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ESTIMATION OF RADIONUCLIDE TRANSFER TIMES IN THE GEOLOGICAL LAYERS OF SALIGNY SITE

Gheorghiţa JINESCU[1], Daniela DOGARU[2]

Lucrarea descrie evaluarea timpului de transfer pentru 60Co şi 137Cs în straturile geologice ale amplasamentului Saligny care este selectat pentru construcţia unui depozit de deşeuri radioactive. Sunt prezentate aspectele geologice ale amplasamentului Saligny necesare pentru evaluarea timpului de transfer al radionuclizilor. În lucrare este prezentat modelul conceptual şi modelul matematic asociat care descrie transferul radionuclizilor din sistemul de depozitare deşeuri radioactive în acvifer, şi este utilizat pentru calcularea variaţiei concentraţiilor 60Co şi 137Cs în fiecare compartiment al sistemului de depozitare. Timpii de transfer al 60Co şi al 137Cs sunt derivaţi din diferenţa dintre timpul când un radionuclid atinge concentraţia maximă în două compartimente adiacente. Timpii de transfer al 60Co şi al 137Cs sunt comparaţi cu timpii de înjumătăţire al radionuclizilor în scopul evaluării rolul straturilor geologice ale amplasamentului să întârzie transferului radionuclizilor şi în acelaşi timp să permită dezintegrarea naturală a radionuclizilor.

This paper describes the assessment of ^{60}*Co and* ^{137}*Cs transfer times in the geological layers of Saligny site which is selected for the construction of a radioactive waste repository. The geological aspects of Saligny site necessary to assess the transfer time of radionuclides are presented. The conceptual model of the repository as well as the associated mathematical model which describes the transfer of radionuclides from the radioactive waste disposal system to the aquifer is used to calculate the variation of* ^{60}*Co and* ^{137}*Cs concentrations into each compartment of the disposal system are presented in the paper. The* ^{60}*Co and* ^{137}*Cs transfer times are derived from the time difference of a radionuclide reaching the highest pick value of concentration into two adjacent compartments. The* ^{60}*Co and* ^{137}*Cs transfer times are compared with the half life of radionuclides in order to asses the role of the geological layers of the site in delaying the transfer of radionuclides, allowing as well the natural decay of radionuclides.*

Keywords: radioactive waste, repository, safety indicators, safety assessment

1. Introduction

Low and intermediate level radioactive wastes generated by the operation and decommissioning of Cernavoda Nuclear Power Plant are planed to be disposed into a new repository located on Saligny site. The repository will be of

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[1] Prof., Dept. of Chemical Engineering, University POLITEHNICA of Bucharest, Romania

[2] PhD Eng., National Commission for Nuclear Activities Control, Bucharest, Romania, daniela.dogaru@cncan.ro

near surface type, with multiple barriers. In order to assess the safety as well as the adequacy of the Saligny geology as host rock for the repository, many evaluations and laboratory tests were done [1], [2] and [3].

The disposal of radioactive waste needs to be carried out in a manner that provides an acceptable level of safety and which can be proved to comply with the established regulatory requirements and criteria. Safety assessment techniques are used to evaluate the performance of a repository and its impact on human health and on the environment. The most common techniques used in safety assessment is to evaluate the impact of the repository on human health in terms of radiological dose, radiological risk, concentrations or fluxes of radionuclides in different compartments of the disposal system as well as the radionuclide transfer time in different compartments of the disposal system.

This paper describes the estimation of ^{60}Co and ^{137}Cs transfer time into the geological disposal layer of the Saligny site. The radionuclide transfer time is used to construct arguments in order to evaluate the performance of the disposal system.

2. Description of Saligny site

Geologically, the Saligny site belongs to the Dobrogean part of the Moessic platform, placed at south of Ovidiu – Capidava fault in the South-Dobrogea platform. The main characteristic of this zone is the deep crystalline foundation covered by thick sedimentary layers. Saligny site structure consists of the sequence of the following geological units: silty loess, clayey loess, Quaternary red clay, Pre-Quaternary clay, Barremian limestone, Vallanginian clay, Jurassic limestone, Paleozoic sediments and the crystalline foundation of the green slates [4].

The significant units to the long-term dose assessment for the population in the surrounding area are the silty loess (Horizon A), the clayey loess (Horizon B), the Quaternary red clay (Horizon C), the Pre-Quaternary clay (Horizon D) and the Barremian limestone. According to the site stratigraphy, the Horizon B is also split in four sub-layers: Iab1, Ib (upper), Iab2 and Ib (lower), having different clay content [5].

From hydro-geologically point of view, the unsaturated (vadose) zone includes the silty loess, the clayey loess, the Quaternary red clay and the upper part of the Pre-quaternary clay, while the saturated zone includes the lower part of Pre-quaternary clay (local small aquifers) as well as the Barremian limestone – host of the main aquifer.

3. Parameters used as input data

The structural parameters of each geological unit of the Saligny site used for assessment of the transfer time of radionuclides are dry density, water filled porosity and depth layer, which are presented in Table 1 [4].

Table 1

Parameters of Saligny site

Horizon	Dry density [kg m^{-3}]	Water filled porosity [%]	Depth layer [m]
Waste form, Slab foundation	2500	0.15	4
Foundation ground	1780	25.68	2
Horizon A – silty loess	1540	12.30	1
Horizon B – clayey loess:			
- layer Iab1	1780	25.68	6
- upper layer Ib	1570	14.03	4
- layer Iab2	1720	25.13	2
- lower layer Ib	1690	25.20	6
Horizon C – quaternary red clay	1760	32.38	8
Horizon D– pre-quaternary clay	1760	30.67	16
Barremian limestone	1800	0.30	-

The values of the distribution coefficients which have been obtained experimentally for the samples collected from Saligny site, for the radio-nuclides ^{137}Cs and ^{60}Co corresponding to different horizons are presented in the Table 2 [1].

Table 2

Distribution coefficients (k_d) for the main horizons of Saligny site

Radionuclide	k_d in the Saligny Horizons [m^{3}kg^{-1}]			
	Horizon A Silty loess	Horizon B Clayey loess	Horizon C Red Clay	Horizon D Pre-Quaternary clay
^{137}Cs	0.774	1.131	4.131	2.366
^{60}Co	0.033	0.030	0.031	0.030

An infiltration rate of about 20 mm/year is specified in [4] for Saligny site and the natural ground. In the first 300 years after the repository closure, its final cover is considered effective and the infiltration rate in the disposal structures is assumed to be zero. Between 300 and 500 years, the infiltration rate in the repository is considered of about 10% from the natural infiltration rate and, after 500 years, the infiltration rate in the disposal structures is assumed to be as in the natural ground.

The initial activity of the radionuclides considered in the evaluation are 8.9E+14Bq for ^{137}Cs and 2.9E+11Bq for ^{60}Co[6].

4. Conceptual and mathematical model

The conceptual model of Saligny repository has been developed using the interaction matrix method in accordance with the requirements of the evaluation with AMBER computer code. The disposal system was split into series of assumed homogenous compartments and the transfer processes between the compartments were expressed as transfer coefficients that represent the activity fraction in a particular compartment transferred from this to another, per unit time. The conceptual model of Saligny disposal facility considered in the assessment of this paper is presented in Fig.1.

The upper compartments of the model are the engineered structures which consist of waste form, slab foundation and improved foundation ground. The lower ones are the geological layers of Saligny site which consist of silty loess, clayey loess, Quaternary red clay, Pre-quaternary clay and Barremian limestone, representing the unsaturated (vadose) zone. The saturated zone includes the lowest compartments of Pre-quaternary clay as well as the Barremian limestone.

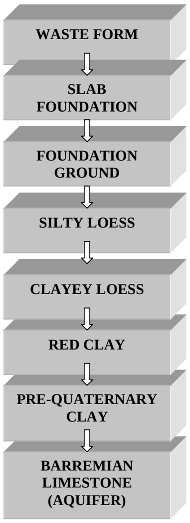

Fig. 1. Conceptual model of the Saligny disposal system

The mathematical model developed in order to assess the distribution of the radionuclide concentration in the geological layers of the Saligny site, and, consequently, the radionuclide transfer time, is mainly based on the release, migration and transport of radio-nuclides from waste, considering only the decay, adsorption, dispersion and advection processes.

The mathematical representation of the inter-compartmental transfer processes takes the form of a matrix of transfer coefficients that allow the compartmental amounts to be represented as a set of first order linear differential equations. For the i^{th} compartment, the rate at which the inventory of radionuclides in a compartment changes with time is given by:

$$\frac{dN_i}{dt} = \left(\sum_{j \neq i} \lambda_{ji} N_j + \lambda_N M_i + S_i(t) \right) - \left(\sum_{j \neq i} \lambda_{ij} N_i + \lambda_N N_i \right) \tag{1}$$

where *i* and *j* indicate compartments, *N* and *M* are the amounts (Bq) of radionuclides *N* and *M* in a compartment (*M* is the precursor of *N* in a decay chain). *S(t)* is a time dependent external source of radionuclide *N* (Bq y^{-1}). λN is the decay constant for radionuclide N (y^{-1}) and λji and λij are transfer coefficients (y^{-1}) representing the gain and loss of radionuclide *N* from compartments *i* and *j*. For simplicity, the above equation assumes a single parent and daughter. The transport equation for unsaturated and saturated geological layers was simplified in order to be solved using the AMBER computer code.

The dispersion was considered by the discretisation of the disposal facility compartments.

Radioactive decay is represented through the decay rate (λ, in y^{-1}), which is given by the equation (2):

$$\lambda = \frac{\ln 2}{t_{1/2}} \tag{2}$$

where $t_{1/2}$ is the half-life period of the radionuclide (y).

The adsorption is described through the retardation phenomenon, which, for a given compartment, is dependent on the radionuclide. The retardation factor R (dimensionless) specific for a compartment is calculated using equation (3):

$$R = 1 + \frac{\rho \cdot k_d}{\vartheta_w} \tag{3}$$

where ρ is the dry bulk density of the compartment ($kg \cdot m^{-3}$), k_d is the distribution coefficient of the element in the compartment ($m^3 \cdot kg^{-1}$) and ϑw represents the water-filled porosity of the analyzed compartment (dimensionless).

For the unsaturated transfers, the advective transfer rate of radio-nuclides (λ_flow, in y^{-1}) is given by equation (4):

$$\lambda_{flow} = \frac{q}{L \cdot \vartheta_w \cdot R} \tag{4}$$

where *q* represents the annual flow rate through the compartment ($m \cdot y^{-1}$), *L* represents the length of the compartment on the direction of the water flow (m), ϑw represents the water-filled porosity of the compartment (dimensionless), and *R* represents the retardation of the compartment (dimensionless). The parameter *q* ($m \cdot y^{-1}$) represents the infiltration rate of water through the repository compartments and geosphere (unsaturated zone).

For the saturated zone, the water infiltration rate is given by equation (5):

$$q = K \frac{\partial H}{\partial x} \tag{5}$$

where K represents the hydraulic conductivity of the compartment ($m \cdot y^{-1}$) and $\partial H/\partial x$ represents the hydraulic gradient (dimensionless).

5. Results and discussions

In this paper, the transfer time is defined as the time period necessary for a radionuclide to transit a disposal system compartment. The transfer time can not be obtained directly, as solution of the transport equation. In the present assumptions, the parameter directly obtained from the transport equation is the radionuclide concentration in each disposal system compartment. Based on this parameter, the transfer time can be calculated taking into account the first moment when the radionuclide reaches the compartment, and the late moment, when the radionuclide leaves the compartment. That is why measuring or calculating that period of time for each radionuclide is not possible, and, because the radionuclide migration process is considered to be continuous, the transfer time is calculated based on the variation of the radionuclide concentration in the disposal system compartments, taking into account the moments when the radionuclide concentration reaches the peak values in two adjacent compartments.

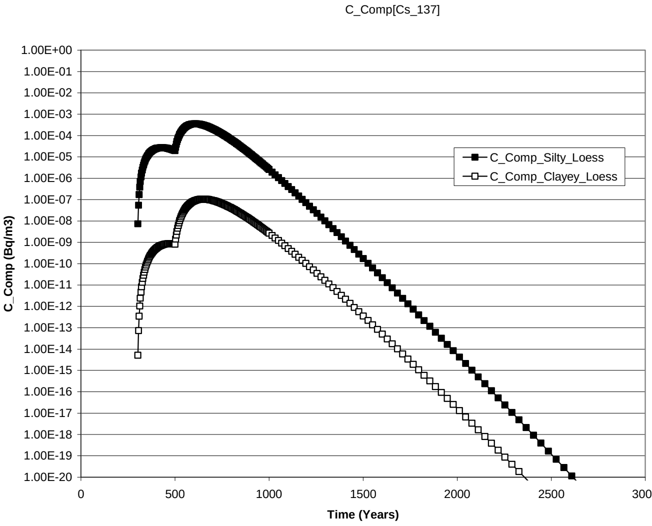

Fig. 2. Time variation of ^{137}Cs concentration in Silty loess and Clayey loess

The time variation of the ^{137}Cs concentration in silty loess and the upper part of clayey loess is presented in Fig. 2, in order to illustrate the transfer time calculation. Based on the results presented in Fig. 2, it is possible to calculate the moments when the ^{137}Cs concentration for each compartment reaches the minimum earlier values, the peak values and the minimum late values. Thus, the transfer time can be defined as the difference between the moments when the peak values are reached, for each compartments as presented in Fig.1.

As Fig.2 shows, the radionuclide concentration in a compartment has two peak values, corresponding to the assessment assumptions, namely the variation of water infiltration rate. Consequently, the transfer time will be calculated taking into account the higher peak values presented for each disposal system compartment.

Figs. 3 and 4 present the transfer times (x axis, years) for the ^{137}Cs and ^{60}Co in the geological layers of Saligny site (the depth under the repository foundation represents y axis, meters).

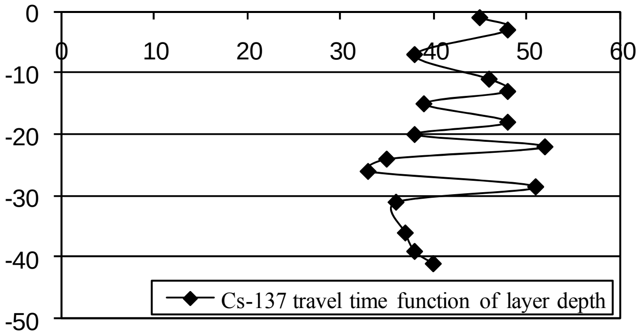

Fig. 3. Transfer time function of the geological layer depth, for ^{137}Cs

Analyzing Figs 3 and 4, it can be observed that the transfer time values are higher than their half-life periods, which will allow the decay of radionuclides in the geological layers, before they will reach the site aquifer.

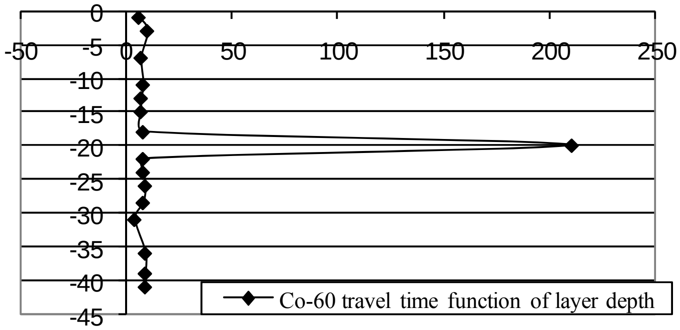

Fig. 4. Transfer time function of geological layer depth, for ^{60}Co

5. Conclusions

Analyzing Figs 3 and 4, it can be observed that the transfer time values are higher than their half-life periods, which will allow the decay of radionuclides in the geological layers, before they will reach the site aquifer.

The transfer time values for 137Cs vary between 33 years and 52 years, and it can be compared with the half-life period of that isotope, which is 30 years.

For ^{60}Co, the lowest transfer time value is of about 4 years, and the highest one is of 210 years. These transfer time values can also be compared with the half-life period of ^{60}Co, which is 5.27 years. Especially for ^{60}Co, the transfer time between the clayey loess and the Quaternary red clay have the highest value, allowing the natural decay of that isotope at very low radioactivity values. The transfer time values are higher than their half-life periods for all radionuclides, fact which allows the decay of radionuclides in the geological layers, before they will reach the site aquifer. In conclusion, the assessments confirm the role of the site as geological barrier of the repository.

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