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Conceptual Model to Determine Maximum Activity of Radioactive Waste in Near-Surface Disposal Facilities

For development of the management strategy for radioactive waste (RW) to be placed in near-surface disposal facilities (NSDF), it is necessary to justify long-term safety of such facilities. Use of mathematical modelling methods for long-term forecasts of RW radiation impacts and assessment of radiation risks from radionuclides migration can help to resolve this issue.

The purpose of the research was to develop the conceptual model for determining the maximum activity of RW to be safely disposed in the NSDF and to test it in the case of Lot 3 Vector NSDF (Chernobyl exclusion zone). This paper describes an approach to the development of such a model. The conceptual model of ^{90}Sr migration from Lot 3 through aeration zone and aquifer soils was developed. The results of modelling are shown. The proposals on further steps for the model improvement were developed.

Keywords: conceptual model, radioactive waste, near-surface disposal facilities, ^{90}Sr, migration, radionuclide distribution coefficient.

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Концептуальна модель визначення максимальної активності радіоактивних відходів у приповерхневих сховищах для захоронення

Для розробки стратегії поводження з радіоактивними відходами (РАВ), які планується розміщувати в приповерхневих сховищах для захоронення, необхідне обґрунтування довгострокової безпеки таких сховищ. Це можна зробити за допомогою методів математичного моделювання для довгострокових прогнозів радіаційних впливів РАВ та оцінки радіаційних ризиків від міграції радіонуклідів.

У статті описано підхід до розробки концептуальної математичної моделі визначення максимальної активності РАВ для безпечного захоронення в приповерхневих сховищах та її апробування на прикладі одного зі сховищ комплексу «Вектор» — Лот 3 (Чорнобильська зона відчуження). Запропоновано концептуальну модель міграції ^{90}Sr з Лоту 3 через ґрунти зони аерації та водоносного горизонту. Наведено пропозиції для подальшого удосконалення моделі.

Ключові слова: концептуальна модель, радіоактивні відходи, приповерхневе сховище, ^{90}Sr, міграція, коефіцієнт розподілення радіонуклідів.

One of the fundamental safety principles of RW management declared by the IAEA [1] is to avoid imposing an undue burden on future generations. Therefore, justification of long-term safety of NSDF is one of the important issues that should be addressed by the safety case.

According to current RW classification in Ukraine, only short-lived RW can be disposed in NSDF. For short-lived waste, levels for clearance from the regulatory control of the nuclear regulatory body are reached earlier than in 300 years [2]. Therefore, criteria for clearance of radioactive materials from regulatory control are established only for specific activity of RW [3]. However, the specific activity in aeration zone and/or the first aquifer can be increased 300 years after the facility closure due to leaching of radionuclides. That is why determination of maximum activity of RW to be disposed in specific NSDF is very important.

According to Ukrainian legislation [4, 5], it is envisaged to dispose all short-lived low- and intermediate-level solid RW on the Vector site. It is located in the Chernobyl exclusion zone (ChEZ) in Ukraine at a distance of approximately 11 km from the Chernobyl NPP.

This paper contains an approach for determining the maximum activity of RW to be disposed in Vector NSDF (called Lot 3) using mathematical modelling methods. The model was tested on ^{90}Sr, which is one of the radionuclides typical for RW inventory of Vector NSDF. The maximum permissible values of ^{90}Sr activity were calculated for the moment of the facility closure; in doing so, radioactive decay was taken into account. The model describes ^{90}Sr migration in the soils of aeration zone and the first aquifer from Lot 3 NSDF, taking into account sorption of radionuclides.

The following potential exposure scenario for Lot 3 is considered for the model development:

300 years after the facility closure, as a result of destruction of the engineered barriers, all radionuclides release from the facility and migrate through soils of aeration zone and the aquifer to the direction of the well with drinking water;

the well is located at a certain distance from the NSDF;

consumption of drinking water from the well is considered as a source of public exposure.

Hence, period of the facility operation was not taken into account. This increases the conservativism of the approach applied in this paper.

Radionuclide migration in geological environment is determined by a large number of interrelated physical and chemical processes. Taking into account the complexity and interdependencies of these processes, it is worthwhile to use mathematical modelling methods to describe radionuclide behavior [6].

Parameter of radionuclide sorption. In this paper, sorption could be considered as a determinative process for migration of radionuclides through the soils of aeration zone and the first aquifer.

The following sorption parameter for conceptual model development was used in this paper.

One of the most important sorption parameters used to develop models of radionuclide migration in a porous medium is distribution coefficient K_d between solid and liquid phases for equilibrium, which is defined as the ratio of specific activities of radionuclide [7]:

$$K_d = \frac{A_s^{spec}}{A_{aq}^{spec}}, \qquad (1)$$

where K_d — radionuclide distribution coefficient, m^3/kg; A_s^{spec} — specific activity of radionuclide in the solid phase

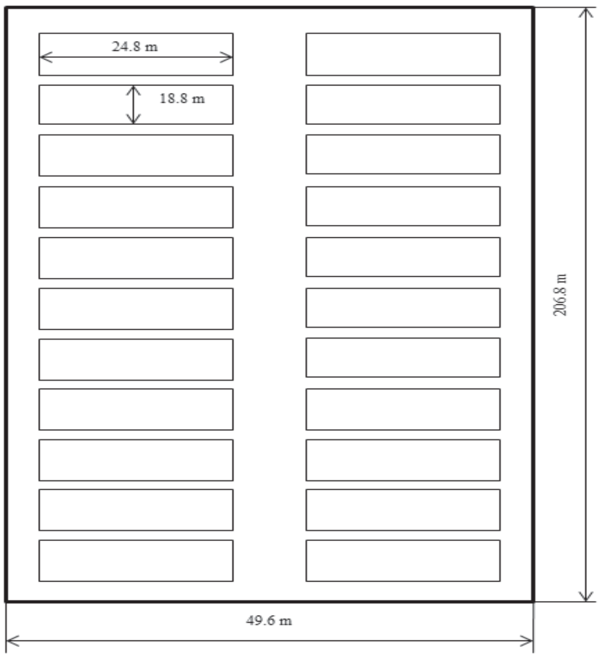

Fig. 1. Illustrative layout of Lot 3 NSDF

of the soil, Bq/kg; A_{aq}^{spec} — volume activity of radionuclide in the aqueous phase of the soil, Bq/m^3.

K_d characterizes sorption of radionuclides by underlying rocks and allows evaluating their mobility in the soil [8]. K_d value depends on numerous geochemical parameters and processes such as pH of solution; mineral composition; presence of organic matter, iron oxides; oxidization / deoxidization conditions; chemical form of radionuclide etc.

The conceptual model description. The conceptual model is based on the following input data:

overall size of storage compartment: 18.8 m×24.8 m×7.5 m; storage configuration: 2 rows of 11 modules, storage sizes respectively: 206.8 m×49.6 m (Fig. 1) [9];

period of the regulatory control of the facility after its closure is 300 years [10];

the volume of drinking water consumption for public is 0.8 m^3/a [11];

^{90}Sr permissible concentration in drinking water based on limit of the individual effective exposure dose for the public (1 mSv/a) [11, 12] is 10^4 Bq/m^3 [11].

Table 1 shows input parameters of the soils used for the model development [9, 13—16].

The following conservative assumptions were made for the model development:

in 300 years after the facility closure, all the engineered barriers are instantly destroyed, all radionuclides in water-soluble form simultaneously release from the facility to underlying rocks and migrate toward the well with drinking water;

the scenario of potential exposure takes place for Lot 3 only because simultaneous destruction of engineered barriers of all the facilities of the Vector site is too conservative approach;

engineering-geologic elements where radionuclide migration occurs are represented as 4 blocks, homogeneous as regards physical and chemical properties: small quartz sand (Layer 3), red-brown sandy loam (Layer 2), fine-grained sand with clay lenses (Layer 1) and soils of the first aquifer (Layer 0);

the dimensions of the above-mentioned blocks are the following: in the case of vertical migration, area of the blocks is equal to the area of Lot 3 (length×width) and height of the block is equal to the thickness of the layer; in the case of horizontal migration through the aquifer, height of the block is equal to thickness of the aquifer, width of the block is equal to width of Lot 3 foundation (conservative approach), and length is equal to the distance from far-end wall Lot 3 to the well;

Lot 3 contains only one radionuclide (^{90}Sr);

the well is located at the distance of 1000 m from Lot 3;

^{90}Sr penetrates into the aquifer only to the depth of several meters because of low vertical dispersion (5 m assumed as aquifer thickness);

the value of ^{90}Sr permissible concentration in drinking water (10^4 Bq/m^3) [11] is not exceeded and is equal to specific activity ^{90}Sr in aqueous phase of the aquifer.

Table 1. Input parameters of the soils used for modelling ^{90}Sr migration from Lot 3 to the well through the soils of aeration zone and the first aquifer

Parameter / Layer	**Layer 3**	**Layer 2**	**Layer 1**	**Layer 0 (The first aquifer)**
Concise characteristic of the layer	Small quartz sand	Red-brown sandy loam	Fine-grained sand+clay lenses	Aquifer, medium size sand fluvioglacial and alluvial- fluvioglacial
Length, m	2.07E+02	2.07E+02	2.07E+02	1.00E+03
Width, m	4.96E+01	4.96E+01	4.96E+01	2.13E+02
Thickness, m	7.50E+00	1.50E+00	1.00E+01	4.00E+01
Volume, m^3	7.69E+04	1.54E+04	1.03E+05	8.51E+06
Porosity	3.87E-01	3.79E-01	3.87E-01	3.75E-01
Density of the solid phase, kg/m^3	1.84E+03	1.95E+03	1.94E+03	2.01E+03
Mass of the solid phase, kg	7.08E+07	1.35E+07	7.36E+07	9.40E+09
Volume of the solid phase, m^3	3.85E+04	6.92E+03	3.80E+04	4.68E+06
Volume of the aqueous phase, m^3	3.85E+05	8.46E+03	6.46E+04	3.83E+06
K_d, m^3/kg	2.10E-03	7.00E-03	3.86E-03	4.10E-03

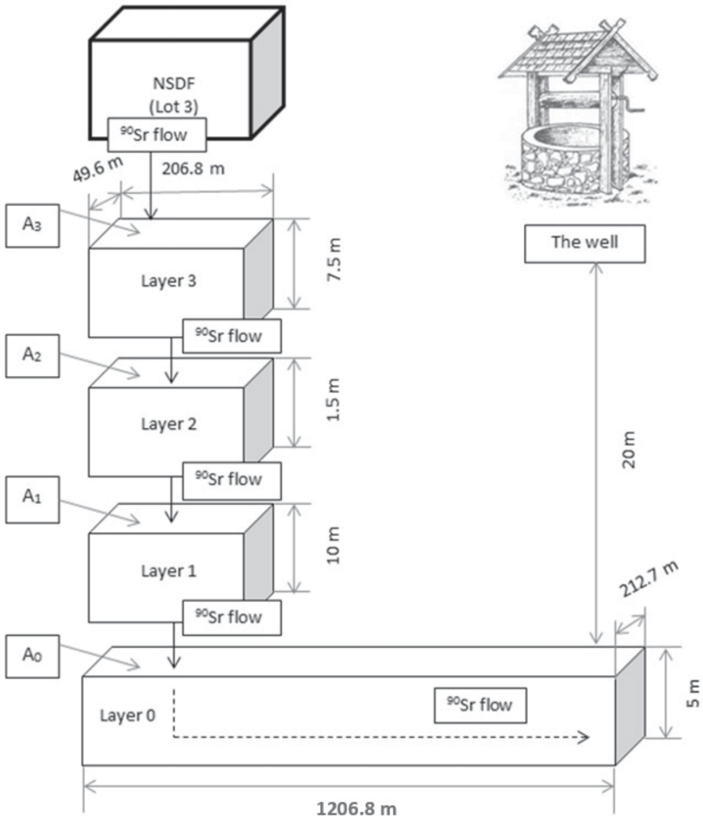

Fig. 2. Simplified illustrative conceptual model of ^{90}Sr migration through engineering-geologic elements represented as blocks

Fig. 2 shows illustrative conceptual model of ^{90}Sr migration using above-mentioned blocks. Hence, it is obvious that the value of ^{90}Sr activity in the place of inlet of ^{90}Sr flow to the next layer (A_0, A_1, A_2, A_3) is equal to the value of ^{90}Sr activity in the place of outlet of ^{90}Sr flow from the previous layer taking into account that the inverse task is resolved.

Approach to development of the conceptual model. In this paper, the ^{90}Sr maximum activity was determined by solving the inverse task provided that the scenario under which the radionuclides are leached from Lot 3 and reach the well would realize.

To develop the conceptual model of calculation of RW maximum activity in case of Lot 3 NSDF, the approach using values of radionuclides distribution coefficient K_d for all layers of soil underlying Lot 3 (soils of aeration zone and the first aquifer) was considered.

Model structure. For the above-mentioned approach the following calculation algorithm of ^{90}Sr maximum activity in RW to be disposed in Lot 3 NSDF is proposed.

1. ^{90}Sr total activity in the layer consists of radionuclide activity absorbed by the solid phase of the soil and radionuclide activity in the aqueous phase transferred through this layer. The value of ^{90}Sr specific activity in aqueous phase is the input parameter. That is why the determination of ^{90}Sr total activity in the aqueous phase of layer 0 (the first aquifer) is following:

$$A_{aq}^{0} = A_{aqi}^{spec} \cdot V_{aq}^{0}, \qquad (2)$$

where A_{aq}^{0} — ^{90}Sr total activity in aqueous phase of the layer 0, Bq; A_{aqi}^{spec} — ^{90}Sr specific activity in the aqueous phase in layer i (for layer 0 it is equal to permissible concentration in drinking water (10^4 Bq/m^3) established by Ukrainian legislation), Bq/m^3; V_{aq}^{0} — volume of aqueous phase of layer 0, m^3.

2. Taking into account the equation (1), ^{90}Sr specific activity in solid phase of layer 0 is determined as

$$A_{s0}^{spec} = K_d^0 \cdot A_{aq0}^{spec}, \qquad (3)$$

where A_{aq0}^{spec} — ^{90}Sr specific activity in solid phase of layer 0, Bq/kg; K_d^0 — ^{90}Sr distribution coefficient for layer 0, m^3/kg.

3. Determination of ^{90}Sr total activity in the solid phase of layer 0:

$$A_s^0 = A_{s0}^{spec} \cdot m_s^0, \qquad (4)$$

where m_s^0 — mass of solid phase of layer 0, kg.

4. Determination of ^{90}Sr activity in the place of inlet of ^{90}Sr flow to layer 0:

$$A_0 = A_{aq0}^{spec} \cdot \left(m_s^0 \cdot K_d^0 + V_{aq}^0\right), \qquad (5)$$

where A_0 — ^{90}Sr activity in the place of inlet of ^{90}Sr flow to layer 0, Bq.

5. Determination of ^{90}Sr activity in the place of inlet of ^{90}Sr flow to i layer (layers 1—3):

$$A_i = A_0 \cdot \left(\left(K_d^i \cdot \frac{m_s^i}{V_{aq}^i}\right) + 1\right), \qquad (6)$$

where A_i — ^{90}Sr activity in the place of inlet of ^{90}Sr flow to i layer (layers 1—3), Bq; K_d^i — ^{90}Sr distribution coefficient for i layer (layers 1—3), m^3/kg; m_s^i — mass of solid phase of i layer (layers 1—3), kg; V_{aq}^i – volume of aqueous phase of i layer (layers 1—3), m^3.

The obtained value ^{90}Sr activity in the place of inlet of ^{90}Sr flow to layer 3 is the value of activity in the bottom of Lot 3 at the moment of destruction of the engineered barriers, i.e. 300 years after the facility closure.

6. Taking into account radioactive decay, the maximum activity of ^{90}Sr at the moment of the facility closure is calculated using the following formula:

$$A_{\max} = A_3 \cdot e^{\lambda t}, \qquad (7)$$

where $A_{\max}$ — maximum activity of ^{90}Sr at the moment of the facility closure, Bq; A_3 — ^{90}Sr activity in the place of inlet of ^{90}Sr flow to layer 3, Bq; λ — decay constant, sec^{-1}; t — time, sec.

The calculations were carried out using MS Excel software.

Modelling results. This paper contains preliminary results of the calculation of maximum activity of ^{90}Sr in RW to be disposed in Vector NSDF (Lot 3). Preliminary results of modelling of ^{90}Sr maximum activity using K_d approach at the moment of destruction of engineered barriers:

Layer name	^{90}Sr calculated maximum activity in the place of inlet of ^{90}Sr flow to the Layer, Bq
Layer 0	1.65E+10
Layer 1	2.13E+11
Layer 2	4.97E+12
Layer 3	3.54E+13

Analysis of the preliminary results shows that the value of ^{90}Sr maximum total activity at the moment of Lot 3 closure using K_d approach (4.85E+16 Bq) could be compared with the values presented in paper [17]. It shows that K_d approach can be considered as generally acceptable. It confirms the statement in [18] that soils of the Vector site play significant role in ensuring long-term safety of RW disposal.

Conclusions

In this paper, a simplified conceptual model for preliminary estimate of radionuclides maximum total activity in RW to be disposed in NSDF is shown. This model is based on representing soil layers where radionuclide migration occurs (aeration zone and the first aquifer) as blocks. The model was tested in the case of Vector site NSDF (Lot 3, ChEZ) where all Ukrainian RW are planned to be disposed. In order to simplify the model, it is assumed that these RW contain only one radionuclide (^{90}Sr). The radionuclide distribution coefficient K_d approach was used for the model development. Based on the above-mentioned assumptions, the estimated value of ^{90}Sr maximum total activity to be safely placed in Lot 3 at the moment of the facility closure is 4.85E+16 Bq.

The modelling results show that K_d approach could be acceptable as a basis. However, this approach has some disadvantages. In particular, it does not reveal dependence of radionuclide activity on volume of the layer where radionuclide migration takes place. Therefore, the above-mentioned value could be used only as indicative value. K_d approach should be further improved taking into account different factors that have an influence on radionuclide migration.

The developed conceptual model would probably be more accurate if values of input parameters, in particular, values of radionuclides K_d, were more reliable. For this purpose experimental data of K_d values of radionuclides are necessary. To obtain this information it is necessary to perform additional comprehensive investigations for soils of Vector site.

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