

PRINCIPLES ENSURING COMPREHENSIVE ACCOUNTING OF PROCESSES ASSOCIATED WITH ENGINEERED SAFETY BARRIERS' EVOLUTION: ASSESSING RADIONUCLIDE SPREAD BEYOND SITE BOUNDARIES

Krychkov D. V., Boldyrev K. A.

Nuclear Safety Institute of the Russian Academy of Sciences, Moscow, Russia

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The article discusses principles and approaches applied in the safety assessment of facilities based on the example of a modern tool DESTRUCT being currently developed. The code takes into account a set of processes associated with the evolution of engineered barrier system and is designed to predict the release of radionuclides from the near field.

Keywords: *radioactive waste storage facility, radioactive waste, disposal, decommissioning, design modeling, engineered safety barriers, evolution, migration, comprehensive safety analysis.*

Introduction

Modern approach to the safety assessment of radioactive waste disposal facilities (RWDF) suggests that changes in their properties should be considered taking into account all the processes being important in terms of assessing the dynamics of radionuclide spread through a barrier cascade. Thus, a comprehensive analysis is required to evaluate the dynamics of the materials' inherent properties evolution, including their mutual influence.

This approach is implemented through the development of computational tools enabling a comprehensive analysis of EBS properties to predict their degradation and the spread of radionuclides outside the studied facility.

Special aspects in describing EBS evolution

Figure 1 presents a general flow chart describing the development an integral model (IM) for contaminant spread through a cascade of evolving

safety barriers. The above diagram shows the potential for the formation of new containment barriers due to the degradation of already existing ones, as well as the degradation driven influence produced by the properties of aggressive medium on the rate of subsequent barrier degradation [1].

A list of most important processes and their parameters and properties should be specified to develop a sound approach enabling to perform relevant calculations and to supply the initial data during the development of IM base-line principles.

Portland cement-based concrete, steel, bentonite, glass matrices of various composition for RW Class 1 immobilization, cement-based matrices designed for the solidification of other RW classes are considered as the key materials in the construction of RW storage and disposal facilities.

The impact produced by the following physical and chemical processes on material properties

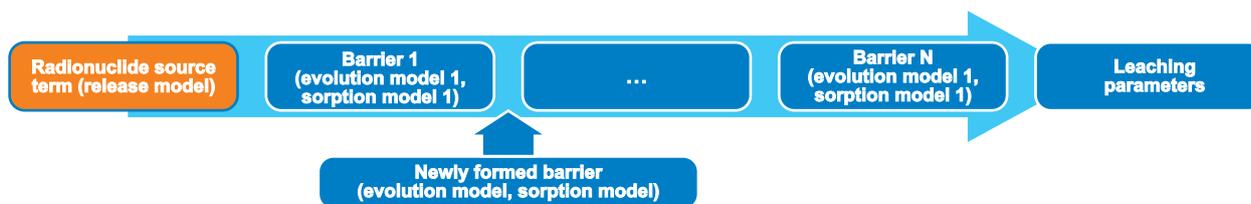


Figure 1. IM block diagram showing radionuclide spread through EBS cascade taking into account the barrier degradation with time

should be taken into account in the development of material transformation IMs:

1. Corrosion of metal structures.

The rate of metal structures corrosion is affected by:

- chemical composition of metal (inherent properties of the barrier);
- chemical composition of the environment in which the metal barrier is located and the composition of aqueous solution interacting with it;
- temperature.

Approaches being based both on the extrapolation of reported corrosion rates and numerical corrosion rate calculations are implemented in the software calculation complex (CC) DESTRUCT developed by IBRAE RAS [1].

Metal structures of insulating container corrode resulting in the loss of their waterproofing capacities. At the same time, corrosion products contribute to the formation of some new barriers producing a significant impact on other materials, which should be taken into account in migration calculations.

2. Degradation of concrete barrier materials.

Durability of Portland cement- (concrete-) based material firstly depends on its structure, chemical composition and also on environmental conditions. According to concrete failure classification proposed by Moskvina, three main types of concrete material corrosion can be distinguished:

- processes accounting for dissolution of the main cement stone components (due to water seepage through the concrete with readily soluble sodium hydroxides leaking at the first place followed by calcium hydroxides);
- formation of mobile corrosion products weakening the cement stone structure (can be also released with a seepage flow);
- interaction of concrete components with the seepage water results in the formation of potentially large amounts of relevant by-products within the massif, its pores, cracks and capillaries which lead to cement stone degradation. Thus, considering the above processes, the following flow sequence is implemented to develop a correct model of contaminant transport through concrete-based materials:

- description of the concrete type, assessment of its mechanical and physical properties;
- development of a model presenting concrete degradation over time considering relevant physical, biological and chemical transformations;
- identification of radionuclides being transported from the source term, selection of optimal sorption and deposition models (for redox-sensitive elements, as well as elements being able to form hardly soluble mineral phases on their way through a concrete barrier);
- final forecast involving the assessment of radionuclide transport and identification of a time dependence for radionuclide migration through a barrier.

3. Bentonite evolution over time. There are several possible scenarios describing bentonite barrier evolution:

- a change in its mineral composition (for example, due to illitization, chloritization or cementation of bentonite by corrosion products of metallic materials);
- a change in its sorption and seepage properties due to evolved chemical composition of groundwater;
- cooccurrence of the above scenarios.

High-temperature illitization of montmorillonite is seen as a most unfavorable process for bentonite. Compared with montmorillonite, illit (micaceous clay) has lower waterproofing and sorption capacities with high temperature and high concentration of potassium in the contacting solution seen as factors favoring the development of this process.

According to numerous earlier studies, montmorillonite illitization is impossible at a temperature below 110 °C, for this reason it is believed that bentonite illitization in storage facilities with a temperature of less than 110 °C can be excluded from consideration [2].

4. Glass matrix evolution

Solution composition does not match the composition of the soluble solid phase when the glass matrix is dissolved, which is explained by the formation of solid glass hydrolysis products and precipitation of poorly soluble compounds. Thus, due to borosilicate glass dissolution soluble silicic acid ions pass into the solution with hardly soluble metal oxyhydroxides constituting to the matrix remaining on the glass surface.

Temperature, acidity and chemical composition of the solution impact significantly on the dissolution rate of the glass matrix.

The above spectrum of the considered material properties evolution is taken into account when a chamber modeling method is used to solve a system of equations describing contaminant distribution within the allocated volumes (chambers) of the investigated space bounded by relevant safety barriers of the facility.

Basically, elements of the multi-barrier safety system are affected not only by the internal mechanisms determining the evolution its constituent elements, but also by the external environment being characterized by evolving properties. In this regard, to provide an accurate accounting of the external factors, not only time-dependent material properties should be considered, but also the boundary conditions being considered external to the barrier system. Thus, in particular, if some radioactive components are available within the system, changes in their concentrations associated with radioactive transformations is taken into in relevant predictions of contaminant spread.

Mathematically, the problem suggesting chamber modeling of contaminant spread is addressed via a system of linear differential equations describing the evolving concentration of substances in the considered space volumes (chambers). Basically, the system can be presented as follows:

$$\frac{dI_i^m}{dt} = -\left(L_r^m + \sum_i L_{i,j}\right)I_i^m + L_r^{m+1}I_i^{m+1} + \sum_j L_{j,i}I_j^m, \quad (1)$$

where I is the concentration (amount) of the considered substance in each chamber, $L_{i,j}$ is the exchange rate between the chambers, L_r^{m+1} is the decay rate of the parent substance relative to the m -th substance; L_r^m is the decay rate of the m -th substance. Indices i and j describe the values of each component in each chamber. This system of equations can be presented in a matrix form:

$$\frac{d\vec{I}}{dt} = \vec{A}\vec{I}, \quad (2)$$

where the vector \vec{I} is composed of all considered substances in all elementary volumes.

In some cases, the system may have an analytical solution, but in the most general one it can only be solved numerically.

Integrated accounting of the interconnected processes describing multi-barrier system evolution

CC DESTRUCT is designed to predict the release of contaminants from the near field taking into

account a wide range of processes occurring during EBS degradation.

The developed CC DESTRUCT elements (calculation units) allow predicting the behavior of each barrier taking into account the conditions of its existence and evolution. Calculation results are integrated into a knowledge base as files with scenarios of time-dependent parameter alterations. Further estimates of material behavior, as well as migration are performed with due account of scenarios presenting evolving properties of barriers and environmental characteristics.

The modeling unit responsible for the safety barriers degradation implies the consideration of their physical destruction along with successive alterations in their physical, chemical and migration parameters.

The unit being responsible for the forecasts on the evolution of bentonite provides the possibilities for illitization evaluation, as well as for assessing the decrease in the amount of montmorillonite over time depending on the temperature and composition of the aqueous medium.

The unit being responsible for the forecasts on the evolution dynamics of concrete materials can evaluate concrete degradation by various mechanisms, including carbonation of concrete and acid and sulfate corrosion based on temperature and water characteristics.

The unit being responsible for the forecasts on the evolution dynamics of steel materials allows to identify the time and the nature of steel structural element degradation based on time, temperature and properties of the aggressive medium.

The unit being responsible for chemical process accounting is used by almost all calculation modules in which various aspects of material evolution is considered to be essential for the performed calculations.

Thus, in particular, CC DESTRUCT is able to evaluate actual characteristics of a package (matrix material). The developed algorithm accounts for the presence of a movable and solid immobile phase, including a semipermeable near-surface transition layer and the monolithic one. During the research, the following algorithms were developed to calculate leaching:

- in keeping with constant composition law based on a linear dependence on the mass fraction for sodium aluminophosphate glass matrix;
- taking into account chemical interactions based on a given dependence taking into account the supersaturation of the solution along the leading mineral phases of the cement matrix, as well as based on the dependence taking into account the supersaturation of the solution along the

leading mineral phase for glass matrices of different compositions.

As regards the opportunities for evaluating the structural elements and materials constituting to the waste disposal facility, CC DESTRUCT is able to assess the state of the main structural elements and materials: concrete, metal and clay structures. As mentioned above, these are considered as safety barriers, the characteristics of which should be evaluated.

Corrosion mechanisms are simulated for metals to identify the time period during which the steel can maintain its protective properties.

It should be noted that an approach based on the identification of concrete degradation stages was used to assess concrete degradation: it was assumed that these stages are characterized by specific redistribution and alterations of the mineral phases constituting to the cement stone that are basically responsible for the pH level of concrete pore water.

Clay material evolution can either cause bentonite destruction at the mineral level (for example, during illitization) or only contribute to some changes in its sorption capacity due to its interaction with waters the composition of which differs from the original pore water.

CC DESTRUCT can evaluate the altered properties of the main safety barriers with non-stationary processes that could be accounted for as well.

Development of EBS models involves the use of a mathematical apparatus to describe physical and chemical mechanisms resulting in barrier destruction and their changing properties. CC implements and integrates models presenting physical properties of the two basic materials: steel and concrete.

Material degradation is simulated with due account of a number of contributing factors including temperature, chemical and biological composition of the medium and etc. Chemical properties can be refined for any parameter based on the developed templates for mineral phase behavior and contaminant migration in each barrier can be calculated. Comprehensive analysis of material degradation based on previously described models yields a dependence presenting the dynamics of contaminant spread characteristics in the near field.

Evaluation of migration parameters enables to obtain a time dependence of their numerical values (distribution coefficient, sorption, filtration, diffusion coefficients, etc.).

The resulting set of numerical values was integrated into a single migration model taking into account the dynamic parameters of the contaminant spread and the main features of the occurring processes.

Thus, the entire system is described by a single IM using time-dependent parameters, taking into account relevant alterations in physical and chemical properties of materials during their evolution and their effects on contaminant spread in the near field.

Consideration of dynamic parameters in the radionuclide spread assessment

Basic structure of a unit designed to predict radionuclide migration in the near field of the studied facility involves:

- a module used to set up some general characteristics of the migration simulation calculation specifying global variables of the initial and final calculation time, maximum readout step, as well as links to chamber and 3D calculation tools and the storage location of the source files;
- a chamber modeling module that can provide relevant estimates on the scale of radionuclide spread beyond the engineered barrier system;
- module enabling 3D calculations of radionuclide migration is used for detailed modeling of radionuclide distribution in the near field taking into account local inhomogeneities.

Material generation module, geometric data import module and model development interface module are considered as the essential ones in setting the parameters of safety barrier degradation.

The materials module allows for setting material migratory properties (water permeability, porosity, sorption interaction parameters (sorption coefficients), diffusion coefficients) via variables based on relevant scenarios and schedules.

Considering the setup presented above, CC DESTRUCT provides coupled solution to the multi-physical problems associated with contaminant spread in the near field. It takes into account relevant changes in the properties of engineered barriers resulting from material degradation and applies coupling methods to combine relevant models.

Examples of comprehensive solutions to the problems associated with the evolution of a multi-barrier safety system

The following examples were chosen to demonstrate the modeling capacities as regards the main processes that should be taken into account in the assessment of material state:

- corrosion of metal structures exemplified by enclosed disposal containers for the NKM deep disposal facility (NKM DDF);
- evolving properties of concrete materials in case of their application in near-surface storage facilities for RW;

- evolving sorption properties of bentonite materials for the NKM DDF;
- radionuclide leaching from sodium-aluminophosphate glass applied for HLW disposal purposes under NKM repository conditions.

Results of complex calculations on radionuclide releases were presented as well:

- release from a waste package for DDF RW Class 1;
- release from a reinforced concrete container;
- release beyond the tailings;
- release beyond near-surface disposal site boundaries.

Corrosion of metal structures exemplified by enclosed containers for RW disposal in a DDF

CC DESTRUCT suggests that numerical methods can reflect the evolving properties and are applied to describe the corrosion damage of steel which is done through the identification of a dependence between the corrosion rate (its penetration depth over time), temperature, pH and Eh of the medium, strain stress, the degree of impact produced by radiolysis and altered properties of the matrix and the groundwater.

First of all, pitting (local) corrosion should be evaluated, since even a smallest hole can result in barrier failure triggering water flow through the barrier. Therefore, a discrete change in the seepage properties of a metal barrier is considered, since seepage would not occur as long as the barrier integrity is not breached due to corrosion or mechanical failure. As soon as the integrity of a wall is breached, the barrier becomes permeable.

A general case suggests that the corrosion rate of this type depends on the intensity of the external load driven by various mechanisms.

Numerical methods developed to calculate the corrosion rate of steel structures were tested enabling to estimate the time required for their

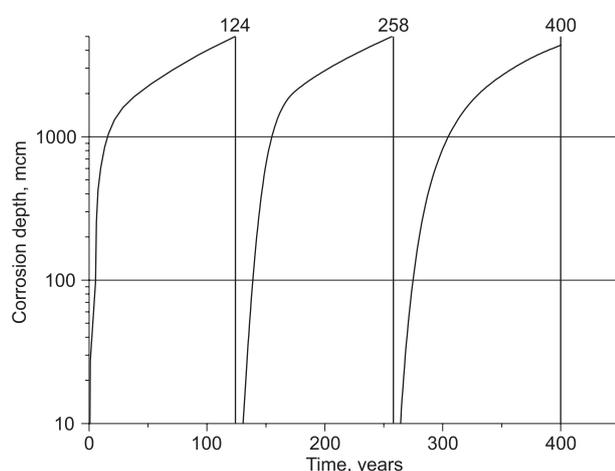


Figure 2. Calculated metal barrier degradation

failure and the time needed for the water to reach the source term taking into account the temperature of the structure surfaces.

Figure 2 shows staged corrosion driven degradation of a few steel barriers being allocated consecutively taking into account structure surfaces temperatures. In this case, vitrified HLW saturation time is estimated to be limited to 500 years.

Filtration properties of concrete materials used in the construction of near-surface storage facilities and relevant dependences

Filtration and diffusion properties of concrete mostly depend on its porosity, which, in turn, depends on the composition of the mineral phases constituting to the concrete. A dependence between filtration (K_f) and diffusion (K_d) coefficients and concrete porosity is described by numerical methods:

$$K_f = c_f \theta^m, \quad (3)$$

where: c_f is the coefficient associated with the resistance of the porous medium (a value of $5,75 \cdot 10^{-11}$ m/s was derived for cement paste based on experimental data); θ stands for the concrete porosity, relative units; m stands for the cementing exponent (a value of 2.5 was assumed for the studied grade M400 concrete).

$$K_d = \theta^{-F} + K_0, \quad (4)$$

where θ stands for the concrete porosity, relative units; F is the coefficient associated with the resistance of the porous medium (a value of 2 was assumed for studied grade M400 concrete); K_0 is the diffusion coefficient for non-degraded concrete ($5 \cdot 10^{-10}$ m²/s).

Based on the estimated changes in concrete porosity, the dynamics of filtration and diffusion coefficient dependences was shown.

Model showing the effects of steel barrier corrosion on bentonite buffer properties for NKM DDF RW

The impact of corrosion products on bentonite was estimated assuming the conditions being considered characteristic for the developed deep disposal facility and taking into account the temperature dynamics in the modeling region shown in Figure 3.

The calculations specified the changes occurring in bentonite upon its interaction with a corroding steel barrier under an evolving temperature field. The model included a kinetic unit presenting steel corrosion with an account taken of its surface inhibition by the resulting corrosion products, as well as a chemical processing unit taking into account sorption along with formation and dissolution of mineral phases. Calculations were performed using the calculation code PhreeQC 2.18 integrated into CC DESTRUCT [3].

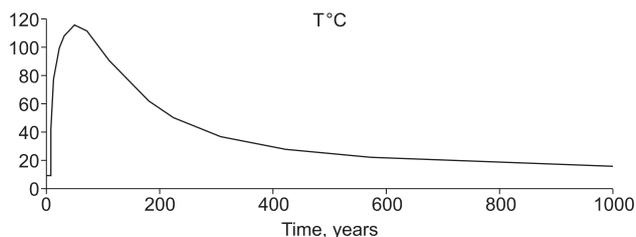


Figure 3. Modeling the time dependence of bentonite barrier temperature

Test calculations aimed at estimating the amount of solid mineral phases in Khakass bentonite over a period of 1,000 years have shown that the content of the essential bentonite component, montmorillonite, remained practically the same with some slight decrease being observed only in its contact zone with the steel corroding barrier. This zone is also characterized by some increased content of the corrosion product, namely, iron hydroxide, whereas chlorite content increases slightly resulting in a decreasing bentonite porosity and, therefore, a decreasing diffusion coefficient.

Sodium-aluminum-phosphate glass leaching model for HLW under NKM DDF disposal conditions

Accounting of actually occurring processes and leaching kinetics seems to be a most optimal approach to leaching assessment that should be followed when it comes to the evaluation of radionuclides constituting to the glass matrix. These are considered as individual mineral phases of oxides, silicates, phosphates, borosilicates with their amount being recalculated based on the total activity of the matrix. The leaching constant and the composition of water contained in the materials of the EBS cascade should be identified taking into account the influence of the processes, as well as the temperature and characteristics of the solution.

A case study with leaching calculations for a sodium aluminum phosphate glass matrix interacting with bentonite is provided below as an example. Unconformity of solution composition with the one the soluble solid phase is believed to be a distinctive feature of glass dissolution which is explained by the formation of solid hydrolysis products of the glass matrix, precipitation of poorly soluble secondary phase compounds (incongruent dissolution).

The calculations were performed using the calculation code Phreeqc 2.18 integrated into CC DESTRUCT [3].

According to model assumptions, aluminum phosphate was considered as a leading phase in the dissolving glass. It was assumed that under the first stage of glass dissolution, its components will dissolve congruently. Then, solution composition was

recalculated based on phase precipitation (leaching products) supersaturating the solution. Thus, the incongruent nature of glass dissolution (leaching) has manifested itself.

An equation was applied in practice to simulate glass matrix dissolution [4] suggesting that the dissolution kinetics depended on solution saturation considering the leading phase of the glass matrix dissolution that could be put down as follows:

$$R_i = S \cdot k \cdot e^{-\frac{E_a}{RT}} \cdot \left(1 - \frac{IAP}{K_s}\right), \quad (5)$$

where S is the interface area with the solution, m^2 ; k is the real reaction rate constant; E_a is the activation energy, J/mol ; R is the universal gas constant, $J/(mol \cdot K)$; T is the absolute temperature; IAP/K_s is the ratio between current ion concentrations being responsible for the phase dissolution and the saturation concentrations. Equation (5) was used as a basis for the developed leaching models. Two models were applied in the calculations. The first one was based on equation (5), the second one was distinguished by the consideration given to the inhibited glass corrosion driven by leaching product formation on its surface based on the following dependence:

$$R_i = S \cdot k \cdot k_T \cdot e^{-\frac{E_a}{RT}} \cdot \left(1 - \frac{IAP}{K_s}\right), \quad (6)$$

where k_T is the inhibition function being set by the following empirical equation:

$$k_T = 1 / (1 + k_1 \cdot \sum_i EQUI_i ("Corrosion product")^{(k_2)}), \quad (7)$$

where k_{1-2} stands for the approximating coefficients; $EQUI_i$ ("Corrosion Product") is the amount of the formed i -th corrosion product, mol/l .

Figure 4 presents the results of model and calculated data comparison for sodium (Na), phosphorus

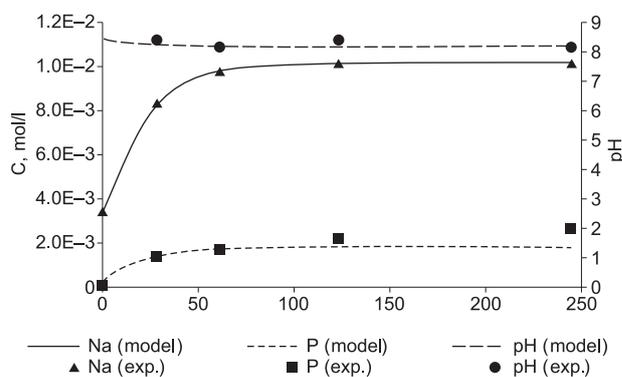


Figure 4. Comparison of model and calculated data on Na, P formation and pH in the solution for an aluminum phosphate matrix

(P) and relevant pH values (a more detailed discussion of the model is presented in [5]).

A number of problems being considered topical in terms of the safety analysis is provided below to exemplify the application of the software package in addressing relevant comprehensive tasks.

Calculations of radionuclide releases beyond the boundaries of RW tailings

The first case study was focused on an SRW storage facility (tailing). The calculations were aimed at evaluating the effectiveness of the safety barrier system.

The following scenario was adopted under the simulations performed:

1. A constant amount of seepage leaks passing through the engineered barrier layer and reaching the aquifer was assumed for the tailing since its commissioning time with a due account taken of the available impervious covering screen.

2. Having passed through the aeration zone, the radionuclides are released into the aquifer, where they further migrate to the groundwater discharge area.

Within the chamber migration model implemented in DESTRUCT, contaminant transfer between the chambers is set via transfer coefficients being expressed as a fraction of the substance migrating from one chamber to another per unit time. Basic mathematical formulas describing such inter-chamber transport are presented in [6]. To predict radionuclide spread through a barrier being assumed as a loam layer virtually remaining unchanged for many thousands of years, a chamber model describing contaminant migration in an unsaturated zone bounded by an underlying loam layer is believed to be sufficient.

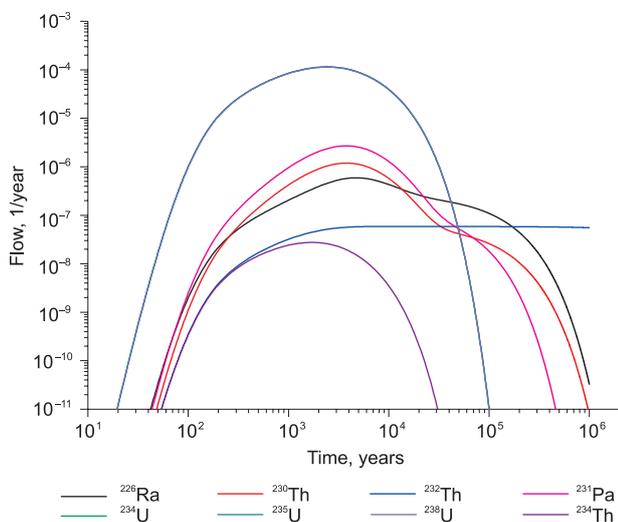


Figure 5. Calculated radionuclide release outside the engineered safety barrier system of SRW storage facility (year⁻¹)

Radionuclide migration into the water-saturated area was calculated with an account taken of the decay chains through the loam layer being considered as a natural barrier.

Figure 5 presents the integrated radionuclide release during their migration through the confinement barriers, normalized to the initial activity of ²³⁴Th, ²³²Th, ²³⁰Th, ²²⁶Ra, ²³¹Pa, ²³⁴U, ²³⁵U, ²³⁸U.

Activity release will reach its peak in ~ 10 000 years evidencing the effectiveness of the underlying loam barrier for the SRW storage facility.

Calculated radionuclide release from DDF RW class 1 package

DDF RW is seen as another example of an urgent task associated with safety analysis with relevant estimates of radionuclide releases from RW class 1 matrices beyond the multibarrier safety system being considered essential.

Steel containers containing drums with HLW enclosed into phosphate glass matrix are proposed as RW packages for RW Class 1 disposal. It was assumed that following “shock” steel container corrosion destruction after 1,000 years (containment preservation time for RW Class 1 packages in accordance with NP-093-14), the dissolution rate for solidified RW Class 1 will amount to 1·10⁻⁵ g/cm²/day (according to the release rate of ¹³⁷Cs from the glassy compound in accordance with NP-019-2000). Based on the developed IM, radionuclide flow escaping the disposal system was normalized to the initial activity (Figure 6).

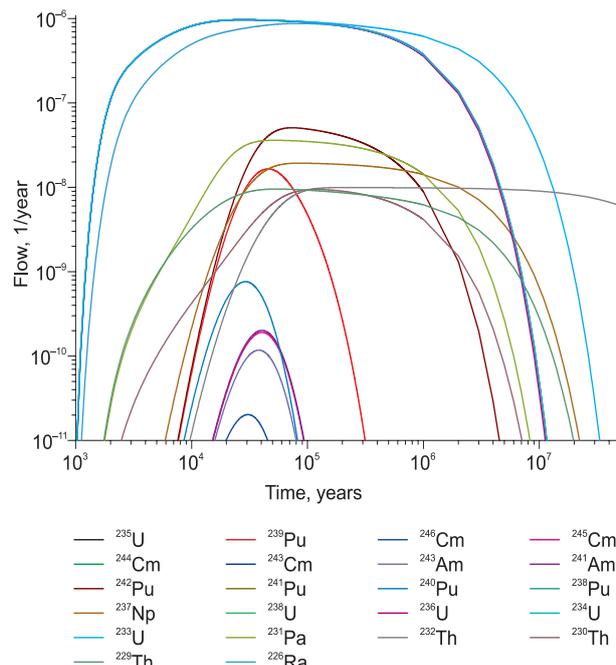


Figure 6. Calculated radionuclide flow escaping the boundaries of DDF RW (year⁻¹)

Calculated radionuclide release beyond a near-surface disposal facility

Near-surface disposal facility (NSDF) was considered as another case study that enabled further refinement of the methods applied to estimate the release of fissile materials.

A 300 years-long time-period was assumed for the concrete barrier during which its uniform degradation will occur culminated by the loss of its waterproofing capacity. Degradation of the upper concrete slab described by a linear function of time was considered in the forecasts of radionuclide releases from the NSDF. Under these calculations, an additional factor was introduced to the seepage value $F(t)$ varying in time from 0 at the beginning to 1 after 300 years, i. e., $I = I_0 \cdot F(t)$.

Figure 7 shows the release of fission products normalized to the initial activity with the peak release (up to $1 \cdot 10^{-5}$ % of the initial content) from a near-surface storage facility corresponding to the range from 10,000 to 1,000,000 years.

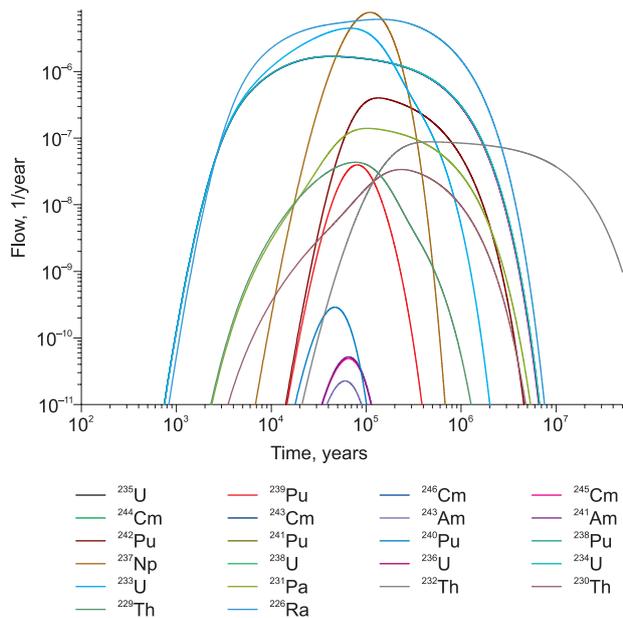


Figure 7. Calculated radionuclide flow escaping NSDF boundaries (year^{-1})

Comprehensive calculation of radionuclide releases from an RW container

The developed software was also applied to evaluate the rate of radionuclide releases from reinforced concrete containers which seems to be quite helpful when it comes to assessing the long-term safety of stored materials being exposed to some external influences.

The scenario assumed that the radionuclides having escaped the source term are released

immediately into a flooded area of the storage facility subsequently spreading within the bedrocks.

A numerical model was developed enabling to identify relevant releases normalized to the initial activity. Figure 8 presents the calculated radionuclide releases normalized to the initial activity.

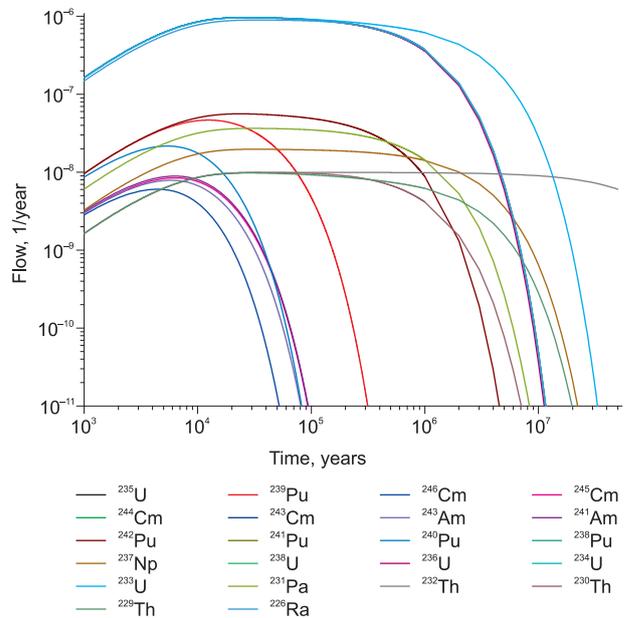


Figure 8. Calculated radionuclide flow through safety barriers (year^{-1})

Conclusion

The article sets out the principles and foundations of an approach to an integrated description of changes in the properties of RW DF engineered safety barriers and methods allowing to consider the impact of these processes on the long-term safety. It presents the main mechanisms resulting in the degradation of these barriers including waste immobilization matrix, packaging materials and basic structural materials used in RW DF construction. It proposes an approach which is based on a comprehensive consideration of the entire EBS evolution under the mutual influence of changes in the properties of each barrier.

The paper discusses currently available capacities of the DESTRUCT computational complex with its modules being integrated to predict the dynamics of evolving EBS properties within a single information environment. The developed tools providing integrated accounting of EBS evolution within the assessments of radionuclide releases beyond facility boundaries enabled more accurate description of the considered mechanisms, the identification of the essential ones, evaluation of their mutual influence and formation of a multiphysical complex

model accounting for the mutual interaction of various calculation tools.

To exemplify relevant practical applications, some results associated with the safety analysis of various common type facilities were presented. Findings of the studies involving the developed approach will reduce the conservatism of the calculations performed to evaluate the spread of radionuclides beyond the engineered barrier system and will provide more accurate estimates of their potential impact on the disposal safety.

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Information about the authors

Kryuchkov Dmitry Vyacheslavovich, PhD, Chief of Laboratory, The Nuclear Safety Institute of RAS (52, Bolshaya Tulsкая str., Moscow, 115191, Russia), e-mail: dvk@ibrae.ac.ru.

Boldyrev Kirill Aleksandrovich, PhD, Senior Researcher, Nuclear Safety Institute of RAS (52, Bolshaya Tulsкая St., Moscow, 115191, Russia), e-mail: kaboldyrev@ibrae.ac.ru.

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