MODELLING OF COUPLED GROUNDWATER FLOW AND RADIONUCLIDE TRANSPORT IN CRYSTALLINE BASEMENT USING *FEFLOW 5.0*

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Abstract. The strategy of radioactive waste management of Lithuania provides for evaluating the possibilities of disposal of spent nuclear fuel and long-lived radioactive waste originated from Ignalina NPP in deep geological formations. The initial studies performed in Lithuania during 2001–2004 focused on screening of all potentially prospective geological formations as host formations. Since most information is available on crystalline basement, this formation was selected for the model case studies. Taking into account the assumptions (canister defect scenario proposed by Swedish experts and evaluated by LEI experts), groundwater flow and radionuclide (iodine-129 as mobile and long-lived one) transport modelling using computer code FEFLOW was performed according to geosphere conditions and parameters characteristic of the southern part of Lithuania (0,8×0,6×0,52 km far-field block). The upward groundwater flow through defected canister located in tectonically damaged zone was simulated. The main results of calculations are the following: in the case of upward groundwater flow, the maximum I-129 volumetric activity in single tectonic fracture above defected canister will not exceed 1 Bq/l, and in the active water exchange zone, it is close to 10^{-2} Bq/l. These figures show that doses obtained by human recipient via aquatic pathway should be below the dose constraint (0,2 mSv/y). More complicated scenarios and assumptions should be investigated in future studies.

Keywords: spent nuclear fuel, geological repository, crystalline basement, groundwater, numerical modelling, finite elements.

1. Introduction

During the operation of Ignalina NPP spent nuclear fuel (SNF) is produced. Since the beginning of Ignalina NPP operation in 1983, all SNF and operational radioactive waste have been stored at the NPP site. Since May 1999, SNF, after being stored in water ponds inside the NPP buildings, has been removed from there, loaded into CASTOR RBMK-1500 or CONSTOR RBMK-1500 casks and transported to an interim storage facility in the industrial area of the NPP. SNF can be stored in these casks for at least 50 years. Due to the danger of exposure arising from long-lived radionuclides to humans and the environment, SNF is prohibited for disposal in the near-surface repositories [1, 2].

Several alternatives related to safe management and disposing of SNF are being analyzed for the future. They are a possibility of having a deep geological repository for SNF and long-lived radioactive waste in any SNF-generating country, the possibilities of constructing a regional repository by joint efforts of several countries that generate or do not SNF, the possibilities of disposing of SNF in other countries, and the possibilities of extending the storage period in an interim storage for up to 100 and more years.

There is an international consensus that high-level and long-lived radioactive waste, first of all SNF, is best disposed of in deep geological repositories using a system of engineered and natural barriers [3]. It is a common opinion that there are few environments for which there exists evidence of stability over hundreds of thousands of years. Modern technology enables assessing old deep geological formations as the most obvious candidate environments.

No geological repository is in operation yet, however, repository projects in Finland, Sweden and USA have advanced to a stage at which decisions can be made to begin their construction [2]. Various types of geological formations are well investigated and can act as host rocks for a SNF repository. Since most information regarding deep geological formation in Lithuania is available on crystalline basement, this formation was selected for the model case studies [4–6].

On the basis of canister defect scenario evaluated by LEI experts, groundwater flow and radionuclide (iodine-129 as mobile and long-lived one) transport modelling, using computer code FEFLOW, was performed in this particular study for geosphere conditions prevailing in the southern part of Lithuania.

2. Outline of the problem of spent nuclear fuel

The expected total amount of SNF produced in Ignalina NPP until 2010 is 21941 fuel assemblies [5]. Each

fuel assembly contains approximately 111 kg of metal uranium, so the total mass of SNF that should be disposed of in the repository is about 2436 t. The radionuclide inventory in SNF will consist of actinides (uranium, plutonium isotopes, etc.), fission products (iodine, selenium, etc), and many activation radionuclides (a product of activation of metallic parts of fuel assembly). Most of radionuclides lie embedded in the fuel matrix of uranium dioxide for a very long time. A few fission products are relatively mobile. One of them is iodine-129 the inventory of which per canister in the model case study is assumed to be the same as in Swedish BWR fuel (2,31E+09 Bq, ^{129}I half life 1,57E+07 years, decay rate 1,3974E-11 1/s).

Repositories for disposal of SNF and high-level waste generally rely on a multi-barrier system to isolate waste from the biosphere. The multi-barrier system usually comprises a natural geological barrier and engineered barrier system [7]. The host rock types currently under investigation are salts (in either salt domes or bedded formations), granite and similar crystalline rocks, argillaceous rocks, tuff and basalt. The most investigated host rocks to date are crystalline rocks.

The Finnish government and local population first have recently approved a preferred site for SNF disposal in granite at Olkiluoto on the Baltic Coast. Sweden has focused onto two granite areas. Other countries that have considered granite-siting options include France, Switzerland, Spain and Canada.

The most developed Swedish concept for a deep geological repository involves encapsulating the fuel in cooper canisters with cast iron inserts, embedding each canister vertically or horizontally (two concepts, KBS-3V and KBS-3H) and surrounding them with bentonite clay at a depth of about 500 m in the Swedish crystalline basement. In recent KBS-3H alternative is proposed so-called super-container, which consists of a perforated steel cylinder with highly compacted bentonite and one cooper canister inside [8].

The KBS-3H concept was chosen as a prototype in modelling case of repository in crystalline basement in Lithuania [4].

3. Hydrogeological setting

Hydrogeological framework of the southern Lithuanian region that includes the study area was outlined on the basis of the information summarized in recent geoscientific generalizations [6, 9–11].

The whole territory of Lithuania belongs to the Baltic artesian basin (BAB) that occurs in the East European platform. Groundwater of BAB occurs in the sedimentary cover and in the fractured rocks of the crystalline basement. The groundwater-forming process depends on the structural geological features as well as climatic conditions, relief and river drainage of the region.

Quaternary deposits (glacial till, various sand and silt) occur at the ground surface and form the uppermost part of geological section of the region. The total thickness of these deposits varies from 83 to 143 m (an average thickness is 119 m). The smallest thickness is characteristic of the nor-

thern part of the region. Quaternary deposits of maximal thickness occur in the highlands of the region.

There are six aquifers (shallow groundwater layer and five confined layers) in the *Quaternary* cover. Shallow (unconfined) groundwater occurs at 1 m depth in the lowlands and at more than 10 m depth in sandy elevations. Confined Quaternary aquifers occur in inter-glacial sands and gravels. Piezometric level and hydraulic properties of Quaternary aquifers are different and depend on the complexity of the geological structure. The groundwater of Quaternary aquifers is fresh (TDS <1 g/l), its total hardness varies from 4 to 7 mg-ekv/l, and pH is about 7,5. Ions of hydrocarbonate and calcium prevail in the chemical composition of water. For modelling purpose, all Quaternary aquifers are considered as a unified aquifer system [6].

The Cretaceous aquifer consists of chalk, silt, and sand. The top of the aquifer dips from an altitude of 61 m a.s.l. to −10 m b.s.l. The thickness of the aquifer varies from 30 to 105 m. The hydraulic head of the aquifer varies between the altitudes of 150 and 155 m a.s.l. The specific yield varies from 0,1 to 2 l/s, and permeability – from 1 to 260 m^2/d. Fresh groundwater prevails in the aquifer except several anomalies in the valley of the Nemunas river.

The Permian aquifer occurs mostly in the northern and northwestern parts of the region. It consists of fractured limestone and sandstone. The top of the aquifer occurs from a depth of 141 m to a depth of 244 m. It dips to the west and southwest. The thickness of the aquifer reaches 20–25 m. The hydraulic head occurs between the altitudes of 160 and 210 m. The specific yield of the aquifer varies from 0,2 to 2,4 l/s. Permeability varies between 2 and 366 m^2/d. The salinity of groundwater reaches 50 g/l. Sodium and chloride ions prevail in the hydrochemical composition of the water.

The Silurian–Upper and Middle Ordovician aquifer system occurs over the studied region except the southernmost edge. It consists of limestone and clay. The top of the aquifer occurs at a depth of 204 to 277 m. The hydraulic head occurs at an altitude 150 m a.s.l. The lowest level is characteristic of the northern part and rises to the southeast. The specific yield of the aquifers varies from 0,002 to 0,296 l/s, and permeability is less than 44 m^2/d. The salinity value of groundwater is low only near the boundaries of aquifer and rises up to 50 g/l in the thicker part.

The Lower Ordovician – Cambrian aquifer system consists of limestone, clayey limestone and sandstone. The altitude of the aquifer top is varying from 150 m to 210 m b.s.l. It dips to the west. The hydraulic head varies between the altitudes of 80 and 150 m a.s.l. The specific yield of the aquifer varies from 0,035 to 0,25 l/s, and permeability is less than 40 m^2/d. The salinity of the groundwater could be from 0,6 g/l near the boundary of occurrence to more than 50 g/l in the north.

The Lower Cambrian aquitard occurs in the northern and northeastern part of the region. It consists of argillite, sandstone and clay. The depth of the layer varies from 259 to 287 m. The altitudes of occurrence are

between 140 m (in the south) and 210 m (in the north-western part of the region).

The Cambrian – Vendian aquifer system consists of sandstone and argillite. It occurs in the whole area of the region at a depth between 300 and 450 m. The aquifer is dipping to the east. Its thickness reaches 100 m in the eastern part of the studied region. The specific yield of the aquifer varies between 0,006 and 0,25 l/s, and permeability could be from 1 to 35 m^2/d. The aquifer has a strong hydraulic connection with the underlying Protherozoic–Archaean aquifer. Therefore, the hydraulic heads of these two systems are similar. It is about 20 m a.s.l. in the southwestern part and more than 110 m a.s.l. in the northeast. The salinity of the groundwater is low (0,3–0,5 g/l) where aquifer occurs at a smaller depth and can reach 100 g/l deeper.

Sediments that were deposited at different stages of the basin evolution overlie the basement rocks. The crystalline rocks were exposed to denudation and weathering for different time intervals under various climatic conditions. Therefore, the upper part of the basement is weathered. The composition of the weathered crust varies significantly, that is related to the composition of the basement rocks and physical-chemical conditions.

The Proterozoic–Archaean aquifer occurs over the region. It consists of fractured crystalline rocks. Groundwater is accumulated in weathering and tectonic fractures. The aquifer occurs at a depth from 300 m to more than 600 m. The permeability of the aquifer could reach 50 m^2/d, and the specific yield could be between 0,25 and 0,35 l/s. The salinity of groundwater is lower than 1 g/l in the southeast and could reach 100 g/l in deep

occurrence areas. Sodium and chloride ions prevail in the hydrochemical composition of groundwater [6].

A typical hydrogeological 2 km long section was chosen from the South Lithuanian region for assessment through modelling (Fig 1).

The site-specific for this section and generic parameters are given in Tables 1 and 2.

4. Conceptual model

According to the Swedish KBS-3H concept, variant MLH (Medium Long Hole) where canisters are proposed to be deposited in a 150–500 m long deposition hole on both sides of the transport tunnel, the evolution of one canister was considered [8]. A copper canister selected as a prototype for SNF from Ignalina NPP is composed of two components: an outer corrosion protective shell of copper and a cast iron insert with channels for the fuel half-assemblies in order to improve the mechanical strength. One canister can hold 32 RBMK-1500 fuel half-assemblies, and for Lithuanian disposal purposes, about 1400 canisters should be employed. A copper canister with 50 mm wall thickness should be made of oxygen-free copper with a low phosphorus content. A canister insert is cast of steel and will have a minimum wall thickness of 50 mm. Preliminary data for a reference canister are 1050 mm in diameter and 4070 mm in length.

The chemical evolution of a damaged canister differs radically from that of an intact canister. Intruding water to a defected canister gives rise to several important chemical reactions, namely: corrosion of a cast-iron insert, corrosion of fuel claddings and other metallic parts, release of radionuclides in these parts, dissolution of the fuel matrix with release of radionuclides.
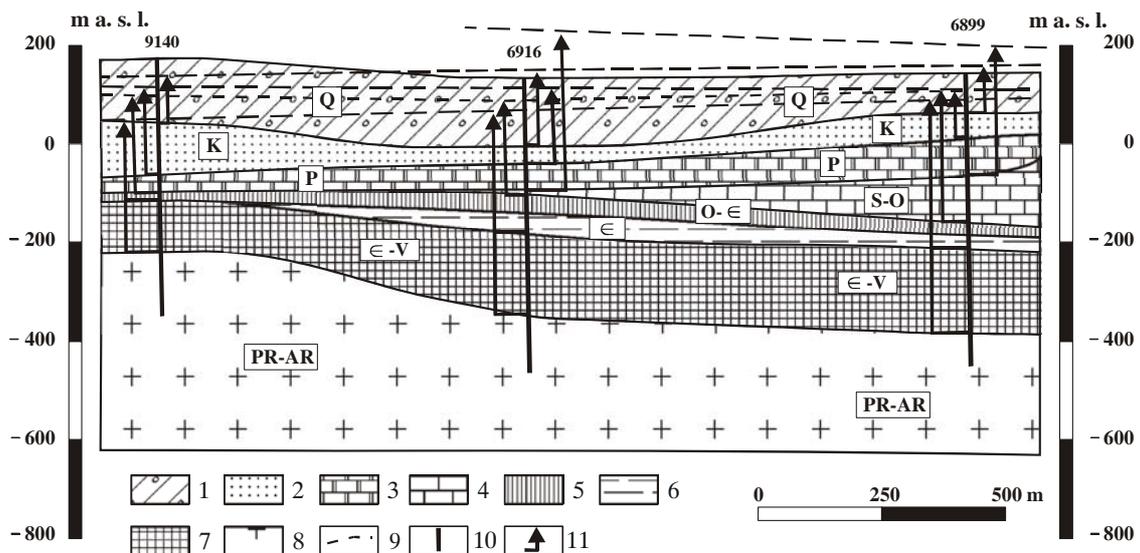
Fig 1. Hydrogeological cross-section: 1 – Quaternary deposits (glacial till, silt and various sands), 2 – Cretaceous (sandy deposits, silt and chalk), 3 – Permian (limestone and sandstone), 4 – Silurian-Upper-Middle Ordovician (limestone and clay), 5 – Lower Ordovician-Cambrian (limestone, clayey limestone and sandstone), 6 – Lower Cambrian (argillite, sandstone and clay), 7 – Cambrian-Vendian (sandstone and argillite), 8 – Proterozoic-Archaen (crystalline rocks), 9 – groundwater level, 10 – well, 11 – hydraulic head

Table 1. Averaged hydrogeological parameters selected for simple modelling domain in the southern part of Lithuania [6, 9–12]

No	Geological formation	Lithological description (prevailing)	Thickness, m	Hydraulic conductivity, 10^{-4} m/s	Porosity	Density, kg/m^3	Henry sorption for ^{129}I	Longitudinal dispersivity, m	Transverse dispersivity, m
1	Quaternary	Glacial loam	100	0,12	0,25	2000	2	0,01	0,001
2	Cretaceous	Sand	100	0,23	0,35	2300	2,3	5	0,5
3	Permian	Anhydrite	50	0,58	0,5	2980	2,98	0	0
4	Silurian-Ordovician	Limestone	20	0,01	0,5	2600	2,6	5	0,5
5	Ordovician-Cambrian	Sandstone	20	0,58	0,05	2700	2,7	5	0,5
6	Cambrian	Clay	20	$1,16 \cdot 10^{-6}$	0,13	2600	2,6	0,01	0,001
7	Cambrian-Vendian	Sandstone	100	0,06	0,05	1800	1,8	5	0,5
8	Proterozoic-Archaean	Weathered rocks	10	$1,16 \cdot 10^{-3}$	0,01	2000	2	0,01	0,001
9		Crystalline rocks	100	$1,16 \cdot 10^{-9}$	0,0038	2800	2,8	5	0,5

Table 2. Parameters of single fracture representing tectonic fault zone [6]

Parameters	Crystalline rocks (PR-AR)	Sandstone (CM-V)
Thickness, m	1	1
Hydraulic conductivity, 10^{-4} m/s	60	5
Porosity	0,3	0,26
Longitudinal dispersivity, m	0,01	5
Transverse dispersivity, m	0,001	0,5
Henry sorption for ^{129}I	2	1,8
Rock density, kg/m^3	2000	1800

Following the results of near-field modelling [5], a simplified canister evolution is given for a canister defect scenario:

- One canister out of the total of 1400 will pass through quality inspection with a penetrating defect of 1 mm^2 in size.
- 200000 years were chosen as a reasonable value for the time when the initial defect grows to a large one.
- After the defect becomes larger, the entire void in the canister at closure, approximately 0,5 m^3, will be filled with water.
- Radionuclides released during the SNF dissolution process dissolve in water within the canister.
- If the concentration of radionuclide exceeds its solubility limit, they start to precipitate. Sorption on the internal parts of the canister is neglected.
- Radionuclides diffuse out through a defect hole.
- The buffer is fully saturated at the time when the release of radionuclides starts. Thus, radionuclides are transported through the bentonite buffer by diffusion mainly.

Radionuclides that reach the boundary of the bentonite buffer diffuse into the water flowing in the fracture intersecting the emplacement tunnel. Thus, radionuclides

are transported to far-field and distributed in the hydrogeological system.

From several cases considered in detail in SR97 and referenced in [5], the most conservative case was selected in this study assuming that SNF is completely dissolved when a continuous water pathway is created (instant release fraction of 100 %).

A very simple conceptual model of radionuclide transport in the repository system, including far-field aquifers, is presented in Fig 2.
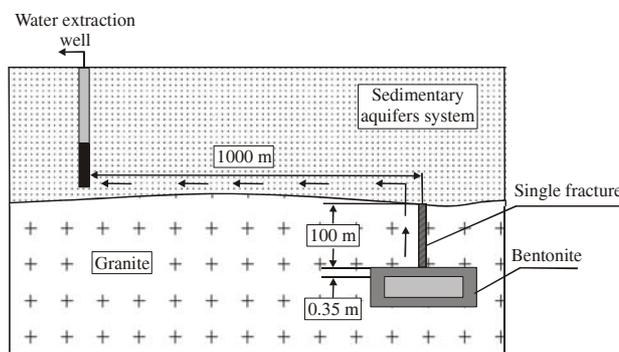
Fig 2. Conceptual model of radionuclide transport (not to scale) [5]

The simple conceptual model is adapted to the hydrogeological conditions of the study area demonstrated in the hydrogeological section (Fig 1).

It is conservatively taken that the canister is in the tectonic fault zone of crystalline basement. The groundwater washing the canister discharges into the upper aquifers. For enhancement of hydrodynamic effect it is taken that a productive well (yield value is 3000 m^3/day) is installed 363 m from the canister in the Ordovician-Cambrian aquifer (sand).

The current understanding of solute migration processes in aquifers involves the following major important radionuclide migration mechanisms: advection in unsatu-

rated and saturated zones, molecular diffusion, hydrodynamic dispersion and retardation [13, 14].

The migration of radionuclides from a repository toward the geosphere (aquifers) and eventually toward to the surface (or biosphere) can only start when the canister containing SNF has been defected and SNF has been dissolved by water and radionuclides have been transported away from the container through the engineered barriers surrounding them. In this study, the time for the mentioned processes is assumed to be 200 000 years.

An additional component of the transport of radionuclides is their retardation by interactions with the solid phase. There are many types of possible interactions but the most common one is adsorption, particularly in the case of small particles (silt, clay, etc). Additional mechanisms may include chemical reactions (acid-base, redox, precipitation, etc) as well as reactions of a solute with organic molecules or with mineral colloids naturally present in groundwater. If radionuclides react with or absorb on such molecules or particles, they may not interact with the solid (no sorption) and be transported faster than expected. The issues of "facilitated" transport are particularly important in solutes with high colloid concentrations [15–17].

An estimated I-129 release rate through engineered barriers to far-field was taken from the study [5] as input function for 3D modelling of radionuclide transport in far-field (Fig 3).

5. Computer code and numerical model

The continuum approach is a standard to describe the fundamental processes of flow and transport in porous media [18]. The assumption of continuum implies that the main physical properties (velocity, pressure, concentration, etc) are continuously distributed in space and thus exist for any infinitely small material point.
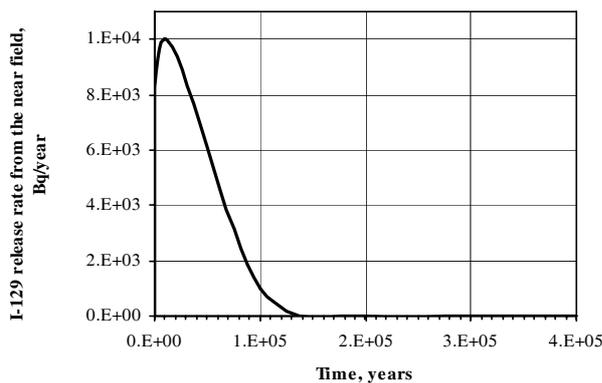
Fig 3. I-129 release rate from near-field starting after 200 000 years after repository closure [5]

In most porous-media problems mass, motion and energy-related quantities can neither be measured nor solved at a microscopic level due to the geometric complexity of real geologic media. These difficulties are resolved by a transformation to the macroscopic level by averaging over volumes, leading to measurable and solv-

able quantities for which the continuum assumption is then invoked. The related macroscopic equations are solved by numerical methods [19, 20]. Popular and powerful strategies are the finite element and finite volume method. Both methods have the same mathematical basis with weighted-integral formulation of the governing balance equations [18].

The finite element modelling has broad applications in various disciplines, including petroleum engineering (hydrocarbon-gas injection and recycling in gas-condensate reservoirs), contaminant hydrogeology and geochemical engineering (contamination of groundwater aquifers and radioactive waste management in the subsurface).

Evaluation of radionuclide environmental effects on a long-term scale requires quantifying properly the hydrogeology of the area under consideration and then mass transport parameters. Constructing of a numerical model for groundwater flow permitted an integration of all the available pieces of site-specific information. However, mass transport parameters are usually absent for particular areas. In order to avoid impermissible uncertainties of simulation results a long-lived and very mobile radionuclide for illustrating the problem of radionuclide transport should be chosen.

The numerical model used in this study is based on three-dimensional finite-element code FEFLOW 5.0 which allows modelling groundwater flow and contaminant transport in layered three-dimensional system taking into account fracture phenomenon. The simulation system based on FEFLOW code models the flow and contaminant mass processes as coupled or separate phenomena [21]. It is based on physical conservation principles for mass, chemical species, linear momentum and energy in a transient and three-dimensional (if necessary, two-dimensional) numerical analysis.

For three-dimensional (3D) and two-dimensional (2D) − vertical and axisymmetric, respectively − processes the representative model general equations are as follows ($i, j = 1, 2, 3$):

$$S_0 \frac{\partial h}{\partial t} + \frac{\partial q_i^f}{\partial x_j} = Q_\rho + Q_{EB}(C,T), \qquad (1)$$

$$q_i^f = -K_{ij} f_\mu \left(\frac{\partial h}{\partial x_j} + \frac{\rho^f - \rho_0^f}{\rho_0^f} e_j \right), \qquad (2)$$

$$\varepsilon R_d \frac{\partial C}{\partial t} + q_i^f \frac{\partial C}{\partial x_i} - \frac{\partial}{\partial x_i} \left(D_{ij} \frac{\partial C}{\partial x_j} \right) + \left(\varepsilon R \vartheta + Q_p \right) C = Q_C, \qquad (3)$$

with constitutive equations

$$\rho^f = \rho_0^f \left[1 + \frac{\bar{a}}{(C_s - C_0)}(C - C_0) \right],$$

$$h = \frac{p^f}{\rho_0^f g} + x_i, \quad K_{ij} = \frac{k_{ij} \rho_0^f g}{\mu_0^f},$$

$$\bar{a} = \left[\rho^f (C_s) - \rho_0^f \right] / \rho_0^f,$$

$$f_\mu = \frac{\mu_0^f}{\mu^f(C,T)} =$$

$$\frac{1+1,85\omega_{(C=C_0)}-4,1\omega_{(C=C_0)}^2+44,5\omega_{(C=C_0)}^3}{1+1,85\omega-4,1\omega^2+44,5\omega^3}.$$

$$\frac{1+0,7063\varsigma-0,04832\varsigma^3}{1+0,7063\varsigma_{(T=T_0)}-0,04832\varsigma_{(T=T_0)}^3},$$

$$\varsigma = \frac{(T-150)}{100}, \ \omega = \frac{C}{\rho^f},$$

$$D_{ij} = (\varepsilon D_d + \beta_T V_q^f)\delta_{ij} + (\beta_L - \beta_T)\frac{q_i^f q_j^f}{V_q^f},$$

$$R = 1 + \frac{(1-\varepsilon)}{\varepsilon}\chi(C), \ R_d = 1 + \frac{(1-\varepsilon)}{\varepsilon}\frac{[\chi(C)\cdot C]}{\partial C}, \quad (4)$$

which are to be solved for three primary variables (h – hydraulic head; q_i^f – Darcy velocity vector of fluid; C – concentration of chemical component or radionuclide).

The main governing parameters are: ρ^f, ρ_0^f – fluid and reference fluid density, respectively; ρ^s – solid density; S_0 – specific storage coefficient (compressibility); K_{ij} – tensor of hydraulic conductivity; e_j – gravitational unit vector; f_μ – constitutive viscosity relation function; Q_{EB} – term of extended Boussinesq approximation; R – specific retardation factor; R_d – derivative term of retardation; D_{ij} – tensor of hydrodynamic dispersion; δ_{ij} – Kronecker tensor; υ – decay rate (λ is most conventional designation for radioactive decay rate); ε – porosity; Q_x – source/sink function of fluid ($x=\rho$) and contaminant mass ($x=C$); $\overline{a}$ – fluid density difference ratio; $\overline{\beta}$ – fluid expansion coefficient; C_0, T_0 – reference values for concentration and temperature, respectively; C_s – maximum concentration; p^f – fluid pressure; g – gravitational acceleration; k_{ij} – tensor of permeability; μ^f, μ_0^f – dynamic viscosity and its reference value,

respectively, of fluid; ζ – normalized temperature; ω – mass fraction; D_d – molecular diffusion coefficient of fluid; V_q^f – $\sqrt{q_i^f q_i^f}$ – absolute Darcy fluid flux; β_L, β_T – longitudinal and transverse dispersiveness, respectively, of chemical species; $\chi(C)$ – concentration-dependent adsorption function.

The relationships of retardation R and R_d, respectively, can be summarized for different adsorption laws as follows [21]:

for Henry isotherm:

$$R = \varepsilon(1-\varepsilon)K, R_d = \varepsilon + (1-\varepsilon)K, \quad (5)$$

for Freundlich isotherm:

$$R = \varepsilon + (1-\varepsilon)b_1 C^{b_2-1}, R_d = \varepsilon + (1-\varepsilon)b_1 b_2 C^{b_2-1}, \quad (6)$$

for Langmuir isotherm:

$$R = \varepsilon + (1-\varepsilon)\frac{k_1}{1+k_2 C}, R_d = \varepsilon + (1-\varepsilon)\frac{k_1}{(1+k_2 C)^2}, \quad (7)$$

where K – Henry sorption coefficient; b_1, b_2 – Freundlich sorption coefficient and exponent, respectively; k_1, k_2 – Langmuir sorption coefficients.

The Henry sorption coefficient of the radionuclides is calculated as:

$$K = k_d \rho^s, \quad (8)$$

where k_d – distribution coefficient for particular radionuclide, m³/kg; ρ^s – density of solid, kg/m³.

From general equations, the horizontal model equations, which differ between confined and unconfined conditions and convective and divergence forms of contaminant transport equations, are derived. The governing model equations have to be supplemented by initial, boundary, and constraint conditions. There are many formulations of initial, boundary, and constraint conditions realized in FEFLOW. For the tensors of conductivity and transmissivity, anisotropy is to be taken into account in FEFLOW as well [21].

A comparatively simple and conservative (in terms of groundwater flow formulation) numerical model was worked out on the basis of a conceptual model (Table 3).

Table 3. Features of numerical model for the domain of southern Lithuania

Problem class	Combined flow and mass transport model	Vertical exaggeration	1:1
Time class	Transient (unsteady) flow, transient (unsteady) mass transport	Problem measure	877,67 m
Time stepping scheme	Fully implicit with constant time steps	Number of layers	11
Upwinding	Full up winding	Number of slices	12
Number of time steps	50	Type	Saturated
Length of time step	$7.305\cdot10^6$ d	Dimension	Three-Dimensional
Error tolerance	$1\cdot10^{-3}$ applied to Euclidian L_2 integral (RMS) norm	Element type	6-noded triangular prism
Maximum number of iterations per time step	12	Mesh elements	56232
Adaptive mesh error	$1\cdot10^{-2}$	Mesh nodes	31884
A posteriori error estimator	Onate-Bugeda algorithm	Mesh optimization	Not done
Velocity approximation	Improved consistent velocity approximation (by Frolkovic-Knabner algorithm) applied to linear elements with local smoothing	Aquifers	Unconfined (phreatic); other aquifers are unspecified (automatically adapted)
Form of transport equation	Convective	Projection	None (3D with free surface)
Solver	Iterative equation solver	Adsorption law	Henry isotherm

Once calibrated, the model was used to predict radionuclide transport within groundwater and a potential effect to the environment. The model is virtually rather conservative and constructed in such a way as to imitate a greater migration of radionuclides to aquifers than it could be possible under the chosen geological conditions. The defected canister is installed in the top part of the crystalline basement, though, following the conceptual design, it should be at a depth of 100 m.

6. Discretization, parameterization, initial, boundary and constraint conditions

Flow in all the aquifers of the numerical model is attributed to 11 layers and represented by 3D finite-element grid of 56232 mesh elements as shown in Fig 4.

The vertical discretization of the model and its parameter values are based on the data presented in Fig 1 and Tables 1 and 2. Vertical discretization of the model in terms of boundary conditions is shown in Tables 4 and 5.

For a numerical model, the created layers are adjusted to the geometry of lithological bodies. In the first simple model approach parameters are assumed to be constant within the whole modelling domain, e g each layer consists of one zone. For a brief illustration of groundwater flow modelling features, a modelled hydraulic head distribution in modelling layer 9 is given which corresponds to crystalline rocks (Fig 5).

By collecting additional hydrogeological information and during further model calibration, the number and outline of zones should be increased to minimize the difference between computed and measured or expected heads at observation points.

The default initial conditions of the hydraulic head and contaminant mass (0 Bq/l) are used for this model.
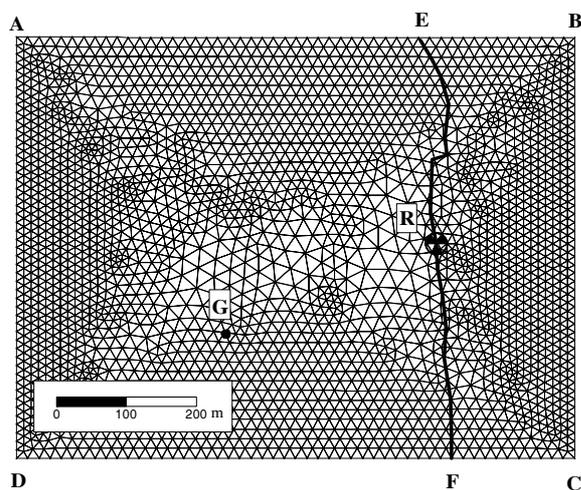
Fig 4. Finite-element grid used for the model domain of southern Lithuania (see Table 4)

7. Model calibration and sensitivity analyses

Model calibration by many repeated runs is routine in numerical simulation [22, 23].

Model calibration as practiced in this study is a process by which the structure and parameters of a nu-

merical model are progressively changed in an iterative manner to obtain an acceptable match between observed or expected and computed hydraulic heads and departing as little as possible from the original conceptual model. In addition to fitting hydraulic heads, calibration was based on the following criteria: computed water levels should always stand below the topographic surface; estimated hydraulic conductivities should be consistent with conductivities derived from available hydrogeological data; spatial patterns of parameters should be consistent with available geological information. A direct experimental substantiation of mass transport is impossible because tracer experiments with long-living mobile radionuclides (I-129) are hardly validated, and the process of large-scale radionuclide transport is very long-lasting.

Table 4. Flow boundary conditions for the model domain of southern Lithuania after model calibration (see Fig 4)

Section or point (layer)	Type	Value	Comment
A–B (1–11)	–	–	Unspecified (impervious) for all 11 layers
B–C (1–11)	Dirichlet (1st kind)	h_1=155 m; h_2=154 m; h_3=153 m; h_4=152 m; h_5=151 m; h_6=150 m; h_7=149 m; h_8=148 m; h_9=147 m; h_{10}=147 m; h_{11}=146 m	Pervious boundary (influx)
C–D (1–11)	–		Unspecified (impervious) for all 11 layers
D–A (1–11)	Dirichlet (1st kind)	h_1=130 m; h_2=131 m; h_3=132 m; h_4=133 m; h_5=134 m; h_6=135 m; h_7=136 m; h_8=137 m; h_9=138 m; h_{10}=138 m; h_{11}=139 m	Pervious boundary (outflow)
E–F (7–11)	Dirichlet (1st kind)	h_{7-11}=147 m	Pervious boundary (influx into fracture representing tectonic fault zone)
G (5)	(4th kind)	Q_ρ^w=3000 m^3/d	Pumping rate of single well

Table 5. Boundary conditions for contaminant mass transport for the convective form

Point (layer)	Type	Value		Comment
		years	Bq/l	
R (10)	Dirichlet (1st kind); time-varying function $C(x_i,t) = C_1^R(t)$	<10000	0	Release from repository (defected canister) to receiving cell with dimensions of 12×12× 1,85 m
		10000–100000	3081	
		100000–200000	308	
		200000–300000	3,1	
		300000–400000	1,5	
		>400000	0	
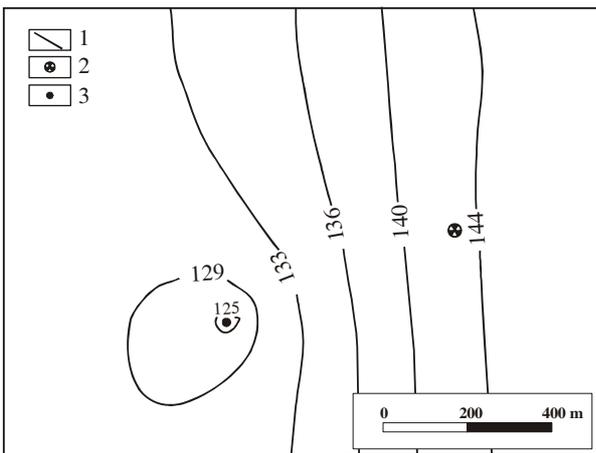
Fig 5. Simulated hydraulic head of layer 9 representing crystalline rocks after 40 000 years: 1 – isoline of hydraulic head, m a.s.l., 2 – repository, 3 – pumping well

The parameters of relevance under far-field flow conditions routinely include hydraulic conductivity (K), areal recharge, hydraulic heads (h) specified along boundaries, dispersivity parameters (β_L, β_T). Areal recharge was assumed to be uniform throughout the study area (1,4·10^{-4} m/d).

A sensitivity analysis was performed to (a) quantify uncertainty in the estimated parameters and that of the parameters that remained fixed during calibration, and (b) evaluate the effect of some hypotheses of the conceptual model (for example, influence of a pumping well). The sensitivity of the model with respect to a given parameter was evaluated by performing calibration runs in which this parameter was assigned a fixed value within a plausible interval, while the rest of the model parameters were estimated. Sensitivity analyses were carried out using the final structure of the model, and its sensitivity was tested for (a) hydraulic conductivities of all the layers and for (b) dispersivity of all the layers (Figs 6 and 7).

For sensitivity analysis, runs of the model were made for four sets of hydraulic conductivity: 1 – normal hydraulic conductivity corresponding to the values of Table 1; 2 – normal conductivity increased by one order of magnitude; 3 – normal conductivity increased by two orders of magnitude; 4 – normal conductivity decreased by one order of magnitude. The change of volumetric
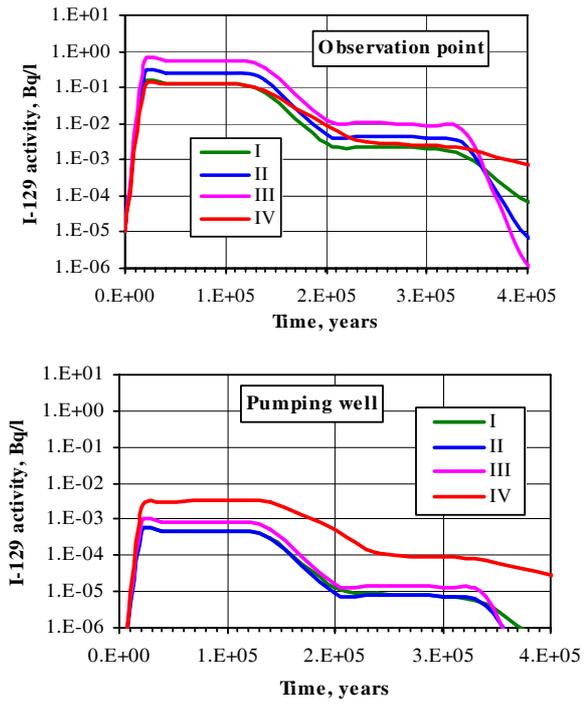
Fig 6. Computed temporal distributions of ^{129}I volumetric activity in groundwater for different sets of hydraulic conductivity values (I – normal hydraulic conductivity; II – normal conductivity increased by one order of magnitude; III – normal conductivity increased by two orders of magnitude; IV – normal conductivity decreased by one order of magnitude)
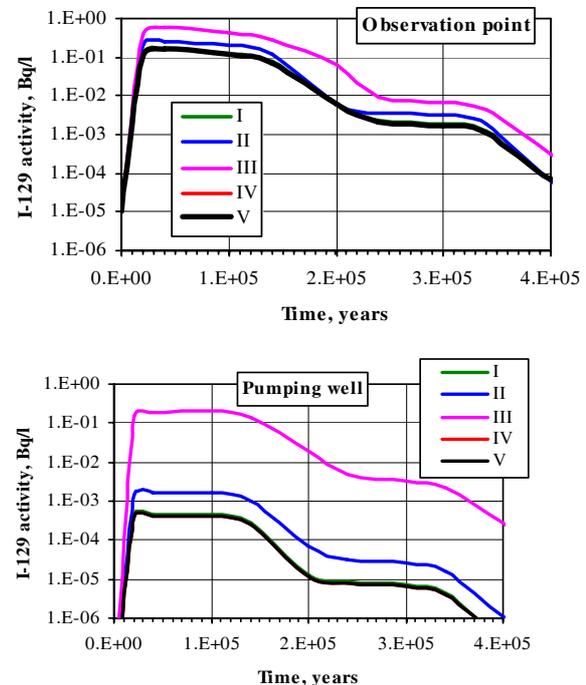
Fig 7. Computed temporal distributions of ^{129}I volumetric activity in groundwater for different sets of hydraulic dispersivity values (I – normal dispersivity; II – normal dispersivity increased by one order of magnitude; III –normal dispersivity increased by two orders of magnitude; IV – normal dispersivity decreased by one order of magnitude; V – zero dispersivity)

activity of I-129 in groundwater for a period of 0 to 10^6 years was reconstructed at an observation point (layer 9 – crystalline rocks, about 90 m from repository) and in a pumping well (layer 5 – sandstone, 363 m from a repository).

The volumetric activity of I-129 in the groundwater at an observation point close to the defected canister did not differ in more than one order of magnitude throughout the whole modelled period. The highest I-129 volumetric activity occurred when the normal conductivity increased by two orders of magnitude. Peak activity value in all the cases did not exceed 1 Bq/l. The volumetric activity of I-129 in the groundwater of a pumping well more remote to the defected canister did not differ in more than one order of magnitude throughout the whole modelled period either. The highest I-129 volumetric activity occurred when the normal conductivity decreased by one order of magnitude. Peak activity value in all the cases did not exceed 10^{-2} Bq/l.

For sensitivity analysis, runs of the model were made for five sets of longitudinal and transverse dispersivity parameters: 1 – normal dispersivity corresponding to the values of Table 1; 2 – normal dispersivity increased by one order of magnitude; 3 – normal dispersivity increased by two orders of magnitude; 4 – normal dispersivity decreased by one order of magnitude; 5 – zero dispersivity. The change of volumetric activity of I-129 in groundwater during the period from 0 to 10^6 years was reconstructed at the same observation point (layer 9 – crystalline rocks, about 90 m from a repository) and in a pumping well (layer 5 – sandstone, 363 m from a repository). The volumetric activity of I-129 in the groundwater at an observation point close to the defected canister in all the cases did not differ in more than one order of magnitude throughout the whole modelled period. Actually calculation results are the same in most of the cases except higher values when the normal dispersivity increased by two orders of magnitude. The volumetric activity of I-129 in the groundwater of a pumping well more remote to the defected canister did not differ in more than one

order of magnitude throughout the whole modelled period either except the higher values when the normal dispersivity increased by two orders of magnitude.

The calibrated model especially in the pumping well area was very sensitive to changes in dispersivity parameters, namely, in the region of very high dispersivity values. The calibrated model in closer to a release point zone was less sensitive to changes in dispersivity parameters. On the other hand, model results for the two cases were not very sensitive to small or even large changes in the hydraulic conductivities. Therefore, the calibrated model was not very sensitive to errors in the hydraulic conductivity estimations.

8. Spatial distribution of contamination plume

Computer program FEFLOW enables spatial and temporal analysis of modelling results in many ways. For demonstration, we present here the calculation results for contamination plume at a moment when peak volumetric activity of I-129 is expected. This is the time approximately 40000 years after the initial moment of release. Yet the initial moment of release will occur 200 000 years after the repository closure. Thus, the migration of the most mobile radionuclide ^{129}I in groundwater will be possible only in a very remote future for normal evolution scenario.

Calculation results in the section show that a contamination plume of 10^{-2} Bq/l does not reach furher than 700 m from the defected canister and includes the Cambrian-Vendian and weathered Proterozoic-Archaean aquifers (Fig 8).

Imitation of discharge conditions show that the groundwater containing I-129 with volumetric activity 10^{-5}–10^{-2} Bq/l is able to reach a productive well or earth surface.

The areal distribution pattern of volumetric activity of I-129 in the most highly polluted water of Proterozoic-Archaean aquifer is given in Fig 9. The highest predicted value of activity near the defected canister in the zone of
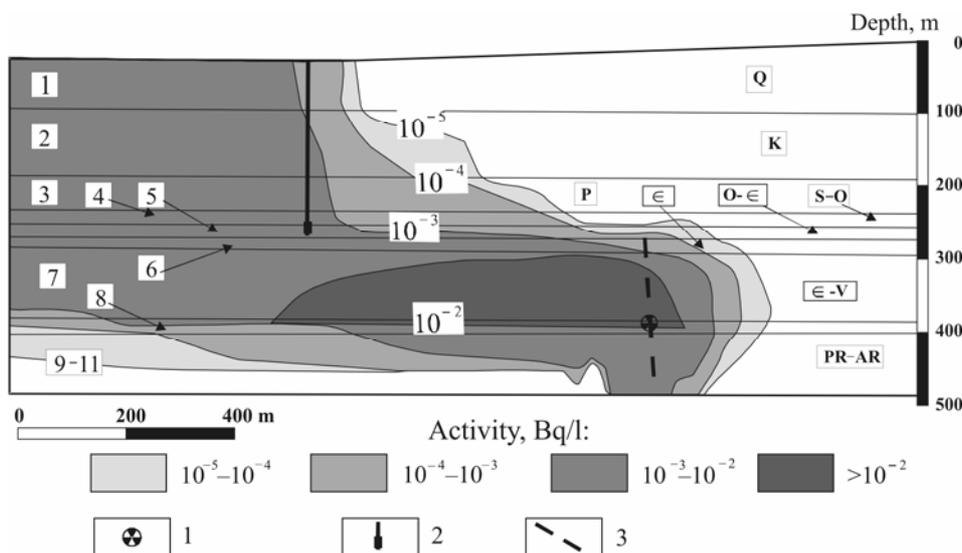
Fig 8. Contamination plume of ^{129}I (cross-section): 1 – repository, 2 – pumping well, 3 – tectonic fault
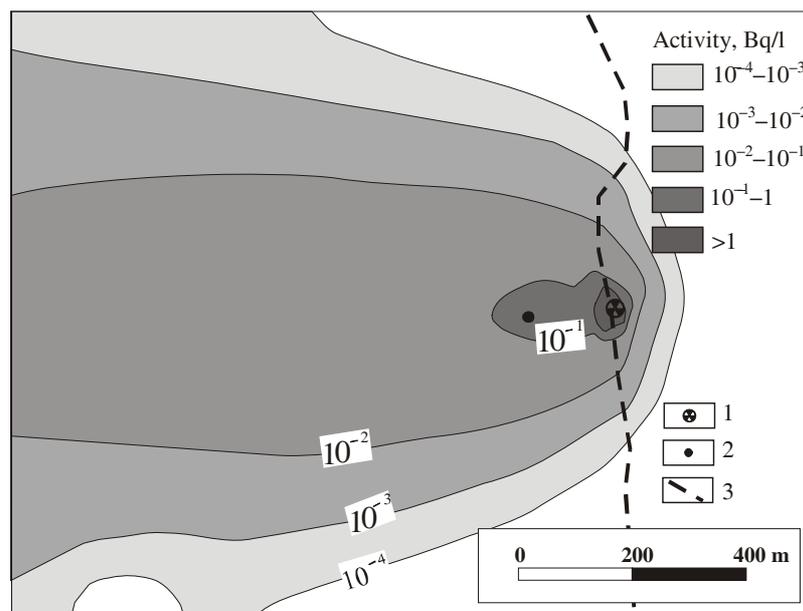
Fig 9. Contamination plume of ^{129}I (areal distribution in Preoterozoic-Archaean aquifer):
1 – repository, 2 – observation point, 3 – tectonic fault

tectonic fault is of the order 1 Bq/l. Contamination plume of 10^{-1} Bq/l reaches the observation point, whereas the groundwater containing I-129 with possible volumetric activity of 10^{-2} Bq/l discharges outside the model boundaries in the flow direction.

The highest permissible value of I-129 volumetric activity corresponding to the 0,2 mSv/years effective dose for drinking water pathway is 5,5 Bq/l. Yet today, for example, the global level of I-129 volumetric activity in the river water is about 10^{-6} Bq/l.

The numerical model should be improved in future and more scenarios of a long-term evolution of repository-surrounded media should be worked out.

9. Conclusions

The crystalline basement is the best worldwide investigated media in connection with disposal of SNF. In Lithuania, thick sedimentary layers of varying origin and properties cover the crystalline basement. Complex structures of overlying layers make performance assessment significantly more sophisticated and complicated.

Modelling of safety of the most relevant radionuclide ^{129}I migration for tectonically fractured domain shows that doses to humans even in such a conservative case will not exceed the existing dose restrictions. Predicted volumetric activity of I-129 at 363 m distance from repository in groundwater of the Ordovician-Cambrian aquifer, which could be used for water supply, is of the order 10^{-3} Bq/l.

On the basis of worldwide experience it would in principle be possible to dispose of SNF and other long-lived high-level radioactive wastes in a repository built in the crystalline basement.

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POŽEMINIO VANDENS TĖKMIŲ IR RADIONUKLIDŲ PERNAŠOS KRISTALINIAME PAMATE MODELIAVIMAS *FEFLOW* KODU

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S a n t r a u k a

Pagal radioaktyviųjų atliekų tvarkymo strategiją Lietuvoje yra numatoma galimybė panaudotą branduolinį kurą ir ilgaamžes radioaktyviąsias atliekas, susidarančias Ignalinos AE, laidoti giliose geologinėse formacijose. Pradiniai tyrimai 2001–2004 metais buvo skirti atrinkti šiam tikslui tinkamiausias geologines formacijas. Modeliui sudaryti pasirinktas labiausiai ištirtas kristalinis pamatas. Požeminio vandens srauto ir radionuklido (itin mobilaus ir ilgaamžio jodo-129) pernašos modelis buvo sudarytas kompiuterine programa *FEFLOW*, laikantis esminės prielaidos (Švedijos ekspertų pasiūlytas bei Lietuvos energetikos instituto specialistų apskaičiuotas konteinerio defekto scenarijus) bei pagal Pietų Lietuvai būdingas geologines sąlygas ir vyraujančius parametrus (0,8×0,6×0,52 km tolimojo lauko blokas). Darbo metu buvo modeliuotas požeminio vandens srautas, kylantis per tektoniškai suardytą zoną, kurioje yra patalpintas pažeistas konteineris. Pagrindiniai skaičiavimų rezultatai: didžiausias ^{129}I tūrinis aktyvumas, kai požeminio vandens srautas, kylantis tektoniškai suardytoje zonoje šalia konteinerio, nesieks 1 Bq/l, aktyvioje vandens apytakos zonoje – iki 10^{-2} Bq/l. Rezultatai rodo, kad žmogaus gaunama dozė, jei jis gertų požeminį vandenį, neturėtų siekti ribinės dozės (0,2 mSv/m). Modeliavimo darbai, remiantis sudėtingesnėmis prielaidomis bei geologinėmis sąlygomis, bus atliekami ateityje.

Prasminiai žodžiai: panaudotas branduolinis kuras, geologinė saugykla, kristalinis pamatas, požeminis vanduo, skaitmeninis modeliavimas, baigtiniai elementai.

МОДЕЛИРОВАНИЕ ПОТОКОВ ПОДЗЕМНЫХ ВОД И ПЕРЕНОСА РАДИОНУКЛИДОВ В КРИСТАЛЛИЧЕСКОМ ФУНДАМЕНТЕ КОМПЬЮТЕРНЫМ КОДОМ *FEFLOW*

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Р е з ю м е

Стратегия обращения с радиоактивными отходами Литвы в качестве одной из альтернатив предусматривает оценку возможностей захоронения отработанного ядерного топлива и долгоживущих радиоактивных отходов в глубоких геологических формациях. Начальная оценка пригодности всех потенциальных геологических формаций для захоронения отработанного ядерного топлива произведена в Литве в течение 2001–2004 гг. Из-за большого объема существующей разносторонней информации кристаллический фундамент был отобран в первую очередь для моделирования потоков подземных вод и переноса радионуклидов.

Принимая во внимание главные предпосылки нормального развития (предложенный шведскими экспертами и оцененный экспертами LEI сценарий дефектного канистра с отработанным ядерным топливом) и подобранные параметры, характерные для кристаллического фундамента и вышележащей толщи осадочных пород Южной части Литвы (0,8×0,6×0,52-километровый блок дальнего поля), моделировался перенос радионуклидов (йода-129 как мобильного и долгоживущего радионуклида) с потоком подземных вод, используя компьютерный код FEFLOW. Через дефектный канистр, гипотетически расположенный в тектонически нарушенной зоне, средствами моделирования был воссоздан восходящий поток подземных вод.

Главные результаты численного моделирования для очень консервативного случая таковы: при восходящем потоке подземных вод максимум активности I-129 в воде тектонически нарушенной зоны рядом с дефектным канистром не будет превышать 1 Бк/л, а в зоне активного водообмена он близок к 10^{-2} Бк/л. Результаты показывают, что получаемые человеком через питьевую воду дозы должны быть меньше ограниченной дозы (0,2 м^3/год).

Ключевые слова: отработанное ядерное топливо, геологическое хранилище, кристаллический фундамент, подземные воды, численное моделирование, конечные элементы.

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