

MODELLING OF RADIONUCLIDE RELEASE FROM ALUMINIUM PHOSPHATE GLASS MATRIX CONSIDERING CHEMICAL PROCESSES

Boldyrev K. A., Bogatov S. A.

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

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The article investigates the mechanisms responsible for aluminum phosphate glass matrix (APM) leaching and quantifies the corresponding release of radionuclides beyond the system of engineered barriers of the deep geological repository (DGR) for radioactive waste. The paper considers a bentonite clay-based material as a buffer material. The leaching model of the APM was developed on the basis of an experimental study with account taken of the kinetics of leaching, surface blocking made by secondary solid mineral phases of the interaction of leached glass matrix with water and contacting bentonite.

Keywords: migration of radionuclides, modeling, radioactive waste, leaching, bentonite, DGR, PhreeqC.

Introduction

The Federal Target Program Nuclear and Radiation Safety for 2016–2030 (FTP NRS-2) states that high-level waste (HLW) is subject to final disposal in a deep geological repository (DGR) to be constructed at the Yeniseiskiy site in the southern part of the Krasnoyarsk Territory. The disposal safety is provided via the multi-barrier principle (NP-055-14 [1] and others) suggesting that a cascade of engineered and technical barriers is installed to prevent the spread of ionizing radiation and radioactive substances into the environment (Fig. 1).

Since 1987, the USSR and later the Russian Federation have been adhering to the practice of industrial liquid HLW immobilization into aluminum phosphate glass (APG) matrix (waste form) [2]. The corresponding disposal concept provides for waste solidification using glass-like matrices given their subsequent emplacement into thick-walled steel containers. The gaps between their outer walls and the surrounding host rock are proposed to be filled with a buffer material. Bentonite clay characterized by certain unique properties (high swelling capacity,

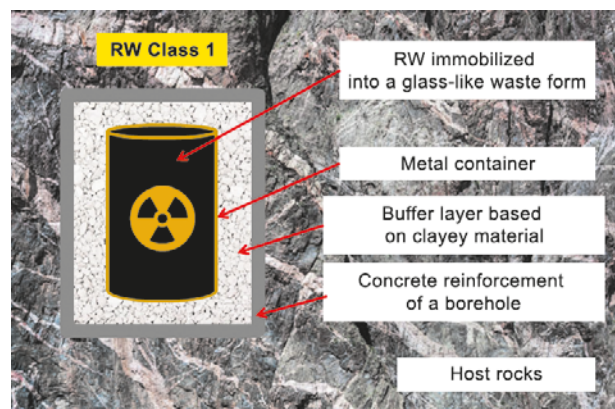


Fig. 1. Conceptual design of a disposal cell for HLW

low water permeability, self-sealing of cracks and effectively absorption of a few radionuclides [3]) is considered as an appropriate backfilling material.

Under long-term safety assessments, radionuclide releases from a term source, their spread through the existing safety barriers with due account of their evolution shall be forecasted and the

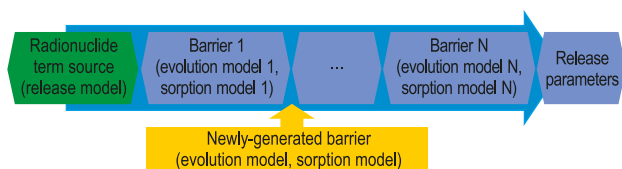


Fig. 2. Schematic model showing the movement of radionuclides through a cascade of evolving safety barriers

expected public exposure levels shall be compared against the regulatory standards. According to NP-055-14 [1] and RB-003-21 [4] provisions, mathematical models shall be used for these purposes. Fig. 2 shows schematically the release of radioactive substances from their term source through a sequence of safety barriers in a DGR [5] with the model of APG leaching due to its interaction with bentonite viewed as its integral part.

This paper presents an approach to the quantitative assessment of APG leaching and its dynamics taking into account the accumulation of chemical reaction products in the leaching zone and the influence of these processes on radionuclide releases from the EBS system. Previously developed models of APG leaching due to its interaction with bentonite [6], [7] served a basis for those considered in this paper.

Approaches proposed to model the leaching of a glass matrix and its interaction with bentonite with an account taken of ongoing chemical processes

The time of laboratory experiments appears to be much shorter than the time scales specific to the processes evolving in a DGR, which requires the radionuclide releases to be estimated on large time scales.

In other countries, leaching processes in glass matrices were mainly simulated for borosilicate glasses mainly proposed as a material for liquid HLW solidification (for example, [8]–[13] and others).

The simplest and most conservative method applied to calculate the radionuclide release rates is based on the assumption regarding constant relative matrix dissolution rate, λ , taken in⁻¹. In this model, the rate, at which the mass of the glass matrix (dissolution) changes, dm/dt , is proportional to its current mass, m , at a time, t :

$$\frac{dm}{dt} = -\lambda m, \quad (1)$$

and λ is determined based on experimental leaching studies.

It is known that its rate decreases considerably with time, which is mainly due to the accumulation

of glass matrix corrosion products on the reacting surface [7], [14].

APG matrix leaching model

In this study, the following dependence was used to describe the leaching rate of the APG matrix at each moment in time, R_{leach} , mol/(s·kg) [6], [7]:

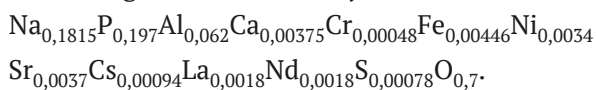
$$R_{\text{leach}} = \frac{k_{\text{leach}} \cdot S \cdot [\text{H}^+]^n \cdot e^{-\frac{E_A}{RT}}}{\left[1 + k_1 \sum_i \text{EQUI}_i(\text{SSMP})\right]^{(k_2)}} \left(1 - \frac{\text{PAI}}{\text{PR}}\right). \quad (2)$$

Where:

- k_{leach} is the leaching rate constant, mol/(m²·s);
- S is the specific surface area of the glass matrix interacting with bentonite (geometric) (m²);
- $[\text{H}^+]$ – proton activity, mol or mol/l;
- E_A – activation energy, J/mol;
- R is the universal gas constant, J/(mol·K);
- T is the temperature, K;
- PAI is the current product of ion activities in the phase governing the dissolution of the glass matrix (aluminum phosphate, AlPO_4 , was chosen for the APG matrix);
- PR is the solubility product of the phase governing the dissolution of the glass matrix;
- n is the exponent;
- $\text{EQUI}_i(\text{SSMP})$ is the amount of the i -th secondary solid mineral phase formed on the glass surface, mol/l.

Formula (2) demonstrates that in addition to the contact area, the dissolution rate depends on the following three factors: chemical composition of the solution (including the proton concentration), the distance from equilibrium (saturation in the phase controlling dissolution) – $(1 - \text{PAI}/\text{PR})$ and the presence of a layer with newly generated products of the glass matrix corrosion.

The model representing the release of glass components corresponds to [6]. It was assumed that 1 mole of glass is described by the below formula:



In the model, during the leaching process, all glass matrix components are released into the solution at the same rate (congruent matrix dissolution), which is adjusted after each time step taking into account the processes of ion form distribution in the solution: secondary phase formation, sorption. Thus, the incongruent nature of the glass matrix dissolution is implemented.

Model showing the evolving mineral composition of bentonite phases

Modeling takes into account the processes important in terms of their influence on the pore

water and its composition with the principal ones that account for the transformation of bentonite and, in particular, of its solid mineral phases (smectite and accessory minerals), gas exchange with the atmosphere (in case of interaction), sorption (ion exchange and complexation on the surface). The model is parameterized according to the following sequence:

- 1) the initial composition of bentonite pore water is specified;
- 2) the areas (m²/g) of mineral phases and kinetic parameters of dissolution are set;
- 3) external conditions similar to the modeled study are adopted.

Experimental data on the bentonite phase evolution in terms of its mineral composition were adopted according to another study [16] focused on the effect produced by concrete leaches at a temperature of 90 °C. The model took into account the dissolution kinetics of smectite, kaolinite, illite, quartz, albite, chlorite, potassium feldspar (in this case, microcline), calcite. In addition, ion exchange and complexation at sorption centers were monitored. Calculations were performed in the PhreeqC geochemical modeling code [17], the *llnl.dat* thermodynamic database was used, and the following equation was applied to calculate the dissolution of mineral phases ([18] describes the parameters of the transformation kinetics (rate) specific of clay phases based on a few literature sources):

$$R_j = S_j \cdot \left(k_{j25}^{\text{acid}} e^{\left[\frac{-E_{jA}^{\text{acid}}}{R} \left(\frac{1}{T} - \frac{1}{298.15} \right) \right]} [\text{H}^+]^{n_j^{\text{H}^+}} + k_{j25}^{\text{neutr}} e^{\left[\frac{-E_{jA}^{\text{neutr}}}{R} \left(\frac{1}{T} - \frac{1}{298.15} \right) \right]} + k_{j25}^{\text{alkali}} e^{\left[\frac{-E_{jA}^{\text{alkali}}}{R} \left(\frac{1}{T} - \frac{1}{298.15} \right) \right]} [\text{OH}^-]^{n_j^{\text{OH}^-}} \right) \cdot \left(1 - \frac{\text{PAI}_j}{\text{PR}_j} \right) \quad (3)$$

where R_j is the mass of the j -th solid phase dissolved per unit time (dissolution rate), mol·s⁻¹/dm³; k_{j25} is the kinetic constant for the dissolution or precipitation rate of the j -th mineral phase at 298.15 K (25 °C), mol/m²·s;

E_{jA} is the activation energy for the j -th mineral phase, J/mol;

S_j is the reaction surface area for the j -th mineral phase, m²;

neutr – corresponds to neutral conditions, acid – to acidic conditions, alkali – to alkaline conditions; n_j is the exponent.

Combined model of APG leaching and transformation of bentonite solid mineral phases

Fig. 3 presents a diagram showing the relationship between the processes occurring during the leaching process assuming the interaction with bentonite. Kinetic parameters of the glass matrix dissolution were determined according to an experimental study [6]: the corresponding indicators adopted for bentonite from the 10th Khutor deposit are given in Table 1.

Simulation results

Simulation of APG matrix leaching assuming its contact with bentonite based on an experimental study

Fig. 4 compares the simulation data reflecting the changes in the content of solid mineral phases of bentonite and the experimental data, whereas Fig. 5 compares the results calculated given the APG matrix leaching assuming its interaction with bentonite in different models. The predicted curves showing the yield of the matrix macrocomponents, sodium and phosphorus, practically reach a plateau. The exchange sorption mass of bentonite and calcite serves as a calcium source, whereas the glass matrix accounts for the source of phosphate ions. The processes accounting for the precipitation of gibbsite, calcium phosphates and other cations, oxyhydroxide phases, as well as the degree to which

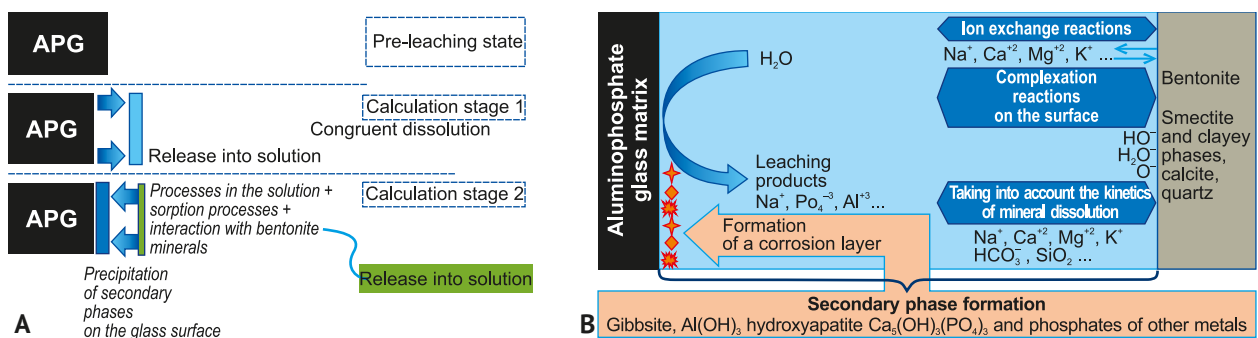


Fig. 3. Conceptual representation of incongruent glass matrix dissolution in the model (A). Relationship between the processes occurring during APG matrix leaching assuming its interaction with bentonite given a corrosion layer formed on the surface of the glass matrix (B)

Table 1. Parameterization of some terms of the equation (3) in a model demonstrating the clay material evolution for bentonite from the 10th Khutor deposit. Kinetic constants (k) are expressed in mol·m⁻²·s⁻¹ and activation energy (E_A) is expressed in k_J·mol⁻¹

Mineral	S, m ² /g	k ₂₅ ^{neutr}	E _{JA} ^{neutr}	k ₂₅ ^{acid}	E _{JA} ^{acid}	η _J ^{H+}	k ₂₅ ^{alk}	E _{JA} ^{alk}	η _J ^{OH-}	k _{pre}
Albite	2	5.1·10 ⁻²⁰	57	8.5·10 ⁻¹¹	58	0.34	1.4·10 ⁻¹⁰	56	0.32	2.15·10 ⁻¹³
Illite	30	3.3·10 ⁻¹⁷	34.6	9.8·10 ⁻¹²	36	0.52	3.1·10 ⁻¹²	48	0.38	2.15·10 ⁻¹⁵
Potassium feldspar	1	1.0·10 ⁻¹⁴	31	1.7·10 ⁻¹¹	43	0.27	1.4·10 ⁻¹¹	31	0.25	2.15·10 ⁻¹³
Kaolinite	5	1.1·10 ⁻¹⁴	38	7.5·10 ⁻¹²	43	0.51	1.25·10 ⁻¹¹	46	0.58	2.15·10 ⁻¹⁴
Quartz	0.03	6.4·10 ⁻¹⁴	76.7				1.92·10 ⁻¹⁰	80	0.34	2.15·10 ⁻¹³
Smectite	40	9.3·10 ⁻¹⁵	63	5.3·10 ⁻¹¹	54	0.69	2.9·10 ⁻¹²	61	0.34	7.59·10 ⁻¹³
OChlorite	1	6.4·10 ⁻¹⁷	16.4	8.2·10 ⁻⁹	17	0.28	6.9·10 ⁻⁹	16	0.34	1.2·10 ⁻¹²
Calcite	0.7	1.55·10 ⁻⁶	14.4	0.5	14.4	1				1.9·10 ⁻³

Glass matrix						
	S _{cont} , m ²	k ₂₅ ^{neutr}	E _A	η _J ^{H+}	k ₁	k ₂
Model taking into account the kinetics of solid bentonite mineral phase transitions						
Glass matrix	0.4	7.9·10 ⁻⁵	17	-0.4	3,000	3.12
Model not taking into account the kinetics of solid bentonite mineral phase transitions						
Glass matrix	0.4	1.4·10 ⁻⁶	-17	-0.4	2,000	2

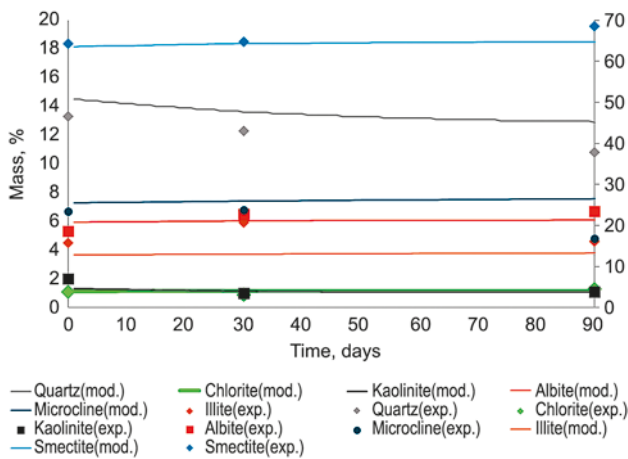


Fig. 4. Comparison of modeling data reflecting the changes in the content of solid mineral phases for bentonite from the 10th Khutor deposit (for smectite, the axis on the right) according to the experimental study discussed in [16]

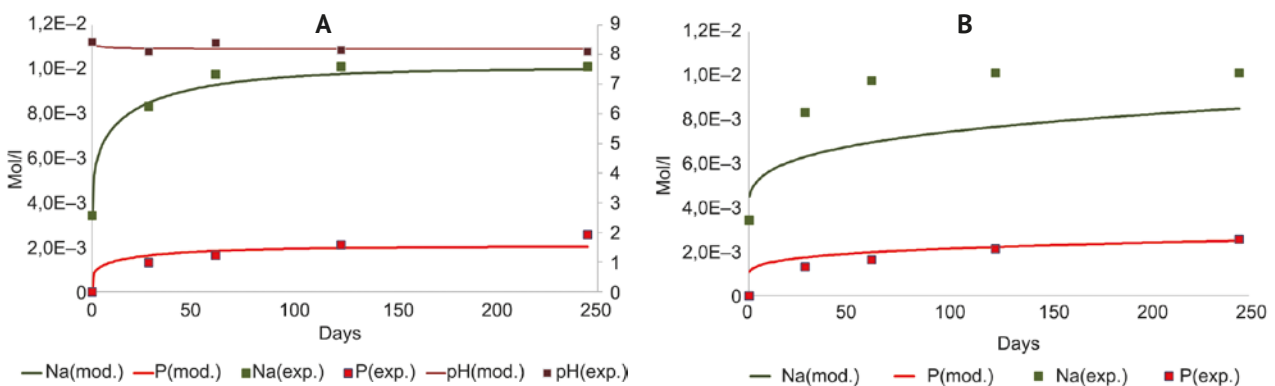


Fig. 5. Comparison of leaching modeling results for an APG matrix interacting with bentonite in different models: A – no consideration given to the kinetics of the bentonite phase transformations, B – accounting the kinetics of bentonite phase transformations

the solution saturates with aluminum phosphate actually govern the glass matrix dissolution process. The model built with no account of the kinetics associated with the transformation of solid mineral bentonite phases (Fig. 5A) demonstrates a more accurate correspondence between the calculated and experimental data in the sodium concentration area, whereas the model built with an account taken of such kinetics (Fig. 5B) shows a better correspondence in terms of phosphorus concentration.

Glass matrix leaching assuming its interaction with bentonite modeled over a time period of 500 years

The evolution of bentonite phases and the pore solution assuming its interaction with the APG matrix was calculated. The model column with water-saturated bentonite was divided into 16 cells of

2.5 cm each. Leaching was assumed in the first of them interacting with the glass matrix.

The matrix material is known to accumulate microdefects during its cooling and due to its internal irradiation, which may result in the evolution of crystallization centers inside the glass and its cracking (see review [19]), as well as an expansion of the APG surface area by up to 40 times [20], thereby, resulting in larger area of interface with water and, accordingly, an accelerated release of glass matrix components into the solution. In the model, it was assumed that the contact area grew up by 40 times compared to the geometric one to account for the cracking effect. In the calculated yield of matrix components, the effective diffusion coefficient for watered bentonite was taken equal to $3 \cdot 10^{-11}$ m²/s. When leaching processes were modeled assuming more ample consideration of the chemical processes, the following two types of simulations were performed:

1. The dissolution kinetics of glass matrix, as well as calcite contained in the bentonite buffer were considered according to the thermodynamic law with no account taken of the transition kinetics as regards other bentonite phases.

2. The leaching kinetics as regards both the glass matrix and the main bentonite phases were taken into account (for the first cell interacting with the APG matrix — for all phases, for the remaining 15 — for calcite only).

Fig. 6 summarizes the simulation results. A decrease in the leaching rate over time appears to

be quite noticeable. The total masses of the glass matrix transformed (dissolved) over 500 years accounted for (per 100 cm² of its geometric surface):

- simulation 1 : 26.3 g;
- simulation 2 : 9.82 g.

The equations reflecting the dependence of the glass matrix mass transformation over time (per 100 cm² of its geometric surface) shown in Fig. 6A were obtained, which were then extrapolated to longer periods (Fig. 6B).

For the model built with no account taken of the kinetics, the dependence of the dissolved APG mass, m (grams), on time t (years) can be calculated as follows:

$$m = 1,3597t^{-0.703}, \quad (4)$$

for the model taking into account the kinetics of bentonite phase transformation it can be calculated as follows:

$$m = 0,6342t^{-0.712}. \quad (5)$$

Forecasting radionuclide transfer in the models of leaching from the APG matrix

The obtained data were used in the current model to assess the DGR safety with an account taken of radionuclide releases from vitrified HLW, their penetration through the EBS and into the near field of the repository, as well as their distribution in the geosphere and biosphere providing an ultimate assessment of the radiation impact on critical groups of population. The analysis was performed to compare the impacts produced by different assumptions regarding the rate of radionuclide releases from the APG matrix of vitrified HLW on the results obtained. In these calculations, the protective properties of the container were not taken into account and the release of radionuclides from vitrified HLW was expected to occur at the initial moment of time. The basic model assumes a constant relative matrix dissolution rate $\lambda = 1.34 \cdot 10^{-4}$ year⁻¹, which was calculated based on short-term (several months) experiments when model APG samples were dissolved and the assumed 10-fold increase in their surface area due to cracking. In alternative models, the APG leaching was modeled numerically based on the above calculations.

The developed model assumed the radionuclide releases to occur in the water layer corresponding to the volume of the unfilled container capacity. Then, their distribution was described by diffusion through a 36-cm-thick cylindrical bentonite layer, where the solubility limits for the considered radionuclides as regards the solid mineral phases of individual radionuclides and their sorption on bentonite were taken into account assuming constant

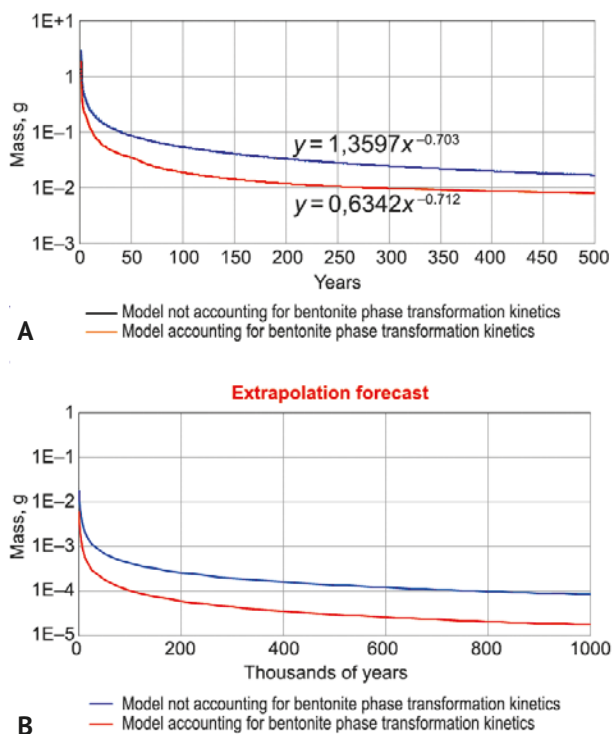


Fig. 6. Dependence between the mass of the dissolved APG matrix and time: A – modeling results and B – their extrapolation to 1 million years

interphase distribution coefficients, K_d . Table 2 presents some parameters for the radionuclides considered in this version of the model. It should be noted that the corrosion properties of the container were not taken into account in the model for the following reasons: on the one hand, the corrosion products of the iron constituting to the steel container have quite notable sorption-precipitation properties with respect to a number of radionuclides, especially with respect to TUE and REE; on the other hand, accounting of additional sorption would lead to an even lower rate of radionuclide release during APG leaching. The authors were aimed at obtaining conservative estimates for the purposes of the safety assessment.

Table 2. Parameters governing radionuclide activity in the glass matrix, threshold solubility levels for their phases and K_d in bentonite buffer

Nuclide	Specific activity, Bq/kg (vitrified HLW)	Threshold solubility levels for individual phases of radionuclides in the pore water, mol/m ³	K_d , m ³ /kg
⁷⁹ Se	1.3·10 ⁷	5·10 ⁻⁶	0
⁹⁹ Tc	1.4·10 ⁷	4·10 ⁻⁶	63
¹²⁹ I*	2·10 ⁴	10 ⁻⁵	0
¹³⁵ Cs	1.1·10 ⁷	∞	0.5
²³⁸ U	5.3·10 ⁴	4·10 ⁻⁶	63
²³⁴ U**	0	4·10 ⁻⁶	63
²³⁰ Th**	0	7·10 ⁻⁴	63
²²⁶ Ra**	0	2·10 ⁻⁸	0.045

*¹²⁹I is assumed to be present in the APG matrix in the form of AgI (silver nuclei have a significant yield during fission, and this compound is viewed as a most likely one in case if iodine is present in the vitrified HLW [23]).

**Daughter radionuclides of ²³⁸U.

Fig. 7 shows the time dependences as regards the fraction of the initial APG matrix mass dissolved in the layer of surrounding water assuming different leaching rates. In the basic version, which includes a conservative source model, over 90% of the APG matrix mass may pass into the solution over the entire modeling period. When glass matrix dissolution is modeled with an account taken of chemical reactions and secondary phase formation, fractions of a percent, i.e., percent of its mass, end up in the leachate. For comparison, Fig. 7 also shows a curve corresponding to a constant linear rate of APG matrix dissolution (0.14 μm/year) obtained from analogous objects [21] (note that this indicator is close to the leaching rate of borosilicate glass K-26 under unsaturated conditions — 0.1 μm/year [22]). In the callouts, the curves in Fig. 7 show the maximum expected annual doses to the critical group of population under the considered model version.

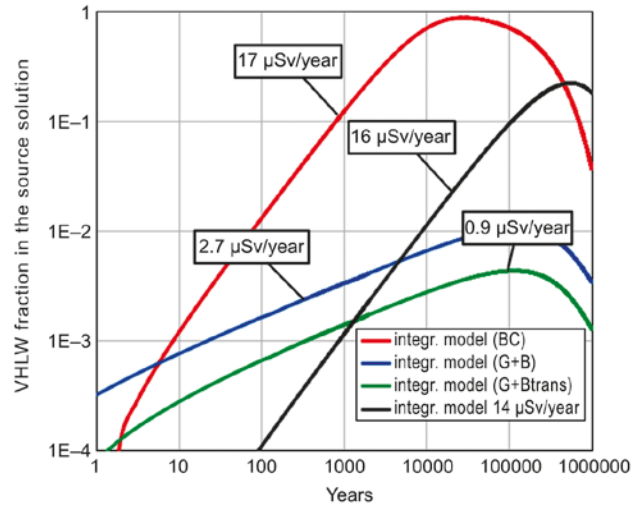


Fig. 7. Dependencies of the initial APG matrix mass fraction in the source dissolution zone: BC – base case; G+B – without taking into account the kinetics of bentonite transformations; G+Btrans – taking into account the kinetics and extrapolation over a long time period; 0.14 μm/year – constant rate of APG matrix dissolution for analogous objects [21].

The callouts show the expected maximum annual doses to adult population

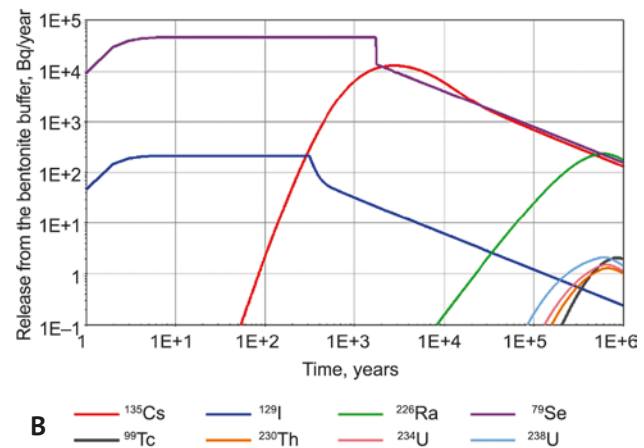
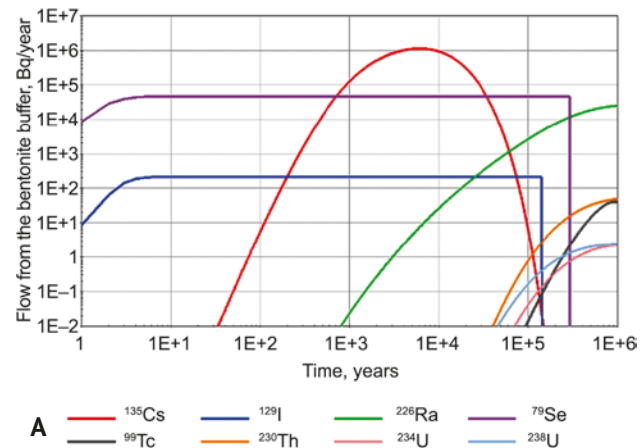


Fig. 8. Comparison of radionuclide flows through a bentonite barrier for the basic option – A and a more realistic one taking into account the interaction of the APG matrix leaches with bentonite – B

Fig. 8 shows the flows of the considered radionuclides beyond the bentonite barrier in the integral model, i. e., for its basic version, and in the model taking into account the chemical processes and kinetics of the bentonite phase transformations (Simulation 2). It is evident that the isotope flows decrease compared to the basic option.

It is interesting to consider the release of phosphorus due to its biotic activity, which is important for the assessment of biogenic processes [23] and the precipitation of phosphate phases in the bentonite massif and on the crack material, i. e., it can be noted that its release slows down as well (Fig. 9). Increased concentrations of phosphorus and sodium were predicted in the early stages – up

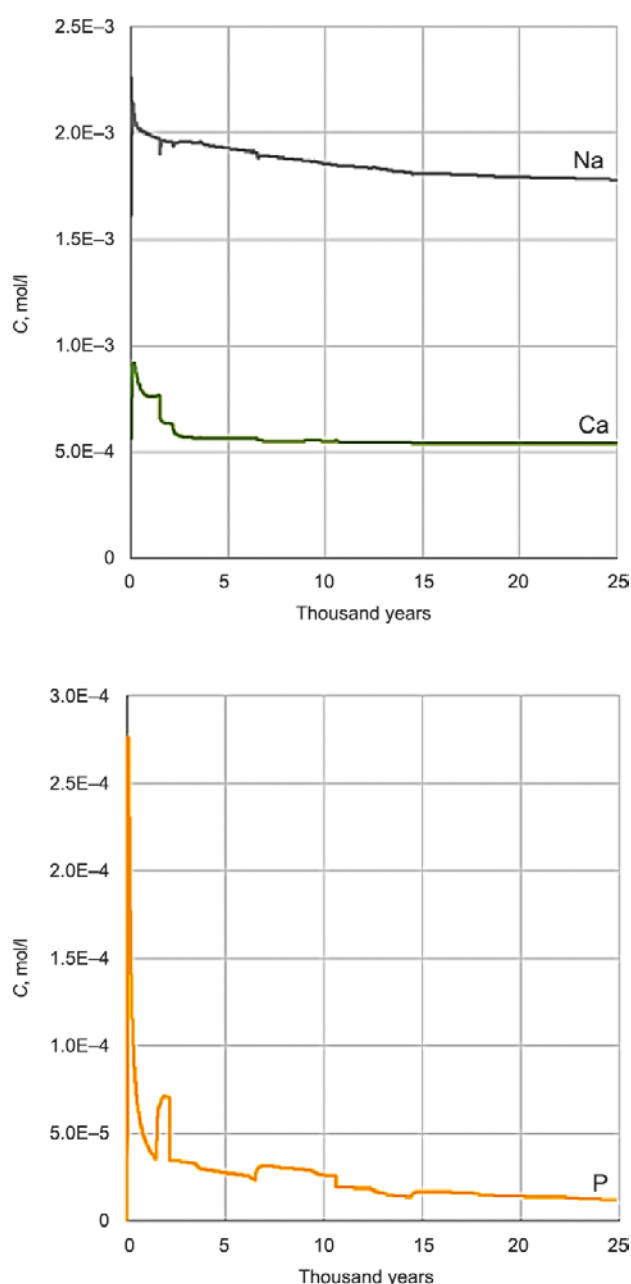


Fig. 9. Calculated concentrations of phosphorus, sodium and calcium in water at the outer boundary of the bentonite barrier

to 1,000 years. It is during this period of time that stimulation of biotic activity by these elements is expected.

Discussion / Conclusions

This paper explores the effect of chemical processes on the leaching rate of the APG matrix of vitrified RW assuming its interaction with bentonite based on literary data and field experiments. The thermodynamic model implemented using the PhreeqC geochemical modeling code was used to account for the effects produced by the chemical composition of the solution interacting with the APG matrix, saturation with respect to the phase governing its dissolution (aluminum phosphate, AlPO_4 , was selected) and the presence of precipitated secondary phases on the glass matrix leaching rate. It should be noted that the formation of new secondary phases is supposed to change not only the properties of the contacting bentonite, including its swelling capacity and water conductivity, but will also cause the capture of radionuclides leached from the matrix during (co)precipitation, which may drive their additional fixation. Preliminary results show that consideration of chemical processes occurring along with APG matrix leaching in the model may considerably decrease both the rate and the fraction of radionuclides released from the engineered barrier system and the potential radiation impact produced by the DGR on the population by more than an order of magnitude compared to another model not taking these chemical processes into account. Great potential impact of the considered effects on the findings of the final safety assessments indicates that model assessments should be continued with more target experiments to explore the leaching of RW matrices interacting with bentonite.

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Information about the authors

Boldyrev Kirill Aleksandrovich, Candidate of Technical Sciences, Senior Researcher, Nuclear Safety Institute of the Russian Academy of Sciences (52, Bolshaya Tulkaya st., Moscow, 115191, Russia), e-mail: kaboldyrev@ibrae.ac.ru.

Bogatov Sergey Aleksandrovich, Candidate of Physical and Mathematical Sciences, Leading Researcher, Nuclear Safety Institute of the Russian Academy of Sciences (52, Bolshaya Tulkaya st., Moscow, 115191, Russia), e-mail: sbg@ibrae.ac.ru.

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