processes, human behavioral patterns and human diets. In the safety assessments, longer time periods are considered based on certain qualitative and semi-qualitative approaches relying upon some general considerations, natural analogues, etc. For a DGR, its protective properties actually last as long as the packages with the RW remain intact. Although this period is expected to last for quite a long time, it may nevertheless be insufficient to provide the radiation safety for the abovementioned radionuclides.

RW disposal in steel containers is considered as a possible design option proposed under the Russian DGR designs (see, for example, [10], [11]). Container corrosion rate and the time period over which they can provide confinement under disposal conditions are much inferior to those of the copper containers. Moreover, this safety approach is not expected to provide zero release of radionuclides into the environment, but rather to substantially slow down this process over an extensive time period, which, taking into account the radioactive decay and some natural environmental processes (dilution in various media and radionuclide transitions into less mobile forms), may turn to be sufficient to reduce the DGR environmental impact to an acceptable level.

If DGR safety is demonstrated, this approach can be considered more sound and reasonable than the assumptions made on the copper containers and their leak-tightness over indefinitely long time periods.

This paper examines the influence of two mechanisms slowing down radionuclide transport: the time-distributed DGR saturation with groundwater (GW) at the post-closure stage and the features of containment failure and radionuclide release from containers. It also provides a comparison with a safety assessment option assuming somewhat instantaneous disappearance of all container confinements.

These mechanisms and their influences were evaluated based on a criterion associated with the expected potential radiation impact on the population at the post-closure stage. To perform these assessments, a simplified integral calculation model was developed based on the GoldSim software [12], [13]. The model considered a hypothetical DGR involving the same engineered barrier designs as those proposed in the Finnish KBS-3V concept [14] except for the container designs: steel containers were considered instead of the copper ones. No description of this model has been provided here as it was considered as being beyond the scope of this paper. So far, let's note that the radionuclide composition of the waste was taken from [15], the parameters of the geosphere and biosphere — from [16], [17], respectively. Cross-verification calculations were performed to demonstrate the model efficiency. Still, the data obtained cannot be considered as a safety assessment of an actual facility: a few key parameters are not available yet. In the latter case, data on analogue objects were used, which can still be used for comparison purposes.

DGR saturation and container failure

The study considers a conventional steel container with RW having the internal dimensions of Ø 0.6 × 3 m and a shell thickness of 0.15 m (the external container dimensions — Ø 0.9 × 3.3 m).

DGR saturation at the post-closure stage

DGR volumes that are supposed to be filled with water at its post-closure stage can be divided into

[1] Radionuclide release from undetected seal failures in the copper container welds was considered as a source term.

easily accessible ones (macroscopic empty volumes such as the under-roof space, gaps between RW packages and backfilling elements in the excavations) and less accessible bentonite pore volume characterized with low permeability.

The saturation time t of the easily accessible volume V_0 can be calculated based on the below equation:

$$t = \frac{V_0}{Q_0}, \quad (1)$$

where Q_0 is the average GW flow saturating the DGR. Q_0 is expected to be quite isotropic and time-dependent — along the DGR saturation process, the air remaining in the excavations is expected to be compressed and no longer able to escape until its pressure is equal to the hydrostatic pressure at the disposal depth, P. At this level, $d = 500$ m, $P = \rho \cdot g \cdot d$, where ρ is the water density ($\approx 1{,}000$ kg/m^3), g is the acceleration of gravity (9.81 m/s^2) and $P \approx 5$ MPa.

A method described in [18] can be used to roughly estimate t providing that:

$$Q_0 = \frac{8rk\rho gd}{\mu}, \quad (2)$$

where r is the equivalent DGR radius, k is the average rock permeability, μ is the dynamic water viscosity. If considered, the back pressure from the compressed air would increase the saturation time by slightly more than 10 % [18] and was not included into these rough calculations.

Considering the DGR planar dimensions of 300×700 m $= 2.1 \cdot 10^5$ m^2, the radius of a circle covering a same area would account for $r = 258$ m. In the assessments, the dynamic water viscosity was assumed as being equal to $\mu = 10^{-3}$ Pa·s. The permeability, k, and the hydraulic conductivity, K, are related in the following way:

$$k = \frac{K\mu}{\rho g}. \quad (3)$$

Assuming a depth-average hydraulic conductivity $K = 10^{-9}$ m/s [16], k would be equal to 10^{-16} m^2, and $Q_0 \approx 3 \cdot 10^4$ m^3/year according to (2). According to an expert assessment of the expected easily accessible empty DGR volume of $V_0 = 3 \cdot 10^4$ m^3, t would amount to approximately one year.

If sodium bentonite with a dry density of 1.6 g/cm^3 is considered as a DGR volume subjected to long-term saturation, its hydraulic conductivity K, according to [19], [20], equals to $5 \cdot 10^{-14}$ m/s, porosity, $\varphi \approx 0.4$ and characteristic pore size, a, accounts to a micron fraction.

Assuming a hydraulic pressure gradient depending on the disposal depth d and on the depth of ground water (GW) penetration into bentonite x, GW speed is governed by Darcy's law and can be approximately calculated according to the below equation:

$$\frac{\mathrm{d}x}{\mathrm{d}t} = \frac{K}{\varphi} \cdot \frac{d}{x}. \quad (4)$$

Saturation time t that can be calculated from (4) assuming a buffer thickness x according to the following equation:

$$t = \frac{\varphi x^2}{2Kd}. \quad (5)$$

This approach does not account for the relationship between K, the pore space saturation and the capillary pressure (the pressure of GW "suction" into the bentonite pores). GW saturation front is assumed to be "flat" and the hydraulic pressure gradient can be calculated based on a ratio between the height of the water layer d and the thickness of unsaturated bentonite layer x. Capillary effects in partially saturated bentonite result in accelerated penetration and blurring of the GW front. Nevertheless, their accurate assessment is challenged by the lack of detailed information. The suction pressure ΔP given a pore size $a/2$ can be roughly estimated as a radius of a capillary curvature assuming that:

$$\Delta P = 2\sigma / a. \quad (6)$$

Assuming a surface tension coefficient of $\sigma = 7 \cdot 10^{-2}$ N/m at the GW-bentonite interface, the characteristic pressure change, given a pore size of 0.1 μm, equals to 1.4 MPa that is less than 30 % of the hydrostatic pressure (5 MPa). In case of larger pore size, it will be negligible. Thus, with regard to accuracy of the calculation, the capillary effects can be neglected. Therefore, given the assumed parameter values, the saturation time t for a 0.5 m-thick (x) bentonite buffer would amount to some 60 years.

The above assessment is considered quite simplified and true only for a buffer located near a water-conducting crack that would provide sufficient GW supply. The actual distance between water-conducting fractures in crystalline rocks (assuming their compliance with the rock suitability criteria) amounts to several meters, therefore, the effective buffer thickness x may turn out to be an order of magnitude higher than the accepted one. According to more accurate assessment of the saturation time in similar crystalline rock may range from several tens (if bentonite is located in the immediate vicinity of water conductive fracture) to several tens of thousands (if located far from it) of years [21].

More realistic assessments would imply that a number of physicochemical processes are considered in a comprehensive manner, including temperature changes, mechanical loads, chemical interactions, etc. Their consideration is beyond the scope of this article, but nevertheless should be taken into account in the safety assessments. Within the

KBS-3V concept, these issues have been addressed through certain criteria developed for the long-term performance of the engineered barrier system. In particular, the maximum temperature at the container-bentonite interface set at a level of no more than 100 °C to prevent water boiling during saturation (the pressure of the saturating water at a depth is above the atmospheric one and the boiling point is higher) and chemical transformations of bentonite; the minimum tolerable dry density of bentonite should ensure sufficiently low permeability, high swelling pressure and to prevent any microbial activity while maintaining certain plasticity level. Moreover, some restrictions are imposed on the content of hazardous corrosive contaminants in bentonite, etc. The features, events and processes accounted for the KBS-3V concept, namely the Finnish DGR design have been overviewed in [22].

Thus, *taking into account the scattering effect, the estimated time of bentonite buffer saturation 2,000 years appears to be conservative enough.*

Container corrosion and perforation

Let's consider holes in the container shell that either may go unnoticed under the packaging quality control process performed after the container is packaged with waste and sealed (most likely in the lid weld) or evolving along with the long-term corrosion process. In the first case, an undetected hole most probably won't exceed 1 mm in its aperture. Issues associated with the carbon steel container corrosion were considered in [23]. Estimates show that under aerobic conditions,[1] corrosion depth would not exceed 1.5 mm in case of homogeneous and 1.5 cm in case of local (pitting) corrosion. After all oxygen in the DGR is consumed, corrosion depth is evaluated to be about ~1.5 cm in the first 5,000 years given the expected elevated temperatures, and then, according to conservative estimates, the corrosion rate would accelerate to ~2 μm/year due to steel-bentonite interaction.

In the long-term, container's sealing capacity would depend on its ability to maintain the integrity of its shell under external pressure conditions (expected range of 5—15 MPa [23]) and thickness thinning due to corrosion. According to preliminary estimates [23], the containment function of a package is expected to last until the shell thickness remains more than ~5 cm. Further on, one may expect severe deformation and ruptures of the shall with apertures amounting to several centimeters.

[1] According to the assumptions ~10 years of storage, and ~100 years of saturation and consumption of the oxygen remaining in the DGR due to chemical reactions.

Assuming an initial container thickness of 150 mm, the estimated *time of its confinement failure* according to the criteria suggesting shell thickness reduction by 100 mm would amount to 5,000 years (correspond to 3 cm of aerobic corrosion) + 35,000 years (correspond to 7 cm/(2 μm/year)) ≈ *40,000 years*.

In the event of confinement failure, the corrosion rate inside the package may increase considerably due to radiolysis of the inflowing GW.

Perforated container filling with water

Before containment failure, the pressure level inside the packages is assumed to be equal to the atmospheric one, whereas after their failure, the water inflow is expected to be limited due to the low water permeability of the water-saturated bentonite.

If the inflow intensity gets higher than the evaporation intensity inside the container and the diffusion of vapors into bentonite, the container may start to be filled with water. However, if the pressure inside it, governed by the hydrogen releases due to the anaerobic corrosion of metal shells, its dissolution in water within container and the release of helium due to alpha decay, exceeds the external pressure, the water, on the contrary, would be squeezed out of the package.

This process can be evaluated quantitatively. The following relation describes the water flow running from bentonite into an opening of up to several centimeters governed by Darcy's law [24]:

$$Q_w = 2\pi\delta K \frac{P_s - P}{\rho_w g}, \qquad (7)$$

where Q_w is the water flow, m³/s, through an opening with a characteristic size (diameter) δ, m; K is the bentonite hydraulic conductivity close to the opening, m/s; P is the pressure inside the container, Pa; P_s is the hydrostatic pressure, Pa; ρ_w is the water density; g is gravity acceleration.

At $P_s = 5 \cdot 10^6$ Pa and $K = 5 \cdot 10^{-14}$ m/s, for decompression apertures $\delta = 0.5$ mm (characteristic opening size for an undetected welding defect and pitting corrosion), $\delta = 1$ cm and $\delta = 10$ cm (expected decompaction during container deformation), the characteristic rates of GW inflows into containers, Q_w, calculated based on (7), would be equal to $2.5 \cdot 10^{-6}$, $5 \cdot 10^{-5}$ and $5 \cdot 10^{-4}$ m³/year, respectively. Assuming the void volume of 0.2 m³ inside the container, one can calculate the corresponding container filling times t_{fil} — $8 \cdot 10^4$, 4,000 and 400 years.

Hydrogen generation inside a perforated container

Iron corrosion under anaerobic conditions occurs according to the reaction:

$$3Fe + 4H_2O \rightarrow Fe_3O_4 + 4H_2, \qquad (8)$$

magnetite and hydrogen are the oxidation products.

Since magnetite volume is approximately three times more than the volume of the original shell metal (one magnetite molecule is formed from three molecules of iron), corrosion products will reduce the empty space inside the package and the size of the opening through which the water inflow occurs. Nevertheless, further on this effect is not considered conservatively.

The rate of hydrogen formation in reaction (8) can be described as follows:

$$\dot{n}_{H_2} = \frac{\dot{s}_{corr} S_{Fe} \rho_{Me} x_{Fe,Me} \nu_{H_2,Fe}}{M_{Fe}}, \quad (9)$$

where $\dot{n}_{H_2}$ is the rate of hydrogen generation, mol/s; $\dot{s}_{corr}$ is the metal shell corrosion rate, m/s; S_{Fe} is the corroding surface area, m^2; ρ_{Me} is the container metal density, kg/m^3; $x_{Fe,Me}$ is the concentration of iron in metal, kg/kg; $\nu_{H_2,Fe}$ is the stoichiometric ratio of H_2 and Fe in the oxidation reaction; M_{Fe} is the molar mass of iron, g/mol.

Assuming $\dot{s}_{corr}$ = 20 μm/year[1], S_{Fe} = 6.2 m^2, ρ_{Me} = = 7,000 kg/m^3, $x_{Fe,Me}$ = 0.95, $\nu_{H_2,Fe}$ = 4/3 (see formula 8), M_{Fe} = 55.8 g/mol, the rate of hydrogen formation amounts to **20 mol/year**.

Required water consumption for the reaction to occur can be calculated as follows:

$$\dot{m}_{H_2O} = \dot{n}_{H_2} M_{H_2O} \nu_{H_2O,H_2}, \quad (10)$$

where $\dot{m}_{H_2O}$ is water consumption, g/year; M_{H_2O} — molar mass of water, kg/mol; ν_{H_2O,H_2} is the stoichiometric ratio of H_2O and H_2 in (8). At $\dot{n}_{H_2}$ = 20 mol/year, M_{H_2O} = 18 g/mol, ν_{H_2O,H_2} = 4/4, $\dot{m}_{H_2O}$ amounts to 360 g/year. This value may be provided only by sufficiently large (several centimeters) opening: hydrogen generation in case of smaller opening is expected to be governed by the rate of GW inflow inside the container.

Experiments [25] show that when the gas pressure within bentonite shall reaches the level amounting to the sum of the hydrostatic pressure and the pressure of water-saturated bentonite, a breakthrough occurs through a crack emerged in the bentonite. Then, the pressure in the container cavity drops by approximately 2 times, which is nevertheless enough to provide long-term preservation of the gas channel formed in the bentonite (about several weeks if the internal pressure is maintained). Assuming the additional pressure from the swelling bentonite of 3 MPa, the calculated "breakthrough pressure" of gases amounts to P_s + 3 = 8 MPa.

If one considers hydrogen in the container as an ideal gas, then to achieve a hydrostatic pressure of 8 MPa in its cavity (0.2 m^3) at a temperature of 20 °C (several hundred years after DGR closure) 658 moles of hydrogen would be required[2]. At a generation rate of 20 mol/year, this amount is expected to be generated approximately every 33 years providing a back pressure sufficient for H_2 to break through the bentonite and, therefore, to slow down the water inflow rate and periodically increase the confinement capacity of the package.

Thus, hydrogen generation plays a somewhat dual role: on the one hand, it contributes to a backpressure inside the container remnants and a decreased GW inflow rate, on the other hand, it triggers periodic breakthroughs of the bentonite barrier which may contribute to partial release of radionuclides in the form of a gas-water phase into the surrounding rocks.

Model describing the effects of container corrosive degradation

The following features and processes govern radionuclide releases from waste containers in terms of the spread observed in their starting times and duration:

- differences in the water conductivity and location of water-conducting fractures in rock relative to the saturated buffer;
- different apertures of the openings formed in a corroded container that has lost its strength;
- back pressure of hydrogen changing due to the previous process, prompted by the anaerobic corrosion and responsible for the GW inflow into the damaged container.

Upon container failure and its saturation with GW contacting with the vitrified high-level waste (HLW), the corrosion rate at the inner container surface gets increasingly accelerated.

Each of the three listed processes may delay the failure time and the release of radionuclides from the container by about 1,000 years.

In the calculation model, process of radionuclide release is described by container failure within certain time interval (40,000, 40,000 + t_{sh}), where t_{sh} can vary within a range of 1,000—10,000 years.

Radionuclide dispersion during radionuclide transport in a fractured medium

Radionuclide transport along water-conducting fractures is seen as the key one for crystalline rocks. Generally speaking, even within a single fracture

[1] Given the radiolysis of GW flows entering the container, the corrosion rate is assumed to get an order of magnitude higher.

[2] These calculations do not take into account hydrogen dissolution in the water inside the container. However, its maximum concentration at a pressure of 8 bar is about 12.5 mg/l (2.5 g in 0.2 m^3), which can be neglected — in 658 moles of hydrogen its mass would be equal to some 73 g.

such transport is not considered uniform, but is concentrated in some individual channels formed due to the changing aperture and the presence of the fracture filling material. Variations in their size and water conductivity, as well as the layout of transport trajectories described by intersecting fracture network governs the spatiotemporal dispersion of radionuclide transport parameters.

Radionuclide transfer into the rocks gets retarded due to diffusion into stagnant GW-saturated zones formed by microcracks surrounding the main fracture and the pore space of the rocks, as well as by radionuclide sorption on the rock and on the fracture filling. These processes may vary within the radionuclide transport trajectories and also govern their dispersion. However, these aspects have not been accounted for in this study.

Considered below is only the dispersion caused by advective transport through a network of water-conducting cracks. The simplified model considers advective homogeneous transfer of any radionuclides emerging from all containers along a single water-conducting crack characterized by its width W and a GW flow Q. It is believed that this process depends on the trajectory length L to the nearest discontinuity in the rock formation with an upward flow being the route for radionuclide inflows into the near-surface sedimentary layer, where a well may be constructed in the future for irrigation and drinking water supply purposes. Radionuclide (RN) transport along a fault with an upward flow is considered to be quite fast compared to RN movement along a fracture, and is conservatively assumed as being instantaneous.

[16] analyzes data on well-studied host rocks of the Swedish and Finnish DGR showing that the average data characterizing advective transport in a fractured medium can be obtained based on appropriately selected parameters W, L, Q, aperture of a conventional fracture (a) and hydraulic pressure gradients. The integral parameter for this process is the so-called crack resistance, or the parameter $F = W \cdot L/Q$. This approach and all available information have been extrapolated to select appropriate parameters for the preliminary characterization of the DGR proposed at the Yeniseiskiy site [16].

However, it seems clear that radiation DGR impact assessments would turn to be overly conservative if the transfer of any radionuclides from all containers dispersed over a large area is limited to a single water-conducting fracture. To take into account the spatiotemporal distribution of radionuclides released into the environment, parameters W, L, Q have been selected so that the experimental distribution of the parameter F for the Finnish Olkiluoto massif, which was considered as the closest analogue of the Yeniseiskiy site, could be reproduced while maintaining the most probable values considered characteristic for the average advective transport. It is well described by the lognormal law with a range of values corresponding to 5—95 % quantiles of two to three decimal orders depending on the fracturing models [26]. It is interesting to note that a similar approach was used in the safety assessment of the Japanese DGR [27], in which the variability of rock properties was modeled by lognormal distribution of the water conductivity of a single fracture. The distribution of logarithm of water conductivity in fractures was modeled assuming normal distribution with a width at half-height of approximately 2.5.

F parameter distribution was selected using the Monte Carlo method by sampling random values from varying distributions of parameters W, L and Q. These were calculated in the GoldSim software.

In this study, the calculation model for parameter W involves a lognormal distribution with a mean value of 5 m, a standard deviation (SD) of 7 m "cut off" within the range of the minimum and maximum values of 0.2 and 20 m. Normal distribution was used for parameter L with an average value of 1,000 m, SD = 1,500 m "cut off" within the range of the minimum and maximum values of 200 and 500 m. Lognormal distribution was used for parameter Q with an average value of $8 \cdot 10^{-3}$ m³/year, SD = $9 \cdot 10^{-3}$ m³/year "cut off" within the range of the minimum and maximum values of $5 \cdot 10^{-4}$ and 0.05 m³/year. Figure 1 shows the resulting parameter F distribution corresponding to their convolution.

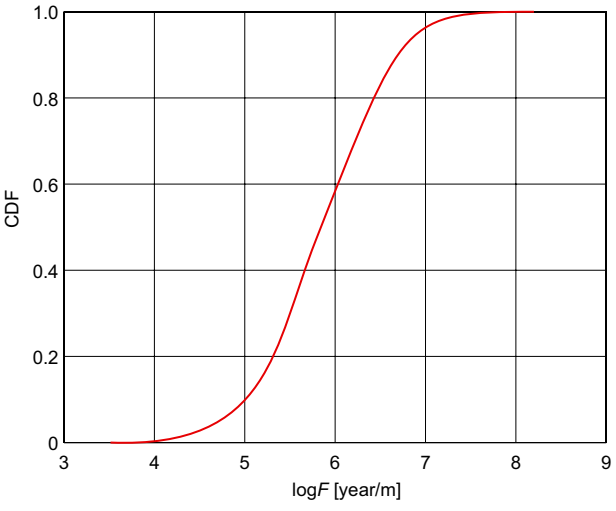

Figure 1. Accepted cumulative distribution of parameter F characterizing the dispersion of the advective radionuclide transport in the calculation model

Assessments based on a calculation model

The results obtained are presented for ^{129}I defining the total predicted dose (Figure 2) for the population under accepted model parameters. This radionuclide is considered almost non-absorbable and "stable" (its half-life is almost 16 million years) and can be viewed as an example of a most conservative contaminant.

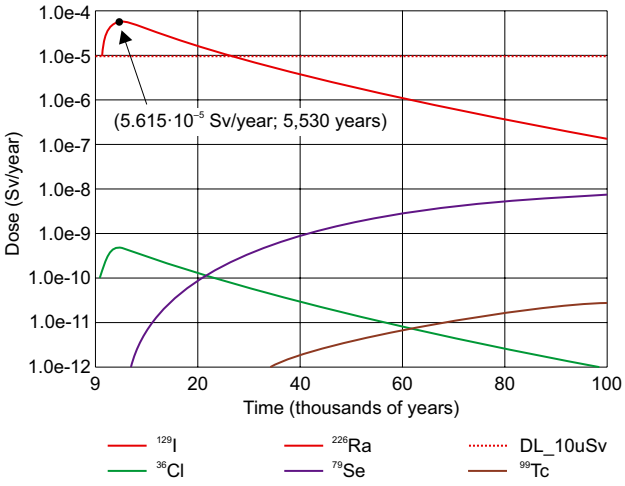

Figure 2. Dependence between the expected annual doses to population and the radionuclide time being mainly responsible for the long-term hazards posed by radioactive waste (corresponding to spontaneous container "disappearance" in 1,000 years; up to 100,000 years after DGR closure)

To evaluate the influence of the studied processes — the time-distributed radionuclide releases from containers and their transport along the rock crack network — a criterion evidencing the achievement of the maximum predicted annual dose at a corresponding point in time was used (Figure 2 shows the initial concept, under which all parameters were assumed to remain constant). In the next calculation model, container failure was described by a uniform distribution covering a time interval of 2,000 years "shifted" by 40,000 years from the time of DGR closure. RN transport along different trajectories in the cracks was simulated using the Monte Carlo method given random samples from the assumed distributions of parameters W, L, Q and crack aperture a (1,000 realizations, Figure 2). The "maximum dose-time" criterion was assumed for the average dependence across the samples. To describe the parameters showing the dependence between the expected average dose and the time, one may select the listed parameters of the fractured medium. By minimizing the sum of relative deviations from the maximum dose and the time of its occurrence, the following values were calculated (two "best" sets of values providing an approximation accuracy of no less than 21%):

W, m	L, m	Q, m³/year	a, m
7.67	4,683	$4.306 \cdot 10^{-2}$	$3.975 \cdot 10^{-4}$
9.56	4,514	$4.977 \cdot 10^{-2}$	$4.350 \cdot 10^{-4}$

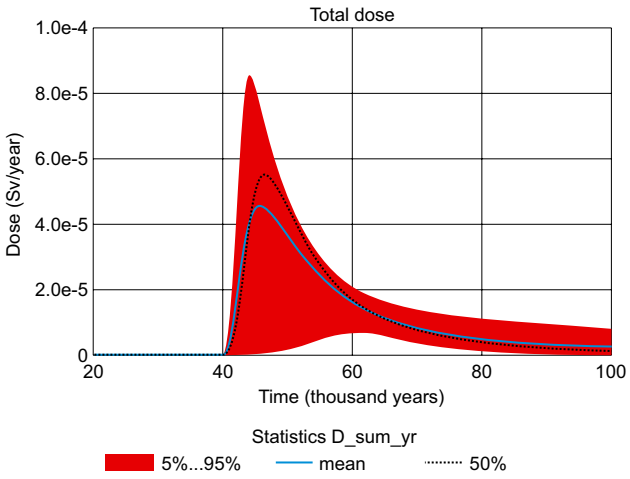

Figure 3. Dependence of annual dose from time modelled with an account taken of the effects associated with container failure and radionuclide release through a discrete fracture network, quantiles 5% – 95%, as well as the mean (Mean) and median values (50%)

Figure 3 shows the time dependence for the expected population exposure modelled in GoldSim with an account taken of all the effects considered including distributions of the parameters responsible for radionuclide transport through a discrete fracture network.

Table 1 presents the criterion "maximum dose — time since DGR closure" calculated for different parameter sets characterizing the spatiotemporal dispersion.

Table 1. The maximum expected doses for ^{129}I and the time of their occurrence taking into account the effects of the time-distributed radionuclide release from the container and their transport in a distributed crack network

Parameter set	All constant	Considering time-distributed radionuclide release from a container	Considering RN transport along different trajectories in the cracks (for an averaged one by realizations curve)	Both effects (for an averaged curve)
Max. dose, Sv/year	$5.616 \cdot 10^{-5}$	$5.564 \cdot 10^{-5}$	$4.612 \cdot 10^{-5}$	$3.263 \cdot 10^{-5}$
Time, upon which the max. dose is attained, years	5,530	46,610	5,000	50,100

Considering a most conservative (stable and non-absorbable) impurity, the influence of all above temporary dispersions corresponding to RN releases from the safety barriers was deemed as insignificant. Thus, the time-distributed leakage of ^{129}I from a container over 2,000 years assuming a delay of 40,000 years reduce the maximum dose by less than 1 % and extend the time needed for reaching the maximum dose by 41,080 years. If the distribution of transport times along the cracks is accounted for, average maximum dose would drop by approximately 18 % and the time needed to achieve the maximum dose would decrease by 530 years. Both effects provide a total reduction in the maximum dose by 42 % and increase the time needed to reach the maximum level by 44,570 years.

Such low impact of release time distribution and its effect is quite specific since all the contaminants flowing relatively quickly beyond the engineered barrier system get released into the geosphere and their further transport occurs at a much slower rate. There the contaminants get somewhat "stacked", which basically explains the retardation and dispersion of radionuclide releases into the biosphere. If one assumes that iodine release from a container is distributed over not two, but 10 thousand years, the maximum dose may drop by approximately 15 %, whereas the time needed to reach the maximum level is expected to raise to 12,390 years.

Naturally, more considerable dose reduction can be achieved in case if such reduction is associated with time-distributed release of radionuclides with their half-lives being comparable to the characteristic delays (some several tens of thousands of years or less).

Conclusion

^{129}I hold a special place among the radionuclides contributing to the long-term DGR's radiation hazard. In HLW generated from the radiochemical SNF reprocessing, its concentrations depend on the content of insoluble phases in process solutions and glass mass, for example, iodate compounds (IO_3^-), and insoluble forms such as colloids of silver iodide (which is one of the uranium fission products) [28]—[30]. The portion of iodine remaining in vitrified HLW is (very conservatively) estimated at 4% of its total activity in SNF [31]. Nevertheless, due to its long half-life (almost 16 million years) and lack of any considerable sorption by the protective barrier materials, it is this radionuclide that basically contributes to the hazard posed by DGR in a timeframe of ~100,000 years. This fact is confirmed by a few international assessments focused on the radiation characteristics of the geological repositories for SNF and HLW (see, for example, [2], [3], [32], [33])[1].

Apparently, **none** of man-made containers can stay tightly sealed for millions of years, thus, environmental releases of radionuclides remaining hazardous within these time frames are expected under all DGR designs. The only meaningful measure is the RN transport retardation and dispersion by the DGR's safety barriers. Apparently, the effect of fracture-wise advective transport distribution, described in this study, is insignificant, and the retardation, as well as the transport "dispersion", are basically provided by matrix diffusion — advective transport process is superimposed on the one of radionuclide penetration into microcracks and the water saturated pore space of the rocks surrounding the fracture. Therefore, in case of "problematic" radionuclides, the DGR safety strategy is not "Concentrate and Retain", but rather "Dilute and Disperse" standing for controlled limitation of releases, which has proven to be a workable method providing safety at existing radiation hazardous facilities [34]. In particular, this approach is applied in France, where most of the iodine activity from SNF reprocessing is currently discharged into the Atlantic Ocean [35]. This solution is considered safer as compared to the one suggesting that high ^{129}I activities are concentrated in a DGR.

Thus, most corrosion-resistant containers can be used to dispose of high activities of weakly absorbable radionuclides with their half-lives of ~ 10^6 years in case of DGR construction in crystalline rocks: these should retain their sealing capacity within a timeframe comparable to the half-lives of most hazardous radionuclides. In addition to already mentioned copper containers, silicon carbide packaging is being considered as well [36]. Addition of selective sorbents ("getters") to the disposal containers can be also considered as an alternative or additional safety barrier option deserving further evaluation. [37] overviews potential materials used as sorbents for ^{129}I and ^{99}Tc. Copper and its oxides found in the RW container materials may provide some slight sorption of iodine in a moderately alkaline environment [38], [39].

It should be noted that the use of any non-naturally occurring materials as long-term DGR safety barrier structures would require some in-depth feasibility study. For clayey rocks, conservative

[1] In [32], ^{135}Cs and ^{79}Se head the list of hazardous nuclides in terms of radiation impacts produced by HLW and considering a timeframe of some 1 million years, with ^{129}I holding the third place. Our estimates covering a timeframe of 1 million years also evidence certain shift in the radiological hazard towards ^{79}Se, ^{99}Tc and ^{226}Ra. However, these effects have not been considered in this paper.

contaminant transport can be retarded for much longer periods, i. e., some 10^6 years [40].

This paper examines the time-distributed processes of DGR saturation with GW after its closure, the features associated with the container's sealing capacity and its loss, radionuclide releases from the container and the dispersion processes due to RN transport in a fractured medium.

Calculations showed that assuming a conditionally stable and non-absorbable contaminant (assuming most conservative option), the influence of its time-dependent release dispersions from the safety barriers would be insignificant. ^{129}I release from a container assuming a retardation of 40,000 years for 2,000 years is expected to yield some slight decrease in the maximum dose (< 1 %). The latter one can drop to about 20 % if contaminant transport time distribution through fracture network is accounted for. If both features are accounted for simultaneously, the effective dose may drop by 40 %. All the released contaminants get "stacked" in the geosphere, which mainly governs radionuclide release dispersion into the biosphere.

To address the disposal challenge associated with high activities of weakly absorbable radionuclides with their half-lives of over 10^5 years, assuming their disposal in crystalline rocks, corrosion-resistant containers can be used. For example, containers fitted with a copper shell or made of silicon carbide. Another possible option — the use of selective sorbents that would sorb most dangerous nuclides. The use of any material having no natural analogs should be preceded by appropriate scientific and experimental feasibility studies.

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