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A RELIABILITY STUDY ON INFLUENCE OF THE GEOSPHERE THICKNESS OVER THE ACTIVITY RELEASE FROM A NEAR SURFACE RADIOACTIVE WASTE REPOSITORY

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ABSTRACT

Infiltration of water into a waste disposal facility and into the waste region is the main factor inducing the release of radionuclides from a disposal facility. Since infiltrating water flow is dependent on the natural percolation at the site and the performance of engineered barriers, its prediction requires modelling of unsaturated water flow through intact or partially/completely failed components of engineered barriers and through the rock layer of the geosphere on which the repository is constructed. The engineered barriers include the cover systems, concrete vault, backfill, waste forms, and overpacks. This paper aims to carry out a performance study regarding a near surface repository in terms of reliability engineering. It is assumed that surface water infiltrates through the barriers reaching the matrix where radionuclides are contained, thus releasing them into the environment. The repository consists of a set of barriers which are considered saturated porous medium. As results, this paper presents the relation between the thickness of the geosphere layer and the radionuclide release rate in terms of activity. Such results represent a useful information for choosing the repository sites in order to keep the released activity in acceptable levels over time.

1. INTRODUCTION

The deposition system safety for the Low and Intermediate Level Radioactive Wastes is determined by the performance of its individual components, by shape and containment of the wastes, engineering barriers and natural barriers (host environment). Previously, multiple barriers systems were regarded as groups of independent individual barriers that worked in sequence [1,2,3]. However, this concept is currently seen in a more integrated and synergic way, as complementary barriers that operate simultaneously and together [4].

A reliability modelling for packages of radioactive waste has been developed. Chesnut [5] has dealt with the mean time to failure (MTTF) for a single package and for multiple packages, while Ananda [6] has used the conditional approach for failure of the packages caused by the high temperature of the high level waste. Ananda [7] has extended the conditional approach for multiple packages, more precisely, for two barriers. Aguiar et al. [8] have discussed the reliability for a large number of packages. This discussion is based on the use of probability distribution that best represents the MTTF of the packages.

Aguiar and Damaso [9] employed genetic algorithm to establish an optimized configuration to the structure of the repository, by determining the thicknesses of barriers and waste packages arrangement, taking into account the engineering and radiological constraints. The radionuclide Tc-99 was chosen because of there is no retention of Tc in the geosphere. Now, the idea is to study the influence of the thickness of the geosphere layer in the radionuclide release rate in terms of activity with the time. This paper aims to carry out a performance study regarding a near surface repository in terms of reliability engineering for the important radionuclides selected among those present in intermediate level wastes [10,11].

2. PROBLEM CHARACTERIZATION

Based on International Atomic Energy Agency (IAEA) recommendations and international practice, Near Surface Repositories are an option for the permanent storage of Low and Intermediate Level Radioactive Wastes. The hypothetic repository is divided in 6 subsystems: A – top cover; B – upper layer; C – packages; D – base; E – repository walls, and F – geosphere. Release of contaminated water from the base is taken into account as well as from the side walls. Subsystems A and B are barriers against water infiltration while subsystems C, D, E, and F are barriers which delay the leaching of the radionuclide into the biosphere. The repository is viewed as a system whose components (barriers) work in a series-parallel configuration according to Fig. 1, where the system failure occurs only when the barriers A and B and C and (D or E) and F fail. The time to failure of each barrier is considered a random variable with a specified probability density function. The probability of system failure is obtained considering the lifetime (random failures). The exponential distribution is adopted to represent the time to fail of each barrier of the system (repository). In addition, this distribution often provides a good representation of the probability for the failure time when failures are rare events resulting from complex interactions of many processes and mechanisms [5] and when many different causes of failures overlap each other [3]. According to Lewis [12], by their nature passive parallel systems involve dependency between components. Thus, this kind of system is satisfactorily analyzed by Markov methods.

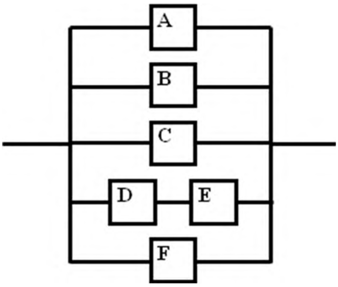

Figure 1: Series-parallel configuration of the repository barriers.

The probability of system failure is calculated by the Markovian approach, since the failure rates of subsystems (barriers) are constants. The failure criterion of the module, for each radionuclide, is the failure of the barriers A and B and C and (D or E) and F, i.e. the module fails if activity is released from the module, through its base or walls, to the biosphere. The transition diagram of Markov states for the probability of repository failure is presented in Fig. 2.

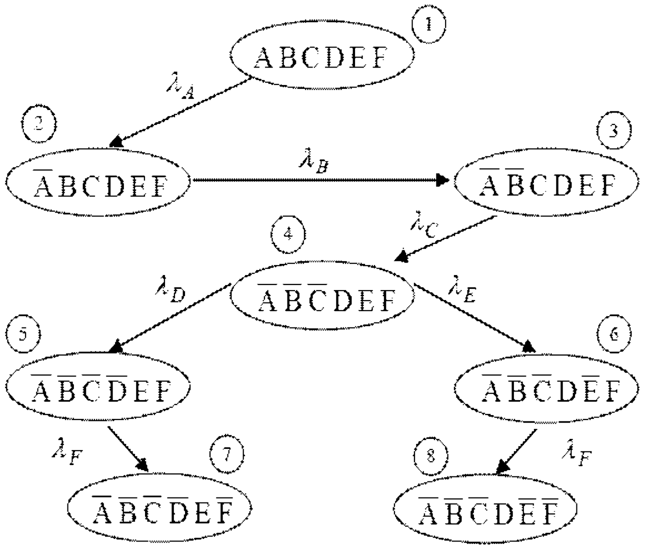

Figure 2. The transition diagram of Markov states for the hypothetic repository.

Solving the Markov model system of differential equations, one obtains the probability of finding the module at time t in each of the diagram states shown in Fig. 2 for each radioactive element. The equation that represents the probability of failure of the module is the sum of the probabilities of states 7 (P7) and 8 (P8) and it is a cumulative distribution function (cdf).

3. ACTIVITY RELEASED

Infiltration predicted by the water infiltration models becomes important only after the failure of a concrete barrier in performance assessment. Before a failure, it is assumed that a concrete vault works as an effective barrier to minimize the contact of water with the waste. The time of failure of concrete vault is important, along with the time interval between the initiation and completion of concrete vault failure. The performance history of modern concrete is short (-100 years) relative to the required time frame of performance assessment, therefore, prediction of concrete service life is difficult [13]. It is admitted that no infiltration will happen due to the active institutional control in the first 100 years [10,14]. Hence, the time the water takes to cross the concrete top cover will be added by 100 years.

A saturated porous medium is considered in order to calculate the superficial speed of the water referent to the total transversal draining area through the barriers. The water speed is obtained from the Darcy's equation for a porous saturated medium, except for the geosphere, Eqn. 1, 2, 3 4, 5, 6 e 7 [9].

$$va_1(t) = k(t)\frac{(h+e_A)}{e_A} = k(t)\frac{H}{e_A} \tag{1}$$

$$va_2(t) = k(t)\frac{(H+e_B)}{e_B} \tag{2}$$

$$va_{3v}(t) = k(t)\frac{(H+e_B+e_{Cv})}{e_{Cv}} \tag{3}$$

$$va_{3h}(t) = k(t)\frac{(H+e_B+e_{Cv})/2}{e_{Ch}} \tag{4}$$

$$va_4(t) = k(t)\frac{(H+e_B+e_{Cv}+e_D)}{e_D} \tag{5}$$

$$va_5(t) = k(t)\frac{(H+e_B+e_{Cv})/2}{e_E} \tag{6}$$

$$va_6 = \frac{K_g.i_g}{p_g} \tag{7}$$

where,
k(t) is the concrete hydraulic conductivity, given by eqn. 8;
h is the water column height above the concrete top cover (subsystem A);
H is the height corresponding to the thickness of the concrete top cover (e_A) plus the height h;
e_B is the thickness of the upper layer (subsystem B);
e_{Cv} is the thickness of the packages in the vertical direction (subsystem C);
e_{Ch} is the thickness of the packages in the horizontal direction (subsystem C);
e_D is the thickness of the base (subsystem D);
e_E is the thickness of the repository walls (subsystem E).

The water speed through each barrier, va_z, is: 1,25E-01 m/y (barrier A); 2,06E-01 m/y (barrier B); 6,38E-02 m/y and 2,08E-02 (barrier C); 4,33E-01 m/y (barrier D); 3,10E-01 m/y (barrier E) and 1,83E02 m/y (barrier F). The radionuclide speed (Eqn. 8) is equals the water speed divided by the radionuclide retardation coefficient of the medium (Eqn. 9).

$$vr_z = \frac{va_z}{Fr} \tag{8}$$

$$Fr = 1 + \left(\frac{1-p}{p}\right)\rho.kd \tag{9}$$

where,
vr_z is radioactive element speed through the barrier z (m/yr).
va_z is water speed through barrier z (m/yr).
Fr is radioactive element retardation coefficient in the geosphere.
k_d is distribution coefficient (m^3/kg).
p is porosity.
ρ is density (kg/m^3).

The most important radioactive elements selected from those present in the medium activity waste are [10]: Co, Ni, Sr, Tc, I, Cs, Pu, Am e Cm. The activity considered for radionuclide selected, at the hypothetical year of the repository sealing, is presented in Table 1.

Table 1: Activity considered for radionuclide selected, in the hypothetic year of the repository closing.

Radionuclide	Activity, Bq	Half-life (y)
^{60}Co	$0.26\text{x}10^{15}$	5.27
^{59}Ni	$0.30\text{x}10^{13}$	$7.5\text{x}10^{4}$
^{63}Ni	$0.33\text{x}10^{15}$	96
^{90}Sr	$0.61\text{x}10^{13}$	29.12
^{99}Tc	$0.19\text{x}10^{12}$	$2.1\text{x}10^{5}$
^{129}I	$0.29\text{x}10^{09}$	$1.57\text{x}10^{7}$
^{135}Cs	$0.27\text{x}10^{10}$	$2.3\text{x}10^{6}$
^{137}Cs	$0.51\text{x}10^{15}$	30
^{238}Pu	$0.14\text{x}10^{11}$	87.7
^{239}Pu	$0.87\text{x}10^{10}$	$2.41\text{x}10^{4}$
^{240}Pu	$0.89\text{x}10^{10}$	$6.537\text{ x}10^{3}$
^{241}Pu	$0.17\text{x}10^{12}$	14.4
^{241}Am	$0.13\text{x}10^{11}$	432.2
^{244}Cm	$0.74\text{x}10^{10}$	18.11

Considering the barrier A and B are barriers against water infiltration and the radioactive waste is in the barrier C, so only from this point there will be leaching of the radionuclide. Therefore, the radioactive element speed, vr_z, will be calculated for the barriers C, D, E e F. After passing through the barrier B, the water will pass through the packages (barrier C) in vertical and horizontal directions. The retardation coefficient, Fr, in the concrete/cement and in the geosphere, as well as radioactive element speed through the barriers, vr_z, are presented in Table 2.

Table 2: The retardation coefficient in the concrete/cement and in the geosphere and speed through the barriers for radioactive element, from Equations 8 and 9.

	Retardation Coefficient		Speed through the barrier z, vr_z, (m/yr).				
Radionuclide	Concrete/ Cement	Geosphere	$vr_{C,v}$	$vr_{C,h}$	vr_D	vr_E	vr_F
Co	162.134	67366.0	$3.93\text{x}10^{-04}$	$1.29\text{x}10^{-04}$	$2.67\text{x}10^{-03}$	$1.91\text{x}10^{-03}$	$2.72\text{x}10^{-03}$
Ni	1612.34	67366.0	$3.95\text{x}10^{-05}$	$1.29\text{x}10^{-05}$	$2.68\text{x}10^{-04}$	$1.92\text{x}10^{-04}$	$2.72\text{x}10^{-03}$
Sr	2.61134	2695.6	$2.44\text{x}10^{-02}$	$7.98\text{x}10^{-03}$	$1.66\text{x}10^{-01}$	$1.19\text{x}10^{-01}$	$6.80\text{x}10^{-02}$
Tc	81.5671	1	$7.82\text{x}10^{-04}$	$2.55\text{x}10^{-04}$	$5.30\text{x}10^{-03}$	$3.80\text{x}10^{-03}$	$1.83\text{x}10^{+02}$
I	2.61134	270.46	$2.44\text{x}10^{-02}$	$7.98\text{x}10^{-03}$	$1.65\text{x}10^{-01}$	$1.19\text{x}10^{-01}$	$6.78\text{x}10^{-01}$
Cs	17.1134	67366.0	$3.73\text{x}10^{-03}$	$1.22\text{x}10^{-03}$	$2.53\text{x}10^{-02}$	$1.81\text{x}10^{-02}$	$2.72\text{x}10^{-03}$
Pu	162.134	673651.0	$3.93\text{x}10^{-04}$	$1.28\text{x}10^{-04}$	$2.67\text{x}10^{-03}$	$1.91\text{x}10^{-03}$	$2.72\text{x}10^{-04}$
Am/Cm	81.5671	53893.0	$7.82\text{x}10^{-04}$	$2.55\text{x}10^{-04}$	$5.30\text{x}10^{-03}$	$3.80\text{x}10^{-03}$	$3.40\text{x}10^{-03}$

In this paper, the determination of the probability of failure of the repository is made only for the actual release of radionuclides from rainwater. However, as the long term behaviour of the system is unknown, it is not possible to precisely determine the instant in which each subsystem and the system as a whole will fail. Therefore, it is necessary to estimate the average time for failure of each barrier (tr_z), which is correlated with the mean time to failure (MTTF) of the distribution function, previously established for each barrier. In this case, the distribution function is the two-parameter exponential distribution, where its *MTTF* is given by Eqn 10. The barrier failure rates for radioactive element is presented in Table 3 for the barriers: A; B; C; D and E. The barrier F (geosphere) does not have a unique value for failure rates because the idea this paper is to correlate the thickness of the geosphere layer and the radionuclide release rate.

$$tr_z = MTTF_z = \frac{1}{\lambda_z} + t_o \qquad (10)$$

where,

λ_z is failure rates of the barriers the barrier z (y^{-1}).

t_o is time after repository closure will water reach the first barrier, 100 years.

Table 3: Barrier failure rates (1/y) for each radioactive element, from Equation 10.

	Co	Ni	Sr	Tc	I	Cs	Pu	Am/Cm
λ_A =	9.69×10^{-03}							
λ_B =	9.85×10^{-03}							
λ_C =	2.44×10^{-05}	2.46×10^{-06}	1.32×10^{-03}	4.85×10^{-05}	1.32×10^{-03}	2.27×10^{-04}	2.44×10^{-05}	4.85×10^{-05}
λ_D =	2.50×10^{-03}	3.24×10^{-04}	9.54×10^{-03}	3.99×10^{-03}	9.54×10^{-03}	7.60×10^{-03}	2.50×10^{-03}	3.99×10^{-03}
λ_E =	2.42×10^{-03}	3.11×10^{-04}	9.52×10^{-03}	3.88×10^{-03}	9.52×10^{-03}	7.51×10^{-03}	2.42×10^{-03}	3.88×10^{-03}

The equation that represents the probability of failure of the module is the cumulative distribution function (cdf), obtained from the Markovian approach, whose derivative provides the failure probability density function (pdf). The amount of activity released per year is estimated from the failure pdf [1,3,15,16]. Therefore, the amount of activity released per year (Bq/y) is the product between the pdf (y^{-1}) and the activity (Bq) per module is given by Eqn 11 [17]. In order to calculate the activity in a period of 1000 years, Bateman's equations [18] are employed for *daughter* isotopes with half-life greater than 10 years.

$$Q^k(t) = f^k(t).A^k(t) = \frac{(\psi_1 + \psi_2 + \psi_3 + \psi_4 + \psi_5)}{\prod_{n=1}^{11} \Lambda_n}.A_0^k.e^{-\alpha^k.t} \qquad (11)$$

where,

$Q^k(t)$ is activity released per year of the radionuclide k (Bq/y)

$f^k(t)$ is failure probability density function of the radionuclide k (y^{-1})

$A^k(t)$ is activity of the radionuclide k (Bq)

A_0^k is initial activity of the radionuclide k (Bq)
α^k is decay constant of the radionuclide k (y^{-1}).

$$\psi_1 = +\lambda_A.\lambda_B.\lambda_C.\lambda_F.e^{-\lambda_F.t}.\Lambda_1.\Lambda_2.\Lambda_3.\Lambda_5.\Lambda_6.\Lambda_8.(\Lambda_{10})^2$$
$$\psi_2 = -\lambda_A.\lambda_B.\lambda_C.(\lambda_D + \lambda_E).\lambda_F.e^{-(\lambda_D+\lambda_E)t}.\Lambda_1.\Lambda_2.\Lambda_4.\Lambda_5.\Lambda_7.\Lambda_9.\Lambda_{10}$$
$$\psi_3 = +\lambda_A.\lambda_B.\lambda_C.\lambda_F.e^{-\lambda_C.t}.\Lambda_1.\Lambda_3.\Lambda_4.\Lambda_6.\Lambda_7.(\Lambda_{10})^2.\Lambda_{11}$$
$$\psi_4 = -\lambda_A.\lambda_B.\lambda_C.\lambda_F.e^{-\lambda_B.t}.\Lambda_2.\Lambda_3.\Lambda_4.\Lambda_8.\Lambda_9.(\Lambda_{10})^2.\Lambda_{11}$$
$$\psi_5 = +\lambda_A.\lambda_B.\lambda_C.\lambda_F.e^{-\lambda_A.t}.\Lambda_5.\Lambda_6.\Lambda_7.\Lambda_8.\Lambda_9.(\Lambda_{10})^2.\Lambda_{11}$$

$$\begin{array}{l} \Lambda_1 = \lambda_A - \lambda_B \\ \Lambda_2 = \lambda_A - \lambda_C \\ \Lambda_3 = \lambda_A - \lambda_D - \lambda_E \\ \Lambda_4 = \lambda_A - \lambda_F \end{array}, \begin{array}{l} \Lambda_5 = \lambda_B - \lambda_C \\ \Lambda_6 = \lambda_B - \lambda_D - \lambda_E \\ \Lambda_7 = \lambda_B - \lambda_F \end{array}, \begin{array}{l} \Lambda_8 = \lambda_C - \lambda_D - \lambda_E \\ \Lambda_9 = \lambda_C - \lambda_F \end{array}, \begin{array}{l} \Lambda_{10} = \lambda_D + \lambda_E \\ \Lambda_6 = \lambda_D + \lambda_E - \lambda_F \end{array}$$

and $\lambda_A, \lambda_B, \lambda_C, \lambda_D, \lambda_E, \lambda_F$ are the failure rates of the barriers A, B, C, D, E and F, respectively.

3. RESULTS AND DISCUSSION

From Eqn 5, the release rates of the 14 radionuclides listed in Table 1 were calculated. The release rate of each radionuclide presents a different behaviour over time, mainly due to the different values of activity and failure rate of the barriers. In order to exemplify these different behaviours observed, four radionuclides – Sr-90, I-129, Tc-99 and Ni-59 – were selected and have their ***release rate*** (*Bq/y*) x ***geosphere thickness*** (*m*) x ***time*** (*y*) graphs shown in Fig. 3, 4, 5 and 6, respectively.

Sr-90 presents its peak of release rate within the period studied, between the 120th and the 130th years, for any geosphere thickness. Due to its lower retardation coefficient in the geosphere and high initial activity, still there is a significant release until after the 300th year, even for a geosphere thickness of 100 metres.

Cs-137 also presents its peak of release rate within the period studied but, due to its higher value of initial activity and lower half-live, there is no observable release after 500 years when the geosphere thickness is very close to zero. Due to its higher retardation coefficient in the geosphere, some few metres of geosphere are enough to reduce drastically the release rate.

Tc-99 has a singular behaviour. Due to the fact of this radionuclide is not be retained by the geosphere, the behaviour is determined by the characteristics and parameters associated to the engineered barriers, independently of the geosphere thickness. After 1000 years, the release rate continues to increase.

Ni-59, similarly to Tc-99, does not present the peak of release rate within the period of 1000 years. But, by the other side, the release rate is strongly reduced by the geosphere.

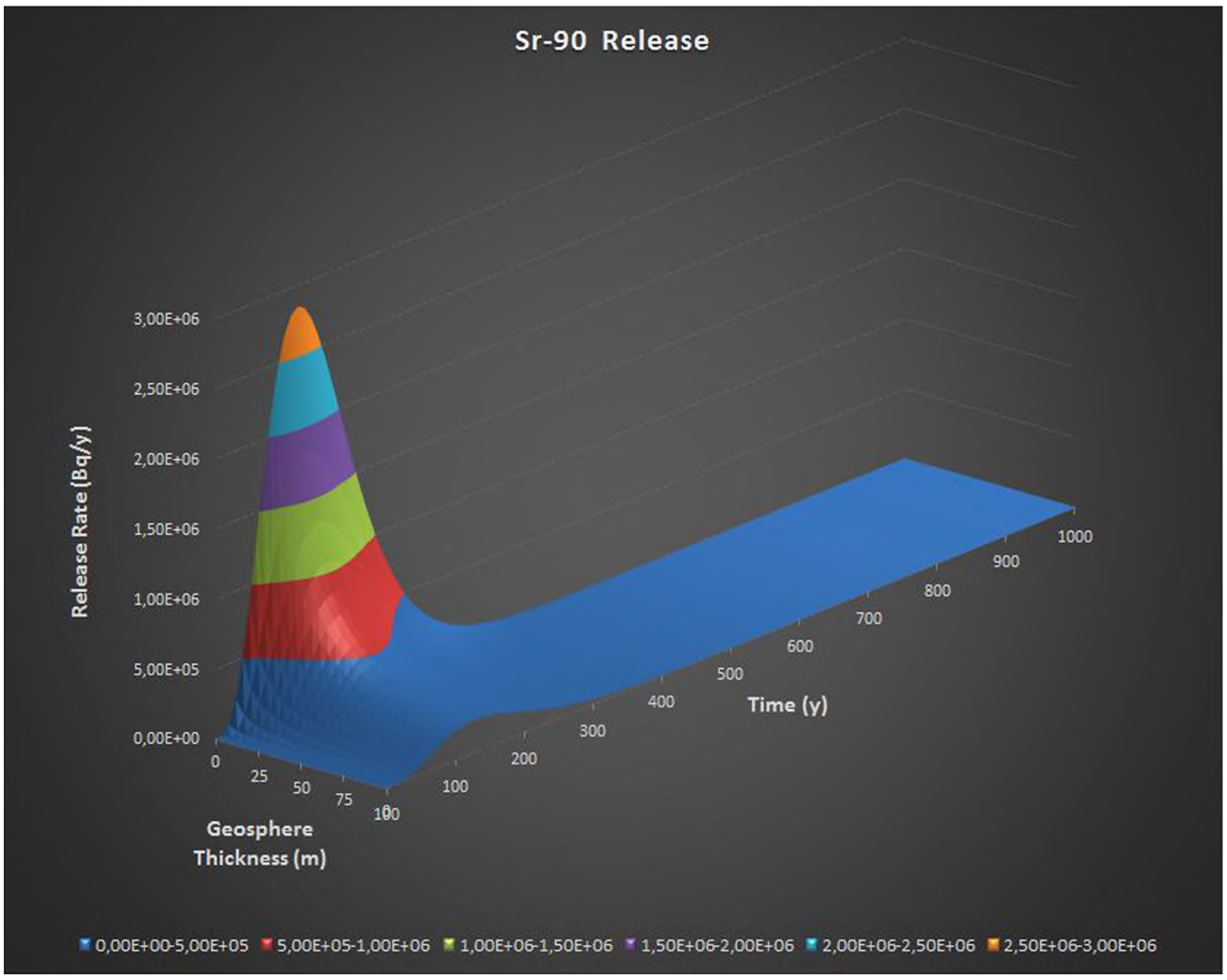

Figure 3. Release rate from the module of the repository to the biosphere for Sr-90.

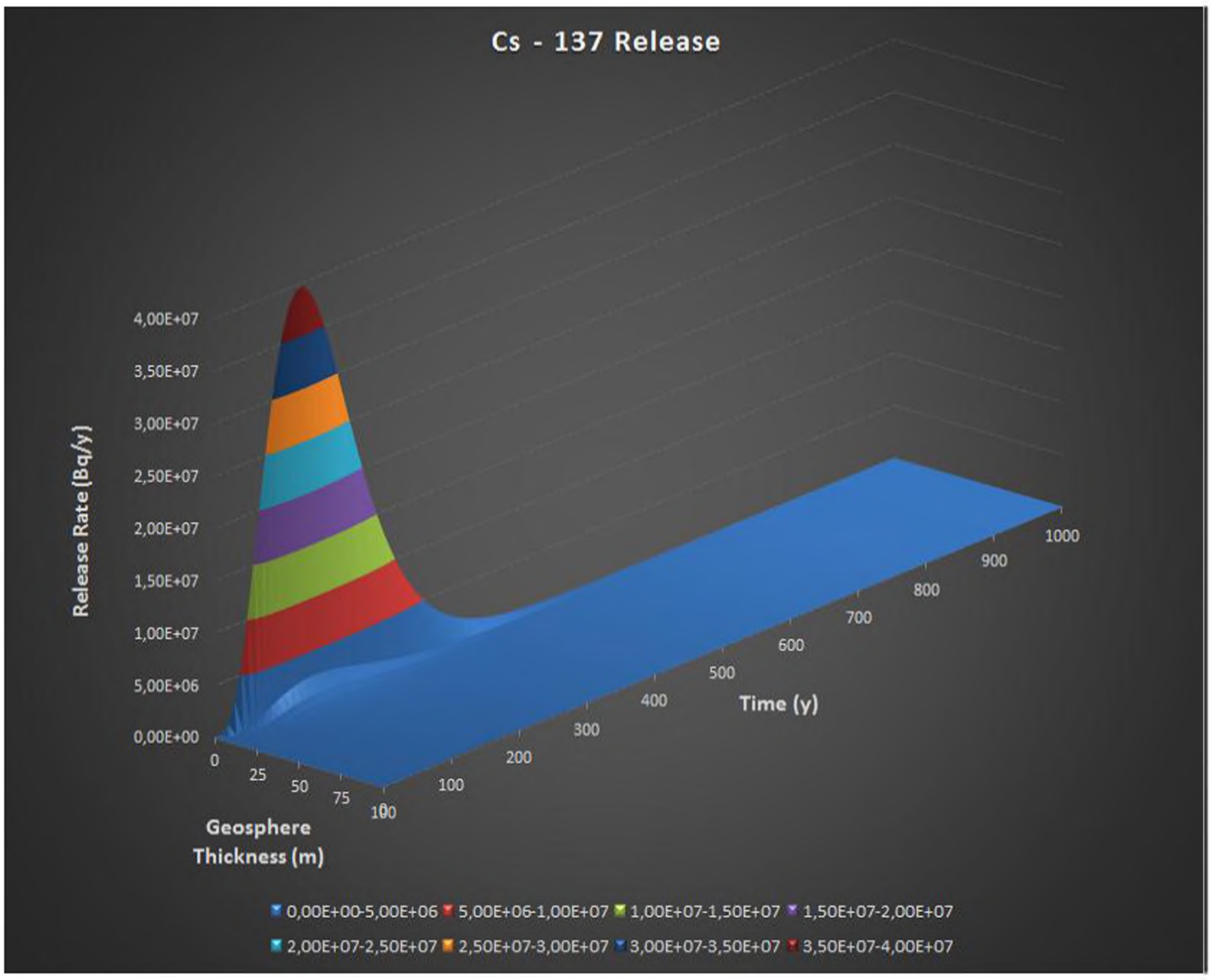

Figure 4. Release rate from the module of the repository to the biosphere for Cs-137.

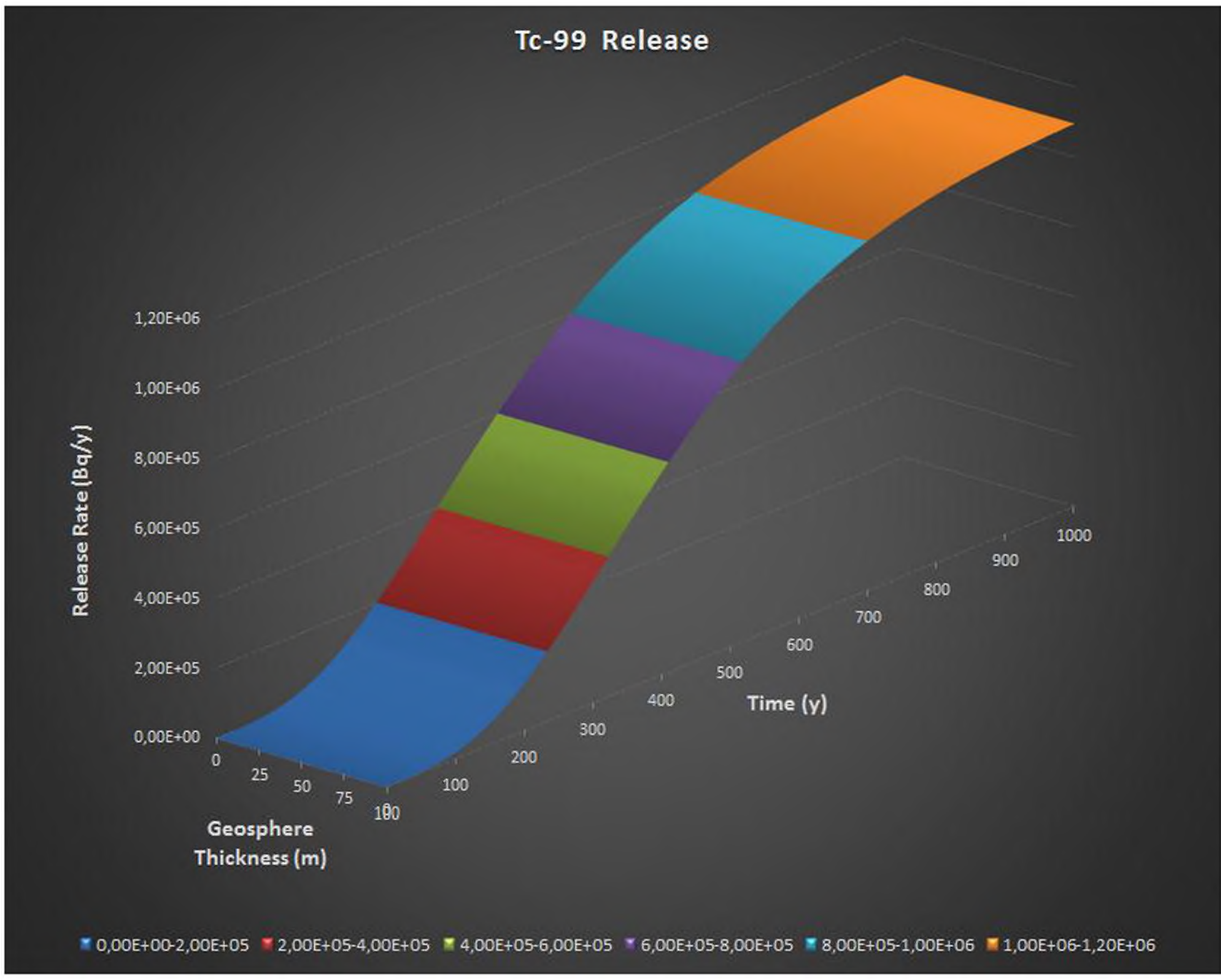

Figure 5. Release rate from the module of the repository to the biosphere for Tc-99.

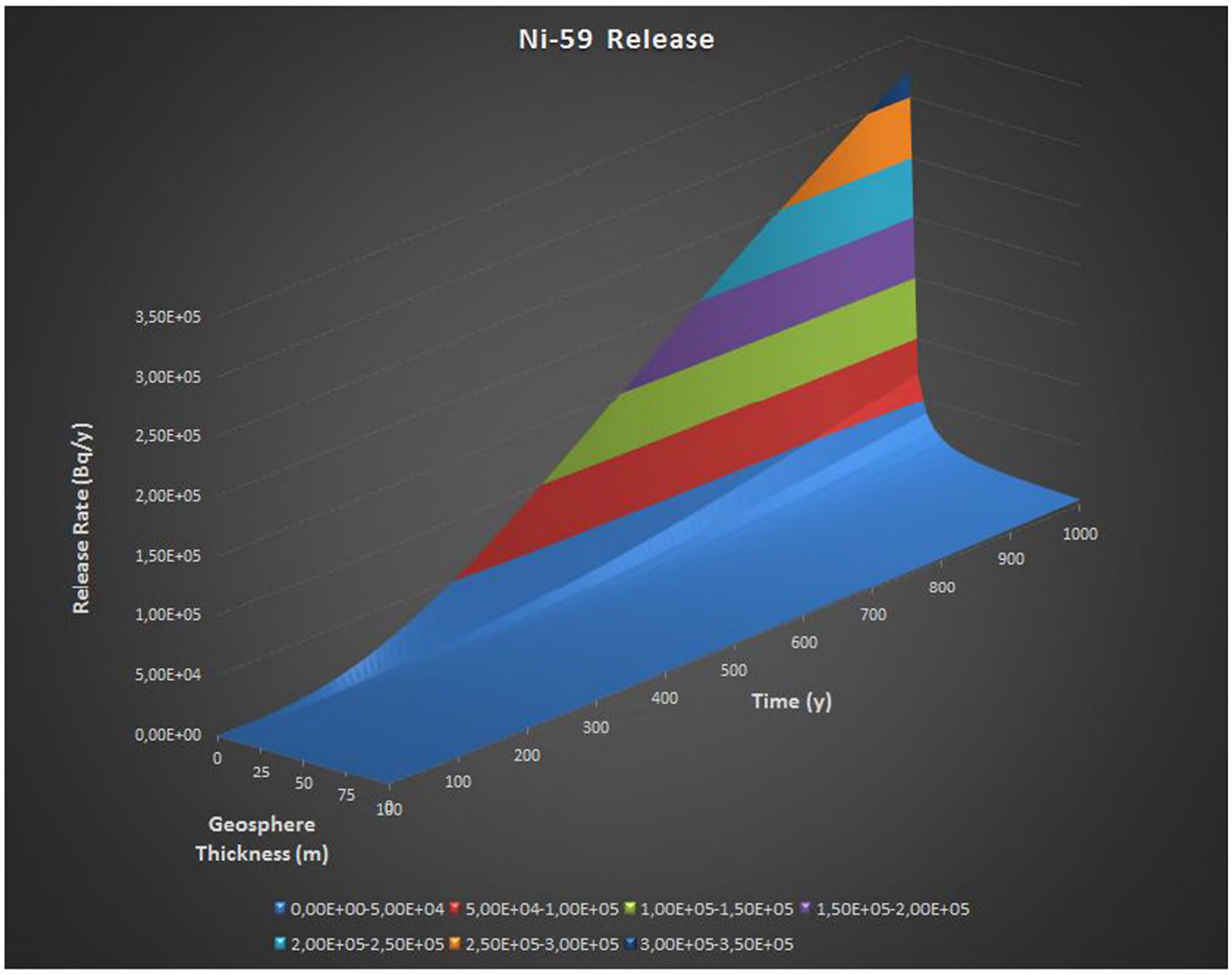

Figure 6. Release rate from the module of the repository to the biosphere for Ni-59.

Figure 7 shows the total release rate, which represents the contribution of all radionuclides listed in Table 1. The graph shape indicates the strong influence of the Cs-137 behaviour, due to the characteristics of this radionuclide, such as high initial activity, when compared to the other elements.

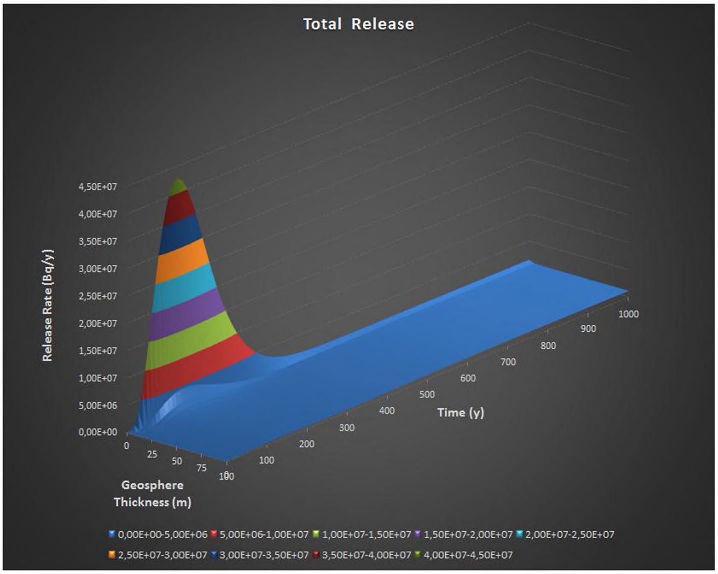

Figure 7. Total release rate from the module of the repository to the biosphere.

The relationship between the maximum release rate and the geosphere thickness is an important information in the process of choice of repository sites, in order to keep the released activity in acceptable levels over time, in accordance with legal requirements. This relationship is shown by Fig. 8, where is plotted the maximum release rate at any time in the 1000-year period in function of the geosphere thickness.

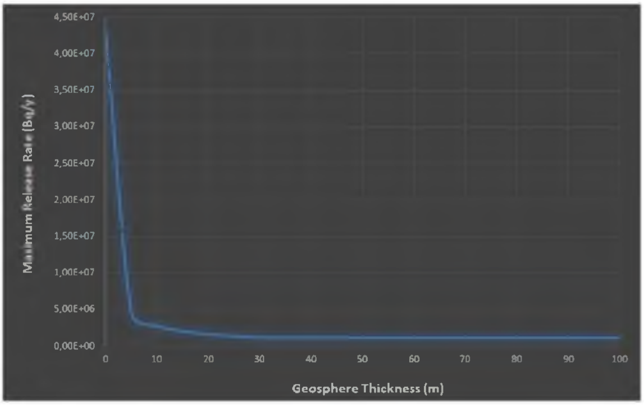

Figure 8. Relationship between the maximum release to the environment rate and the geosphere thickness.

4. CONCLUSIONS

The objective of this paper was to carry out a performance study regarding a near surface repository in terms of reliability engineering, particularly on the probability of failure of the engineered barriers and the geosphere, releasing radioactive waste to the environment. The most important radionuclides present in an intermediate level waste were target of this study. The influence of the particular behavior of each radionuclide allowed identifying the impact on the repository performance. As an important result, the knowledge about the relationship between the release rate and geosphere thickness is a very useful tool for choosing repository sites. As a suggestion, this work may be improved by carrying out, simultaneously, a study on performance optimization of the engineered barriers structure and the repository layout, as well as a sensibility analysis of the main parameters of the model.

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