INFLUENCE OF DEGRADED CEMENT BACKFILL ON RADIONUCLIDE RELEASE CONSIDERING RW CLASS 1 DISPOSAL IN SUPER-CONTAINERS

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The paper evaluates the influence of degraded cement backfill on contaminant release under geological radioactive waste disposal concept. The concept provides that the waste surrounded by compacted bentonite is emplaced in "super-containers" into vertical boreholes within cement backfill. The paper demonstrates that simple compartment and more precise 3D-model assessments yield similar results. It also shows how the calculations are affected by mesh discretization and ground water flow in degraded backfill. Considering moderate rock suitability criteria, degraded backfill increases maximum contaminant release not more than by an order of magnitude, which is substantially less compared to other safety assessment uncertainties.

Keywords: *radwaste geological disposal, crystalline rock, "super-container" with compacted bentonite, cement backfill, advective transport, diffusion transport, radioactive waste.*

Currently, the so-called super-containers (SC) placed into vertical boreholes with the voids backfilled with cement-based material are considered as an alternative concept for RW Class 1 disposal (RW-1) at the Yeniseiskiy site [1]. The SC is a metal shell made of low-carbon steel (possibly with a copper coating) holding a primary package with RW-1 surrounded by compacted bentonite blocks. The primary package itself holds drum(s) with vitrified high-level radioactive waste (HLW).

This concept avoids a number of problems considered typical for the "traditional" concepts of SNF and HLW disposal, in particular, the Scandinavian KBS-3V concept [2] proposed for crystalline fractured rocks. In particular, the hardening cement backfill provides RW-1 package disposal in relatively deep 75-meter boreholes as specified under the repository designs with less thick-walled containers, thus, increasing the disposal capacity, providing much lower steel corrosion rate compared with the bentonite environment, as well as the compatibility of compact RW Class 1 and 2 disposal in a deep repository with a cement buffer [1].

However, in practice, the concept [1] has been studied little and suggests the use of materials considered poorly compatible as regards the long-term integrity of the safety barriers. Degradation of the cement backfill in the long run (thousands - tens of thousands of years) and corrosion of steel containers is seen as a key drawback of this concept suggesting that the bentonite backfill with RW packaging will be surrounded by a permeable material with its barrier properties as regards poorly sorbed radionuclides being similar to the sand.

This study evaluates the characteristics of engineered barriers used under a new concept and the way they will differ from those expected under the "traditional" one, when the container is emplaced into a borehole surrounded by compacted bentonite. The study involves consistent comparison of a "traditional" model considering the emplacement of a bentonite block with a contaminant into a borehole intersected by a water-conducting fracture and the SC concept with the same bentonite block emplaced into a borehole and surrounded by a permeable material. Both semi-analytical method and more accurate numerical model were used in the study.

The simple assessments presented in the article should reveal the problems having most important effect on the safety of the engineered barriers proposed under the disposal concept and, possibly, requiring more accurate analysis under further studies.

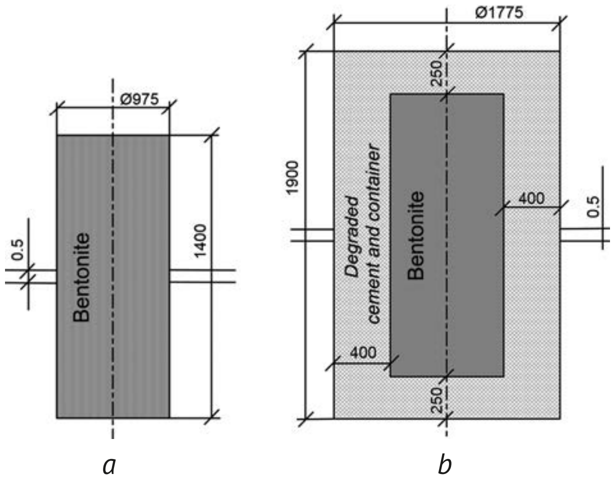


Figure 1. Bentonite cylinder crossed by a water-conducting fracture (a); bentonite cylinder in a permeable shell (b)

Initial data

Two models were considered. In the first case (option A), a bentonite cylinder 975 × 1,400 mm containing 1 kg of a non-absorbable and stable tracer (contaminant) is crossed in the middle by a water-conducting fracture in the host rock (HR) with an aperture of 0.5 mm (see Figure 1a). In the second case (option B), the same cylinder is surrounded by a shell, conditionally called "sand", which is literally represented by degradation products of the cement surrounding the super-container in the new deep disposal concept and a steel SC. The thickness of the "sand" on the sides amounts to 400 mm, at its side walls — 250 mm each.

The HR was considered waterproof and the advective transport of contaminant in bentonite was considered negligible. Relevant material parameters are listed below.

Bentonite:

bulk density	1.7 g/cm^3;
porosity	0.4;
effective diffusion coefficient	10^{-10} m^2/s.

Bentonite is considered water-saturated, the fracture is filled with water (the diffusion coefficient of water is 10^{-9} m^2/s).

Degraded backfill ("sand"):

bulk density	1.7 g/cm^3;
porosity	0.2;
effective diffusion coefficient	$2{\cdot}10^{-10}$ m^2/s;
water conductivity	1 m/day.

Two groundwater flow rates (GW) in the fracture crossing the borehole were considered: 1 and 20 m/year. The first rate corresponds to a relatively undisturbed rock, the second — to a large water-conducting fracture. The task was to calculate the dynamics considering a conservative tracer release into a water-conducting fracture.

Option A. Analytical estimates

In many cases, radionuclide transfer through a system of multiple barriers depends on their mutual interaction and the time of their distribution within each barrier is much less than the characteristic time of radionuclide transport between them. Thus, assuming their "instantaneous mixing", in each case, the system of safety barriers (SB) in a deep disposal facility can be considered in a chamber model approximation, where each chamber is characterized by a mass fraction of the contaminant (radionuclide) contained in it and transferred per unit time, λ. The permeability of engineered SB such as bentonite is quite low and diffusive transport prevails.

The following approach provides a transition from the diffusion equations to the chamber model: the flow of a radionuclide J [kg/s] in a porous medium through area A [m^2] depends on its concentration gradient in solution C [kg/m^3]:

$$J = -D_e{\cdot}A{\cdot}\mathrm{grad}\,C, \tag{1}$$

where D_e [m^2/s] is the effective diffusion coefficient. In many cases, expression (1) can be obtained as a product of a fictitious flow carrying a contaminant, Q [m^3/s], and the difference in the impurity concentrations:

$$J \approx -Q\Delta C = -Q(C_1 - C_2), \tag{2}$$

where C_1 and C_2 are the radionuclide concentrations at the front and the back edge of the engineered barrier, respectively. If $C_2 \ll C_1$, equation (2) can be represented as follows:

$$J = \frac{\partial m}{\partial t} \approx -QC_1 = -\frac{Q}{R\varepsilon V}m = -\lambda m, \tag{3}$$

the solution of which is the dependence of the type $m = m_0 e^{-\lambda t}$, which is commonly used in chamber models. The expression providing transition from the radionuclide concentration C to its dissolved mass m in the volume V ($m = CVR\varepsilon$) takes into account the porosity of the medium (ε) and its sorption retardation coefficient $R = 1 + \frac{\rho K_d}{\varepsilon}$ (where ρ is the bulk barrier density and K_d is the sorption distribution coefficient for the considered radionuclide).

Let us analyze the contaminant diffusion through the surface of a bentonite cylinder with radius r and height S into a plane infinite fracture intersecting it with an aperture of $2b$. If diffusion through the ends of the cylinder is not considered for the reasons of conservatism, then solution of a similar problem in [3] can be used: it considers diffusion through an annular layer with an inner radius r_1 and an outer radius r_2. In this case, the problem is reduced to a diffusion through a cylinder with an equivalent radius ρ. In [3], for the contaminant flow at the entrance of fracture J, the following expression was obtained:

$$J = \pi^2 D_e (r_1 + r_2) \frac{C_1 - C_2}{\ln\left(\frac{\rho}{b}\right)}, \qquad (4)$$

where C_1 and C_2 are the contaminant concentrations in the layer and in the HR in the fracture, $2b$ is the fracture aperture. In (4):

$$\rho = \sqrt{\frac{2dS}{\pi}}, \quad d = r_2 - r_1, \qquad (5)$$

where D_e is the effective diffusion coefficient of the layer or in approximation (3):

$$Q = \frac{\pi^2 D_e (r_1 + r_2)}{\ln\left(\frac{\rho}{b}\right)}. \qquad (6)$$

Assuming that a cylinder can be considered as a special case of an annular layer with an inner radius $r_1 = 0$ and $r_2 = r$, the volume of the cylinder can be calculated as follows:

$$V = 2\pi r^2 S. \qquad (7)$$

Diffusion of radionuclides through the annular bentonite layer can be approximated by a chamber model with a time constant, λ_1 [year^{-1}]:

$$\lambda_1 = \frac{\pi D_e}{rSR\varepsilon \ln\left(\frac{\rho}{b}\right)}, \qquad (8)$$

where ε is the open bentonite porosity, and the retardation coefficient for the non-absorbable contaminant is $R = 1$.

In addition to diffusion in bentonite, the contaminant penetration into the fracture is limited by the time of GW contact with the backfill, and the impurity concentration in the GW at the borehole boundary is less than in the pore water of the barrier material contained in it. This mechanism is called “fracture resistance”, and the effective flow for approximation (3) was calculated [4, 5] based on the balance between the influx of contaminants into the fracture driven by diffusion and the advective release of contaminants along the fracture. The corresponding water flow through the boundary layer Q is:

$$Q = 4 \cdot 2b\sqrt{urD_w}, \qquad (9)$$

where D_w is the groundwater molecular diffusion coefficient and u is the GW rate in the fracture. The corresponding time constant in approximation (3), λ_2 is calculated by dividing Q in (6) by the bentonite volume (V) and its porosity (ε).

Thus, for option A, contaminant release from bentonite is described by two time constants λ_1 and λ_2, and the ultimate time constant describing the release into fracture λ_b in a chamber model approximation is calculated based on the following expression:

$$\frac{1}{\lambda_b} = \frac{1}{\lambda_1} + \frac{1}{\lambda_2}. \qquad (10)$$

Figure 2 presents analytical solutions obtained under these assumptions for the contaminant flow from bentonite into the fracture.

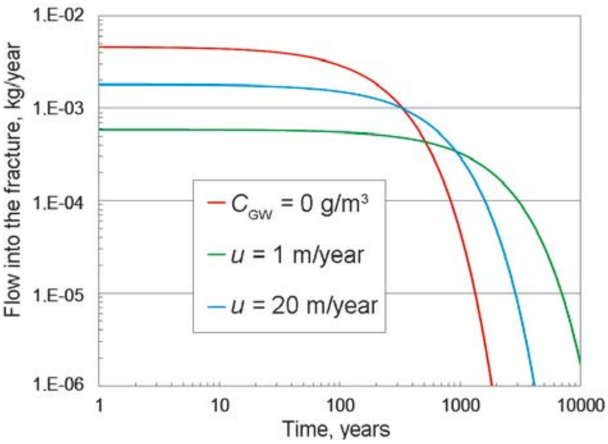


Figure 2. Conservative contaminant flow into a fracture crossing a borehole filled with bentonite considering different groundwater flow rates in the fracture (option A)

The upper curve ($C_{GW} = 0$ g/m^3) corresponds to the "zero" boundary conditions (GW rate $u = \infty$); it is assumed that the contaminant concentration in the GW at the fracture boundary is always equal to 0, i. e., the contaminant is instantly carried away. The higher GW flow rate, u, corresponds to a greater dilution of the contaminant in GW and its increased flow from bentonite.

Option B

Under option B, bentonite is surrounded by a permeable backfill and advective contaminant transport from the bentonite by groundwater flow is initiated through it. Diffusion and advection were modeled using GoldSim [6, 7]. To describe the diffusion radionuclide release into a fracture from the backfill, the chamber model approximation (3)—(10) was used: relevant capabilities were also provided for by GoldSim.

Since water permeability of the degraded backfill is several orders of magnitude higher than the water permeability of the HR, the so-called flow focusing occurs — the GW is “drawn” into the borehole and their flow through it turns out to be greater than in the undisturbed GW. In [8], the planar GW flow through a cavity with a characteristic size w was considered. It was found that the flow "focusing" occurs in a region where GW flows around the cavity with a characteristic size of $2w$. For a three-dimensional cavity, it can be approximately assumed that the flow through it is determined by a hypothetical fracture, which has a characteristic area the size of which is calculated as the product of its aperture ($2b$) and the width being equal to the double dimension of the cavity occupied by the backfill (in our case, the diameter of the backfill can be taken equal to the borehole diameter $2r$). GW flow through backfill q will be equal to:

$$q \approx 2 \cdot 2b \cdot 2r \cdot u. \quad (11)$$

As the numerical calculations presented below and in [8] show that the backfill degradation mainly occurs in the area adjacent to the fracture crossing the borehole, and the GW flow also tends to focus in this direction. Thus, even for a completely degraded backfill the GW flow will mainly pass only through a relatively thin layer of material adjacent to the fracture. It was shown in [9] with adequate accuracy that the GW flow, q, flowing through the borehole can be calculated as the flow through the equivalent volume (“plug”) of a continuous porous medium (Figure 3), the length of which is 3—8 fracture apertures depending on the relative orientation of the fracture and the intersected volume. Given a fracture aperture of $2b$, the plug length dplug was taken equal to $10b$. For the plug model, the hydraulic GW head gradient in the undisturbed

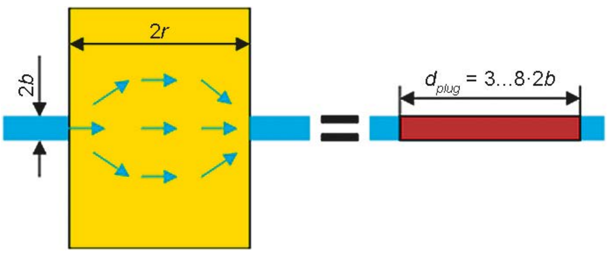


Figure 3. Equivalent volume model for a backfill capable of advective water transport

HR, i, will increase by $r/d_{plug} = r/5b$ times (in this case, r is the borehole radius) since the head drop previously occurred at the borehole diameter ($2r$) will be now observed along the length of the "plug". If the water conductivity of the "plug" is equal to K_{plug}, then, by equating the flows flowing through the borehole and governed by the GW transmissivity T the following equation can be obtained:

$$q = iTW = i \cdot \frac{r}{5b} \cdot K_{plug} \cdot 2b \cdot W = K_{plug} W i \frac{2r}{5}, \quad (12)$$

where W is the width of borehole intersection with a fracture, or:

$$K_{plug} = \frac{5T}{2r}. \quad (13)$$

If the water conductivity coefficient for the backfill $K \gg 5T/2r$, then the GW flow into the borehole can be described by formula (11). At $K = 5T/2r$, the water flow through the disturbed volume depends on the flow in the fracture, and if $K < 5T/2r$, then the backfill hydraulic resistance is sufficiently high and the GW starts flowing around the borehole.

In [9], the distribution problem for an advective flow along the height of the borehole was addressed as well. In case of $K = 5T/2r$, when the backfill is already slightly resisting to the GW flow, over 90% of its total volume passes through the area, the height of which does not exceed 10% of the borehole one. In the considered case, the flow is concentrated in the backfill layer adjacent to the fracture being no more than 20 cm in its height.

Given the expected transmissivity of fractures at the target depth of the Yeniseiskiy site $T \sim 10^{-9}$ м²/с [10] and $r \approx 0.89$ m, the criterion (13) is equal to $5 \cdot 10^{-9}/2 \cdot 0.89 = 2.8 \cdot 10^{-9}$ m/s $\approx 2.4 \cdot 10^{-4}$ m/day. When the water conductivity coefficient of the degraded backfill is $K = 1$ m/day, $K \gg 5T/2r$ and the GW flow through the backfill is described by formula (11).

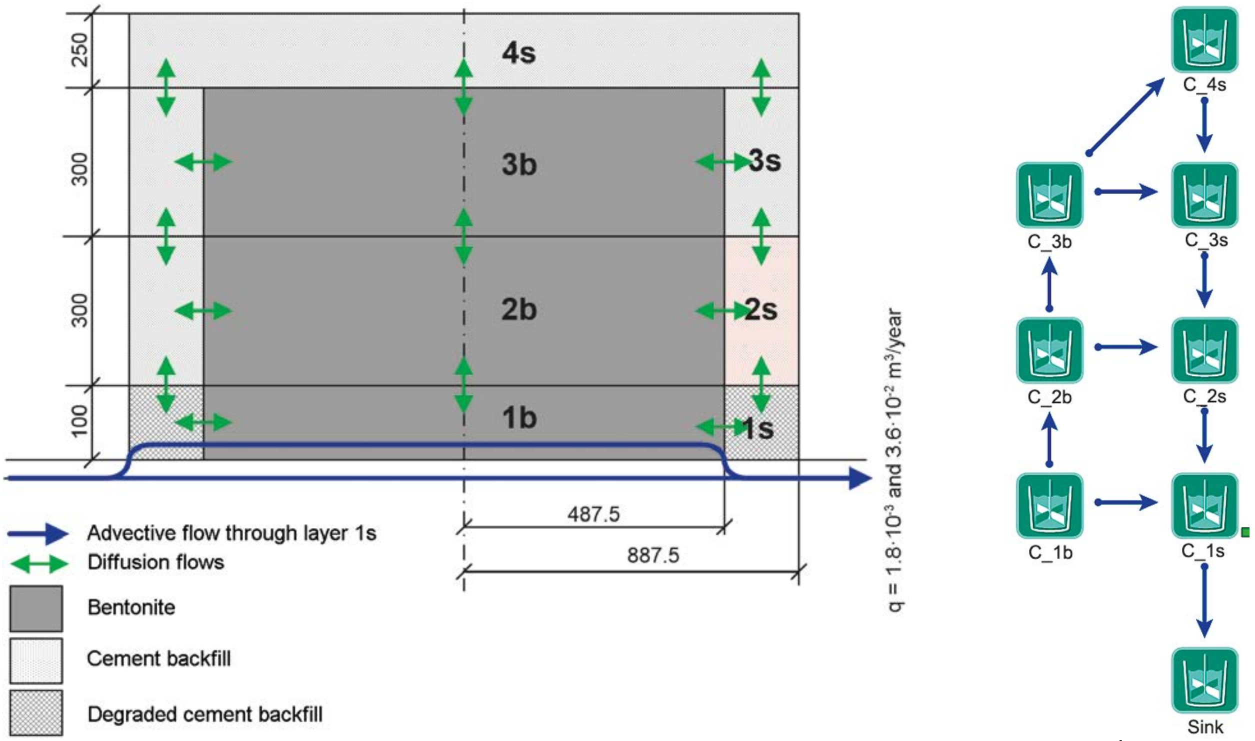
Further assessments were based on the geometry shown in Figure 4a (the upper part of the model is shown in the Figure, while the lower part is identical).

The advective flow through the degraded cement backfill layer (1s in Figure 4) q is equal to:

$$\begin{aligned} &q\,(1\ \text{m/year}) = 2 \cdot 2b \cdot 2r \cdot u = \\ &= 2 \cdot 5 \cdot 10^{-4}\ (\text{m}) \cdot 1.775\ (\text{m}) \cdot 1\ (\text{m/year}) = \\ &= 1.78 \cdot 10^{-3}\ (\text{m}^3/\text{year}) \\ &q\,(20\ \text{m/year}) = 2 \cdot 2b \cdot 2r \cdot u = \\ &= 2 \cdot 5 \cdot 10^{-4}\ (\text{m}) \cdot 1.775\ (\text{m}) \cdot 20\ (\text{m/year}) = \\ &= 3.55 \cdot 10^{-2}\ (\text{m}^3/\text{year}) \end{aligned} \quad (14)$$

This flow is concentrated on the area of the degraded backfill being 20 cm tall.

Figure 5 presents the results calculated for option B. It can be seen that the maximum contaminant flow for $u = 1$ m/year is 1.65 10^{-3} kg/year (after



a *b*

Figure 4. Geometry of a model with a degraded backfill adjacent to the fracture (a) and its representation in the form of "cell" elements in GoldSim (b)

19 years); for $u = 20$ m/year — 0.0179 kg/year (after 9 years).

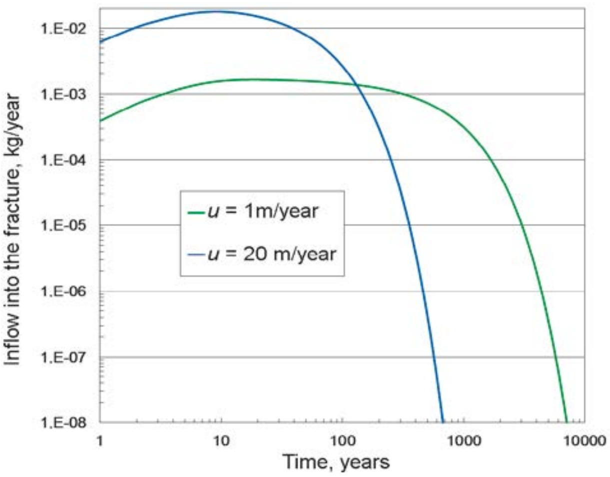


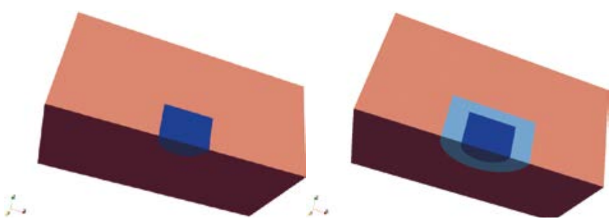
Figure 5. Conservative contaminant flow into a fracture crossing degraded backfill considering different groundwater flow rates (option B)

Problem modeling in the GeRa code

For the cross-verification of the obtained data, this problem was also solved using three-dimensional flow and transport modeling in the GeRa code [11]. The modeling area was represented by a rock fragment with a fracture, a bentonite cylinder and a "sandy" shell (under option B) according to a conceptual model, Figure 1. The size of the rock mass 5 × 5 × 5 m was selected purposely so that the model could cover the entire area of GW flow perturbation by the container. The flow was set using boundary conditions of the 1st kind (the rates of flows being far from the borehole, u, were equated to Darcy velocities and were set through the corresponding head gradients, i: $u = i{\cdot}K_f$. Using the empirical dependence of the crack aperture ($2b$) on its transmissivity (T) from [12], $2b\,(\text{m}) = 0.117\ (T\,(\text{m}^2/\text{s}))^{1/3}$ and $K_f = 2b{\cdot}T$, the water conductivity coefficient in the fracture was assumed as $K_f = 13.5$ m/day.

The rest of the model parameters are given above. For symmetry, the calculation covered only 1/4 of the simulation area (as shown in Figure 6).

Triangular prismatic grid was used in the calculations involving numerical schemes of the finite volume method implemented in GeRa. Computational



a *b*

Figure 6. Geometric model in GeRa considering options A (a) and B (b)для вариантов А (а) и Б (б)

experiments have demonstrated (see Figure 7 as a case in point) that the calculations depend strongly on the meshing at the boundaries between bentonite/fracture (for option A) and "sand"/fracture (for option B). Ultimately, to find a compromise between the convergence of the result and the calculation rate, a fineness of $d = 3$ mm at the fracture boundary was chosen as the main computational grid (see Figure 8). In this case, flow underestimation of about 40 % is possible.

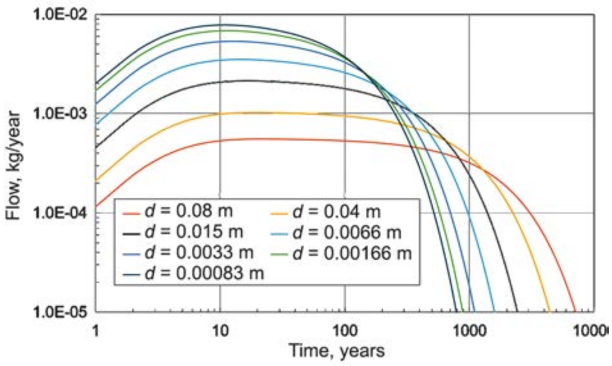


Figure 7. Intensity of contaminant release given different partitioning steps near the fracture boundary (option B without advection)

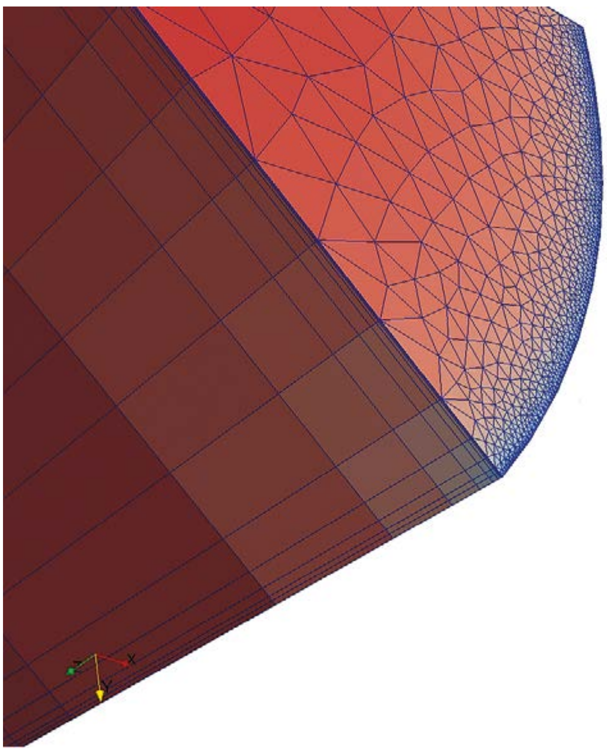


Figure 8. Partitioning the computational grid near the fracture boundary (partitioning fineness $d = 3$ mm)

Figure 9 shows the calculation results for a bentonite cylinder crossing a fracture (option A). As can be seen from the Figure, the obtained fluxes basically agree with the results of analytical calculations presented in Figure 2. The maximum discrepancy (approximately 2 times) is observed for a flow rate in the fracture of 1 m/year. The model has adequately reproduced the effect of "fracture resistance" (an increase in the outflow intensity along with an increased flow in the fracture). The discrepancy is most likely due to both the above-mentioned flow underestimation for the chosen partition discreteness and the fact that the analytical model was built based on simplified assumptions [3—5].

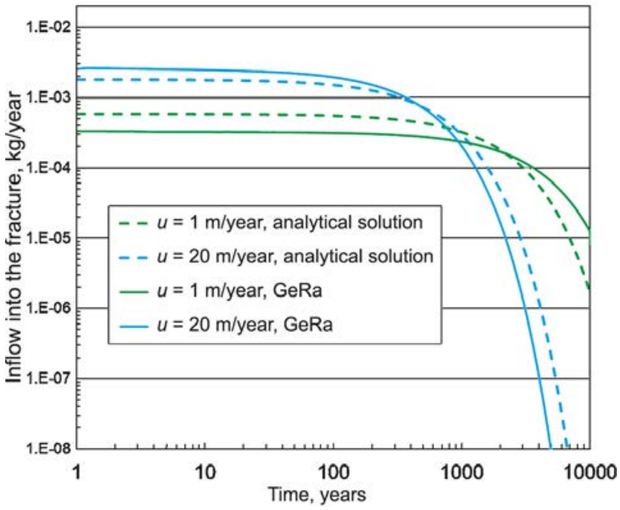


Figure 9. Results of calculations in GeRa in comparison with the analytical solution: contaminant flows into the fracture considering different GW flow rates for option A

Figure 10 presents the calculation results for option B. The flows obtained correspond to the data calculated in the chamber model B (Figure 5): the maximum flow for $u = 1$ m/year is $1.6 \cdot 10^{-3}$ kg/year

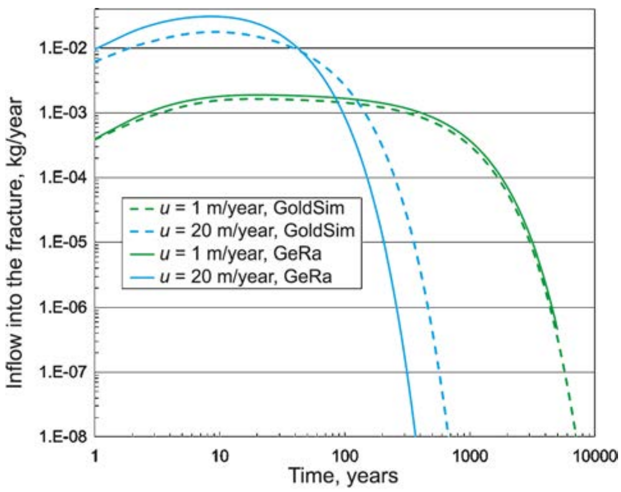


Figure 10. Comparison of calculation results for GeRa and GoldSim: impurity flows into the fracture given different GW rates for option B

(after 20 years); for $u = 20$ m/year — 0.0275 kg/year (in 9 years). At a flow rate of $u = 1$ m/year, the values practically coincide; at $u = 20$ m/year, they are higher by about 30 %

The effect of the above flow "focusing" in the backfill area adjacent to the fracture can be illustrated. Figure 11 shows the dependence of the calculated Darcy velocity in the backfill on the distance to the fracture plane along the "sand"/bentonite and "sand"/host rock boundaries (points were selected along the extreme transverse flow forming bentonite and "sand" cylinders). As it was assumed earlier, it can be seen that the main GW flow through the backfill is concentrated in a 20 cm thick layer.

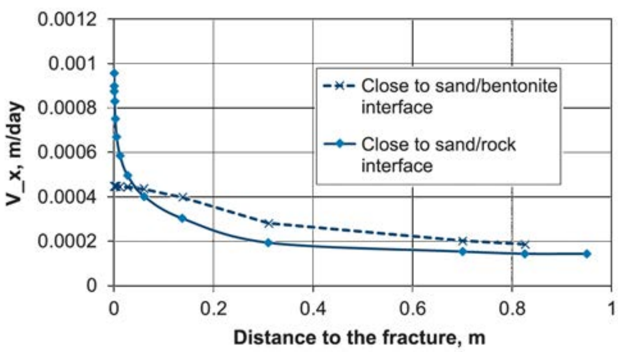


Figure 11. Dependence of the horizontal component constituting to the Darcy velocity in the backfill on the distance to the fracture plane (option B, u = 20 m/year)

Conclusions

1) Estimates based on approximate analytical and three-dimensional numerical models agree fairly well (see Figures 9, 10), so that their cross-verification may be considered successful. It should be noted that the results of the three-dimensional calculation (GeRa) appeared to be very sensitive to the meshing near the fracture boundary — an inaccurate choice of this parameter could result in a more than 10-fold underestimation of the desired value.

2) The three-dimensional numerical model (GeRa) has reproduced adequately the results of analytical calculations focused on contaminant release from bentonite (Figure 9). The maximum discrepancy (approximately 2 times) is observed for a flow rate in the fracture of 1 m/year. The model has adequately reproduced the "fracture resistance" effect and the discrepancy was caused both by the assumptions made under the analytical estimates [3—5] and by some underestimation of the flow at the chosen mesh fineness.

3) In most cases, even considering a completely degraded external backfill, ~ 90 % of the GW flow is concentrated in a relatively narrow layer (~ 10 % of its height), which was confirmed by calculations based on a three-dimensional numerical model.

4) It was shown that the degraded backfill around the block of compacted bentonite leads to an increase in the contaminant inflow to the fracture. The maximum groundwater flow rate $u = 20$ m/year is slightly more than an order of magnitude (about 11 times according to the simplified model and 12 times according to the 3D model). At the same time, at the target horizon, the GW flow rate in the fracture of 20 m/year is likely to be unacceptable according to the acceptance criteria of crystalline rocks for deep RW disposal [13], thus, barrier property deterioration under the concept [1] will be assumed to be several times less.

5) The accuracy of the calculated maximum contaminated flows (governing the radiation impact of the deep disposal facility) was expertly assessed within factor 2 (the difference between the given results and the "accurate" ones for this model is no more than two times).

6) The degree of other uncertainties currently available in the safety assessments performed for the deep disposal facilities to be developed in crystalline rocks is significantly higher than an order of magnitude, thus, at this stage, the concept from [1] may be considered as an alternative design solution for the deep disposal facility to be sited at the Yeniseiskiy site.

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