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Multibarrier system preventing migration of radionuclides from radioactive waste repository

**Wioleta Olszewska,
Agnieszka Miśkiewicz,
Grażyna Zakrzewska-Kołtuniewicz,
Leszek Lankof,
Leszek Pająk**

Abstract. Safety of radioactive waste repositories operation is associated with a multibarrier system designed and constructed to isolate and contain the waste from the biosphere. Each of radioactive waste repositories is equipped with system of barriers, which reduces the possibility of release of radionuclides from the storage site. Safety systems may differ from each other depending on the type of repository. They consist of the natural geological barrier provided by host rocks of the repository and its surroundings, and an engineered barrier system (EBS). The EBS may itself comprise a variety of sub-systems or components, such as waste forms, canisters, buffers, backfills, seals and plugs. The EBS plays a major role in providing the required disposal system performance. It is assumed that the metal canisters and system of barriers adequately isolate waste from the biosphere. The evaluation of the multibarrier system is carried out after detailed tests to determine its parameters, and after analysis including mathematical modeling of migration of contaminants. To provide an assurance of safety of radioactive waste repository multibarrier system, detailed long term safety assessments are developed. Usually they comprise modeling of EBS stability, corrosion rate and radionuclide migration in near field in geosphere and biosphere. The principal goal of radionuclide migration modeling is assessment of the radionuclides release paths and rate from the repository, radionuclides concentration in geosphere in time and human exposure to ionizing radiation.

Key words: engineered barrier • migration • radionuclides • repository • radioactive waste

W. Olszewska, A. Miśkiewicz✉,
G. Zakrzewska-Kołtuniewicz
Institute of Nuclear Chemistry and Technology,
16 Dorodna Str., 03-195 Warsaw, Poland,
Tel.: +48 22 504 1270, Fax: +48 22 811 1917,
E-mail: a.miskiewicz@ichtj.waw.pl

L. Lankof, L. Pająk
The Mineral and Energy Economy Research Institute
of the Polish Academy of Sciences,
7 Wybickiego Str., 31-261 Krakow, Poland

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Introduction

Existing site selection requirements for radioactive waste repositories, undertaken measures to improve safety barriers, radioactive waste processing and solidification technologies and reprocessing of nuclear fuel, as well as the work on the behavior of radionuclides in natural ecosystems, are the components of safety and determinants of safety culture. At the same time all these elements contribute to the strategy of sustainable radioactive waste management and to sustainable development in general [1].

Poland faces the challenge of choosing a location for the new surface disposal site for low and intermediate level radioactive waste, which is to replace the repository in Rozan. Rozan repository will be closed-up after 2020, which implicates establishing of the long-term program of site monitoring with permanent inspection of the system of barriers. The new facility, which is under selection now, will need a preparation of safety assessment that involves evaluation of the security of protected barriers with mathematical codes developed for this purpose. This will be a guarantee for long-term safety of the closed repository and safe operation of the new facility.

The problems which are raised in this article concern prevention of radionuclides migration in the surroundings of radioactive waste repository. This is an important issue at every stage of repository performance: from siting to final closure and during the phase of long-term post-closure monitoring. The appropriate method of waste conditioning before disposal, engineering multibarrier system and researches which help to learn and understand the migration routes and the accumulation of radionuclides in the environment, should effectively prevent their penetration into the biosphere as a result of black scenarios [2].

The simulation example of radionuclide migration in geosphere was also presented in this article. The example concerns hypothetical release of radionuclide in saturated porous media from constant source. Usually a multibarrier system tends to isolate the waste of such an environment but presented example has only demonstrative character of the TOUGH2 simulator usage in radionuclide migration in geosphere.

Multibarrier system

The multibarrier system is used to provide the safety of radioactive waste storage in near surface repositories. The following requirements must be met for proper isolation of radioactive waste:
– reduction of waste volume,
– appropriate waste form preventing radionuclides leaching and dispersion,
– storage in an environmentally acceptable way.

There are many protective barriers (multibarrier system), which consist of natural (geological) and artificial (engineered) barriers. The system is designed to provide good protection against release of radioactive substances from a repository and their further migration. The main safety objective of the multibarrier concept of disposal system is the isolation of the waste and the containment of the radionuclides in the waste. Nevertheless, it happens that the engineered barriers could lose their ability to provide complete containment in time. Because some radionuclides decay extremely slowly and/or are mobile in deep groundwater, their complete containment is not possible [3].

The EBS comprises a variety of subsystems or components, such as the waste form, canister, buffer, backfill, seals, and plugs. The major purpose of an engineered barrier system is to prevent and/or delay the release of radionuclides from the waste to the repository host rock. EBS should function as an integrated system and ensure proper physicochemical conditions so that all barriers can fulfill their intended function [4].

Prior to storage the radioactive waste has to be treated, including volume reduction and reduction of radioactive compounds and other solute in the effluent. For liquid radioactive waste treatment, many methods as precipitation, sedimentation, ion exchange, thermal evaporation and membrane methods are applied [5, 6]. To provide effectiveness of EBS, many natural sorbents (e.g. clay, bentonite or zeolites) as well as synthetic ones (e.g. synthetic zeolites) are applied to reduce the migration of radionuclides from contaminated sites [7]. They could be used as buffer, backfill and sealing materials in the repository.

Barriers can provide partial containment and/or complete isolation of waste. Their effectiveness depends on the complex interactions of natural and engineered barriers. The barriers can be described in terms of their function, for example, a cut off wall is used to limit horizontal migration of groundwater [8].

The main EBS barriers and their functions are presented in Table 1.

Table 1. The main EBS barriers and their functions [8, 9]

Engineered barrier system (EBS)	
Barrier	Function
Waste form (binders of concrete, asphalt, organic polymers and ceramic mass)	Solidification of radioactive waste, in order to prevent scattering, dispersion, spraying and leaching of radioactive substances.
Waste container (metal and concrete containers)	Isolation of waste from the environment, protection against mechanical damage, extremes of weather and contact with water.
Backfills (clays, clay mixtures, cement, rock materials, soil)	Limitation of water infiltration, sorption and precipitation of radionuclides, gas control and if necessary facilitation of waste retrieval. Limitation the release of radionuclides from the waste-form to the far-field (geosphere) after container failure.
Structural material and liners (concrete, reinforced concrete, clay and asphaltic or organic membranes)	Ensuring physical stability of repository structure, additional protection of waste, especially from the extreme weather, containment barrier.
Drains (combinations of clay and gravel blankets or conventional ceramic or concrete drains)	Water and leachate management, particularly during the disposal facility's institutional phase.
Cap	Covers the surface layer of concrete, prevents the infiltration of rainwater into the waste storage area, prevents corrosion of packaging, prevents leaching of radioactive substance.

In multibarrier system, EBS is complemented by the natural barrier system comprising geological conditions as well as surface conditions. Location of radioactive waste repository is strictly limited by the siting criteria. In site selection for near surface waste repository, geological environment is evaluated in terms of host rock lithology, lithological units the presence of discontinuities, possible contaminant transport paths, mechanical, hydrogeological, geochemical and thermal processes and conditions. Surface environment is evaluated in terms of, e.g., topography and morphology, types of soil sand sediments, hydrological and atmosphere conditions and the rate of erosion and deposition.

Site for radioactive waste selected according to the criteria is very efficient barrier ensuring:
- isolation of the waste from people and the natural environment surface by providing a massive shield against radiation;
- protection of the engineered barrier system by providing a stable mechanical and chemical environment;
- appropriate rock properties and a weakly dynamic hydrogeological environment that controls the rate at which deep groundwater can move to, through and from the backfilled and sealed facility, or completely prevent flow;
- stable chemical, mechanical and hydrogeological evolution of the storage system;
- sufficient properties that retard the movement of any radionuclides in groundwater – these include sorption onto mineral surfaces and properties that promote hydraulic dispersion and dilution of radionuclide concentrations;
- dispersion of gases produced in the facility so as to prevent mechanical disruption of the engineered barrier system [3].

The study of migration of radionuclides in repository surroundings

To evaluate the long-term safety of the multibarrier system, the safety assessment is developed. Usually they comprise modeling of EBS stability, corrosion rate and radionuclide migration in near field in geosphere and biosphere. The principal goal of radionuclide migration modeling is assessment of the radionuclides release paths and rate from the repository and radionuclides concentration in geosphere in time. These data complemented by the results of biosphere modeling allow for evaluation of human exposure to ionizing radiation [10].

Degradation of barrier system may result in radionuclide release and migration in repository surroundings. The radionuclide release types, main mechanisms of release and type of primary receiving media are shown in Fig. 1.

Behavior of radionuclides in natural ecosystems depends on chemical and physical interactions with mineral and organic matter of the soil. Migration of pollutants, e.g., in the soil depends on sorption properties (absorption in a soil and adsorption with accompanying chemical reaction). One of the most

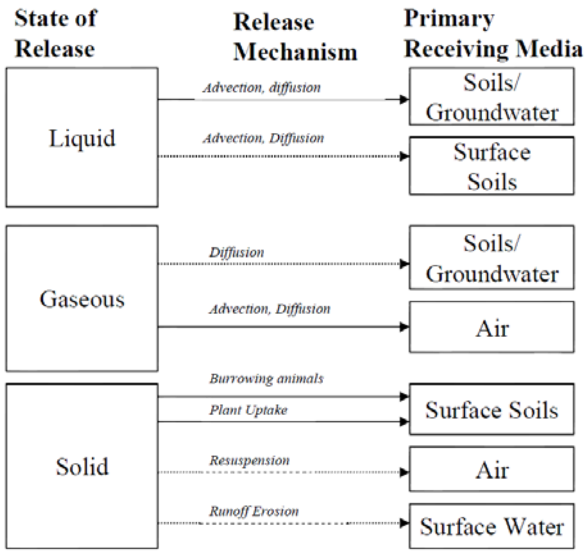

Fig. 1. Example of release mechanisms due to state of release [11].

important sorption mechanisms is the sorption by ion-exchange sorption of exchangeable. It depends on exchange capacity relative to the cations, pH, composition of soils, content and quality of organic matter, a surface area of soil colloids, the type of cations in the solution, kind of competing anion and temperature. The sorption by ion-exchange sorption of exchangeable relative to the cations is a reversible process and dependent on soil parameters.

The main physical and chemical characteristics of the soil influencing the migration of the radionuclides are: granularity, mineral component, density, porosity, permeability, natural humidity and the pH [12]. The rate of migration in soils depends among others on the water saturation and on clay content resulting slower migration of radionuclides. It is necessary to study sorption parameters of material, which can bind radionuclides. A good location of a repository for radioactive waste must have high capacity for ionic exchangers, small porosity and lowest permeability of soil [13].

The next mechanism of accumulation of pollutants is a sorption of biological matter. This is immobilization of metal ions by living organisms or plants present in the soil environment. One of the mechanisms of penetration of the metal to organisms is transport in the form of complex with organic substances.

The diverse redistribution of radionuclides in the soil depends on: physicochemical properties of soil, intensity and kind of rainfall, deposition time, sorption and re-deposition by plant, and other biological factors [2].

The studies of migration and accumulation of radionuclides in environmental components are important in the context of the siting process of a new radioactive waste disposal facility and the closure of the old repository. Closure of the repository binds to organizational and technical activities. It is necessary to isolate waste of the biosphere, therefore the analyses of kind of radioactive waste disposed, its activity, type of storage, security barriers, kind of repository and applicable regulatory provisions should be analyzed. The closed repository in Rozan will be monitored for many years. The scientists and engineers will work on measurements of radionuclides in the environment and on inspection of designed engineered barriers.

Mathematical codes

Numerical modeling of radionuclide migration requires development of conceptual models comprising description of radionuclide release scenarios in spatial and temporal terms and their implementation into mathematical models with governing equations of relevant processes. Mathematical simulation is a set of operations comprising:

- development of computational models based on physicochemical processes;
- their verification using available experimental setups;
- analysis of variants associated with prediction of the development of the processes and optimization of possible consequences.

In the context of mathematical modeling, it is necessary to properly define the mechanism, which influences the migration of radionuclide. The wording of the flow model, which consists of the equation of continuity, momentum balance, resistance to motion with the initial and boundary conditions is a next step of this approach. It is necessary to prepare a model of mass flow and to obtain physicochemical data from the literature, geological reports and technical documentation. The migration of radionuclides depends on the size of the modeled area or division into compartments. Selecting the appropriate code or computer program that will create a grid discretization (triangular or rectangular) is necessary to carry out computer simulations and present the results in graphical form.

Four groups of mathematical codes are identified for modeling the radionuclide migration in radioactive waste storage systems. There are codes for simulating processes in the near field, geosphere, biosphere and in a total system of radioactive waste disposal. A great number of modeling works are carried out to determine the pathways of and rate of radionuclide migration in geosphere.

The following codes are generally used to simulate the migration of radionuclides in geosphere: TOUGH2, MODFLOW, MT3D, FEFLOW, and PORTFLOW (Fig. 2). One of the most popular is TOUGH2 – a numerical simulator for nonisothermal flows of multicomponent, multiphase fluids in one-, two-, and three-dimensional porous and fractured media.

TOUGH2 is designed for geothermal reservoir engineering, nuclear waste disposal, environmental assessment and remediation, and unsaturated and saturated zone hydrology. The first version of TOUGH2 code provided five different equation-of--state modules (EOS). The new Version 2.0 release of TOUGH2 includes improved versions of these five EOS-modules and provides a more flexible and comprehensive description of flow systems and processes, e.g., new user features, such as enhanced

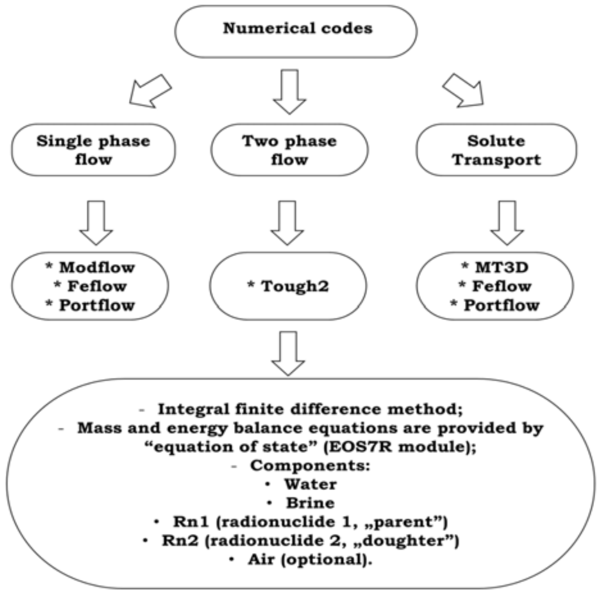

Fig. 2. The computer codes used to model migration of radionuclides in geosphere.

linear equation solvers, and direct outputting of graphics files [14].

In order to simulate the migration of radionuclides in environment the EOS7R is used. This is an enhanced version of the EOS7 module in which two additional mass components have been added. The module enables selection of radionuclides with user-specified half-life that are of the user's interest. There are two types of radionuclides in this model. The radionuclide 1 (Rn1) is named the 'parent' and radionuclide 2 (Rn2) – the 'daughter' [15].

The software is based on the finite difference method, which is one of the simplest and of the oldest numerical methods to solve differential equations. The method has many advantages. The mathematical formulation and computer implementation are easily understood for simple problems, for example, one-dimensional or steady-state groundwater flow. The textbooks are very helpful to the beginner. Efficient numerical algorithms have been developed for implementing the finite difference method on computers.

The example of modeling using TOUGH2 code

Numerical modeling of radionuclide migration in geosphere

The computational abilities of TOUGH2 simulator are presented in hypothetical simulation of radionuclide migration in saturated porous media from constant source of radionuclide.

Simulator uses finite differential method for multiphase and multicomponent modeling in porous and fractured media in unstable conditions [16].

Model is located in Cartesian coordinate system X, Y, Z. The model covers an area of 1000 m (X from 0 to 1000 m) to 1000 m (Y from 0 to 1000 m) to 300 m (Z from –300 to 20 m). In the middle of the area in the element (X = 485 m, Y = 512.5 m and

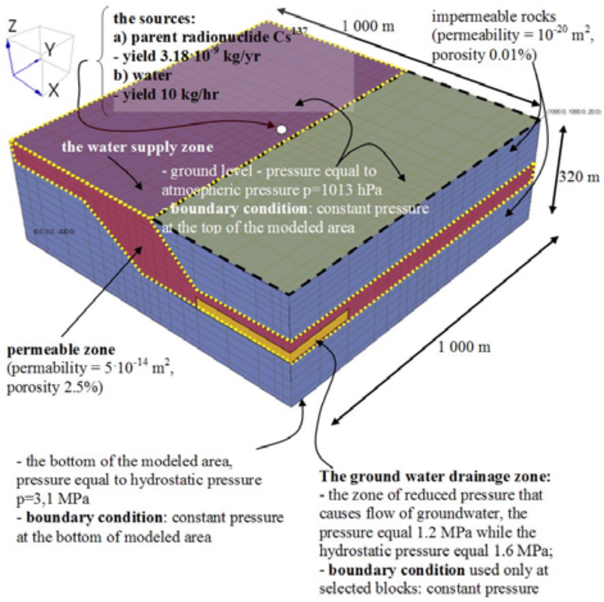

Fig. 3. Discretization scheme, area location in the coordinate system and the boundary conditions of the model.

Z = 9 m) the source of the radionuclide is located. Discretization scheme, area localization in the coordinate system and the boundary conditions of the model are shown in Fig. 3.

Two types of geological formations were distinguished in the modeled area – the permeable and impermeable ones. Parameters of these formations are as follows: permeability 10^{-13} m² (100 mD) and 10^{-20} m² (10^{-5} mD), respectively, effective porosity – 2.5% and 0.1%, respectively. The location of geological formations is presented in Fig. 3. The numeric calculations were carried out for isothermal conditions using the module (equation-of-state) EOS7R intended for modeling the transport of radionuclides in geological media [16].

The initial conditions were defined by modeling the steady-state pressure distribution in the modeled area using the boundary conditions presented in Fig. 3. In Fig. 4, the pressure distribution diagram for the initial conditions is shown. The adopted boundary conditions caused asymmetric flow of groundwater in the permeable formation from the feed zone located upon the surface (Fig. 5). Groundwater flow to the discharge area located at a depth of approx. –155 m in the corner of the modeled area (Fig. 3).

The discharge zone was generated by setting the first kind boundary condition in the corner grid elements of the modeled area (Fig. 3). In these grid elements the decreased pressure of approx. 1.25 MPa was applied (Fig. 4). The rate of groundwater flow is highest in the vicinity of the discharge zone and decrease to feed zone located in the opposite corner of the modeled area.

In this hypothetical example the migration of ^{137}Cs radionuclide was modeled. Cesium-137 has a half-life of about 30.17 y and about 95% decays by beta emission to a stable nuclear isomer of barium. Cesium radionuclide source was located approximately in the center of the modeled area (Fig. 2). Source generates parallel radionuclides and water. The rate of source is 0.1 kg of cesium per year and 10 kg of water per hour. Parallel generation of radionuclides and water simulates permeation of contaminated leachate into saturated geological formation.

In Figs. 6 and 7, the calculation results of contamination propagation after 100 years are presented. The shape of contamination isosurfaces reflects the hydrodynamical conditions in the model area. To facilitate interpretation of results presented on diagrams, the vectors of groundwater flow are presented additionally in the range as in Fig. 5. The ^{137}Cs contamination plum extend is significantly larger than for its decay product. However it does not reach the model borders in modeled span of time. The extend of cesium contamination plum of 10^{-9} mass fraction concentration ranges up to 500 m and barium plum of the same concentration ranges up to 200 m in 100 years.

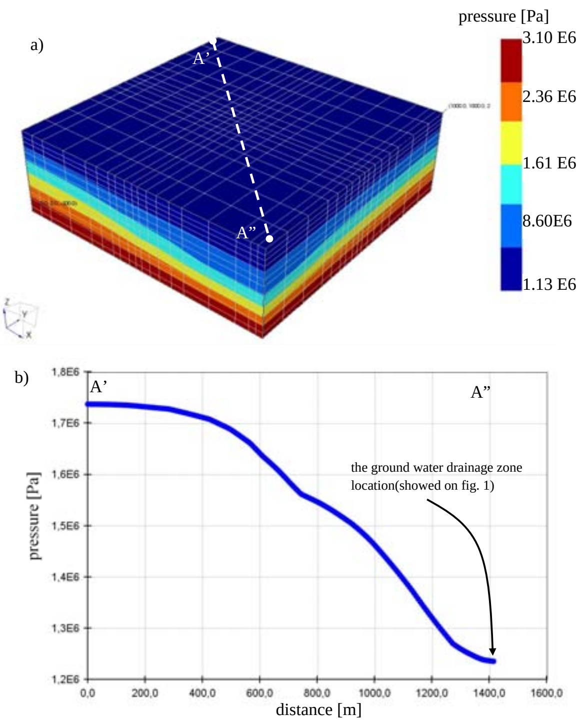

Fig. 4. Initial pressure distribution at the modeled area: a) view similar to that presented in Fig. 3, b) pressure changes along the profile of A-A indicated in fig. a) at a depth of 155 m – water discharge zone depth (showed in Fig. 2).

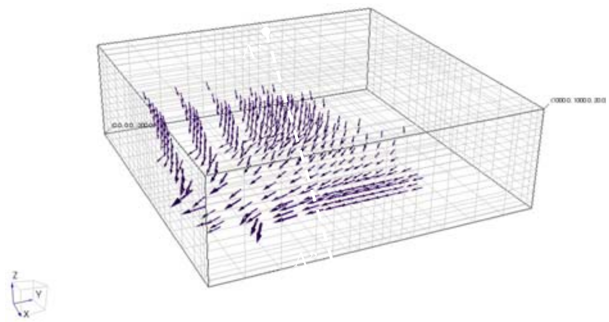

Fig. 5. The rate of groundwater flow at the range from 10^{-6} to 7.5×10^{-6} m/s (31.56–236.68 m/year) – the length of the arrows is proportional to the rate value within the given range.

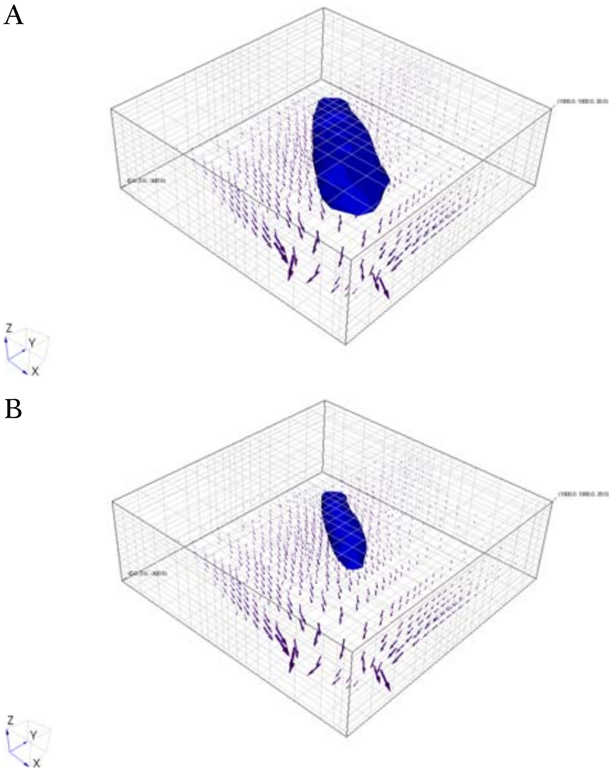

Fig. 6. The range of isosurface of 10^{-9} mass fraction concentration of ^{137}Cs (A) and ^{137}Ba (B) after 100 years from the beginning of radionuclide release from the source and groundwater flow rate (range of rate similar to Fig. 5).

The aim of the calculations was testing and presenting the computational capabilities of the TOUGH2 simulator used for modeling radionuclide contamination propagation in the geological environment taking into account the decay of radionuclides in time.

Despite the simple conceptual model presented problem confirms the expediency and possibility of using this software for modeling complex three-dimensional issues related to the subject issue.

The simulator is quite common in issues related to groundwater flow modeling, with special emphasis on geothermal energy – as evidenced by the extensive literature related to this subject [17–19].

Summary and conclusions

Radionuclides could migrate through the engineered barriers and fractures in the host rock by various transport mechanisms. In order to evaluate the suitability of a proposed repository site, it is necessary to understand the hydrogeological conditions at the site, and estimate the rates at which radionuclides could migrate along fractures and other permeable structures that may be present. Very strict criteria for site selection for the disposal of nuclear waste are solely dictated by safety issues.

The geological hydrogeological and climatic conditions are continuously changing. Therefore, it is necessary to design barriers to prevent the release of radioactive material at the place of storage, and barriers to prevent migration of the substance.

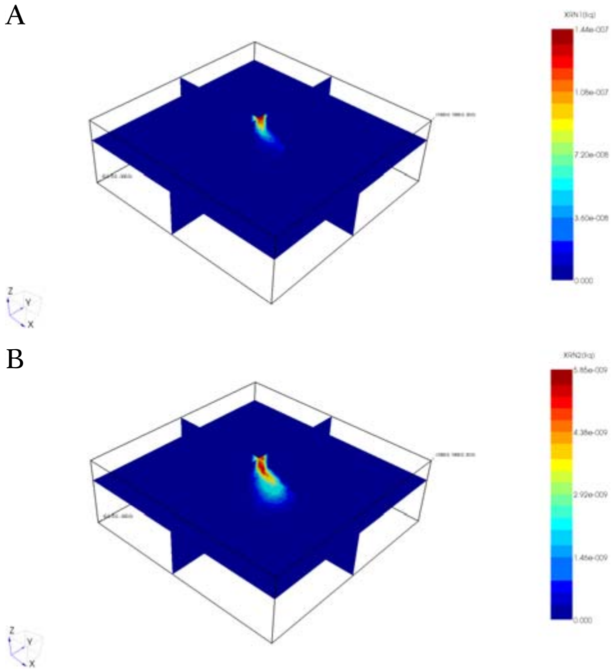

Fig. 7. The concentration of ^{137}Cs (A) and ^{137}Ba (B) mass fraction after 100 years from the beginning of radionuclide release from the source.

To evaluate the safety of radioactive waste repository multibarrier system detailed long-term safety assessments are developed which comprise modeling of EBS stability, as well as radionuclide migration in geosphere and biosphere. Radionuclide migration modeling provides data concerning the radionuclides release paths and transport rate from the repository; radionuclides concentration in geosphere in time and human exposure to ionizing radiation.

The multibarrier system and long-term safety assessment comprising numerical modeling should be considered as a coherent protection system against the various black scenarios of uncontrolled radioactive release to the environment. The construction of the new disposal sites and safe closure of the old repositories require proper design of multibarrier system, and involve permanent inspection of this system, as well as safety assessment with application of developed according to the site-specificity mathematical codes.

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