

## COMPLEMENTARY SAFETY INDICATORS OF SALIGNY RADIOACTIVE WASTE REPOSITORY

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*Abstract.* This paper aims at presenting the geological aspects of the Saligny site necessary for developing the conceptual model of the repository as well as the associated mathematical model which describe the transfer of radio-nuclides from radioactive waste disposal system to the aquifer. The transfer time of some important radio-nuclides through the geological layers of the Saligny site, as a complementary safety indicator, is herein calculated. The transfer time of  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^{63}\text{Ni}$ ,  $^{59}\text{Ni}$  and  $^{99}\text{Nb}$  is derived from the time difference when a radionuclide reaches the highest value of concentration into two adjacent compartments. It is compared with the half-life of radio-nuclides in order to assess the role of geological layers of the site to delay the transfer of radio-nuclides in the surrounding environment.

*Key words:* radioactive waste, disposal, safety indicators, safety assessment.

### 1. INTRODUCTION

The low and intermediate level radioactive waste generated by the operation and decommissioning of Cernavoda Nuclear Power Plant are planned to be disposed into a new repository located on Saligny site. The repository will be a "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, a significant number of evaluations and laboratory tests have been done [1–3].

The disposal of radioactive waste needs to be carried out so as to provide an acceptable level of safety and, at the same time, to comply with the existing legal requirements. The fulfilment of the safety objective is demonstrated by applying a

safety assessment methodology in order to determine the safety indicators *i.e.* the parameters used to build arguments proving safety performance of the disposal system. Based on such indicators we may judge whether the disposal system is acceptably safe in terms of risk to human health and harm to the environment. Safety indicators are characteristics or consequences that can be measured or calculated, and compared to rigid or more loosely defined measures or ‘yardsticks’ in order to formulate such arguments [9].

Usually, the main safety indicators are the radiological dose or radiological risk. Taking into account the specific case of the Saligny repository inventory (low level waste with certain quantities of long-life radio-nuclides), the radiological dose does not seem the most appropriate safety indicator after a period of 10.000 years, determining us to consider some complementary indicators, mainly for the long term period of the repository. Therefore, we have chosen the transfer time of the radio-nuclides between the different compartments of the disposal system, better characterized in their half life, similar to those reported in [8].

Accordingly, in this paper we present the assessment of transfer time of both short ( $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ) and long ( $^{63}\text{Ni}$ ,  $^{59}\text{Ni}$ ,  $^{94}\text{Nb}$ ) life radio-nuclides into the geological layers of the Saligny site, within a normal evolution scenario developed for the repository.

## 2. DESCRIPTION OF SALIGNY SITE

Geologically, the Saligny site belongs to the Dobrogean part of the Moessic platform, placed in the south of Ovidiu – Capidava fault in the South-Dobrogea platform. The main characteristic of this area is the deep crystalline basement covered by thick sedimentary layers. Saligny site structure consists of a sequence of different geologic units: an Upper Pleistocene loessoid deposit, with silty and clayey loess, a Lower Pleistocene red clay, an Aptian - Sarmatian complex of clay, limestone and sands laying on Berriasian – Valanginian limestone with marls and sands intercalations, Jurassic marls with limestone intercalations, Paleozoic sediments and, finally, a crystalline basement of green slates [7].

The significant units for 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 complex (horizon D) and the berriasian-valanginian 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 contents [5].

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

### 3. INPUT DATA

The structural parameters of each geological unit of the Saligny site used for assessment of transfer time of radio-nuclides, such as dry density, water filled porosity and depth layer, are presented in Table 1 [4]. The distribution coefficients of radio-nuclides in the model compartments are presented in Table 2. As presented in reference [4], the values of distribution coefficients for  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  have been obtained experimentally on the samples collected from Saligny site for different horizons while the numerical values for the other radionuclides reproduced in Table 2 have been collected from literature [10, 11].

Table 1

Parameters of the disposal system compartments considered in the long term safety evaluation

Compartment	Depth [m]	Dry bulk density [kg/m <sup>3</sup> ]	Water filled porosity [-]	Hydraulic conductivity [m/yr]
Waste form	4.4	2300	0.15	0.31
Slab foundation	1	2500	0.15	0.31
Foundation ground	3	1760	0.32	1.58
Silty loess	1	1540	0.12	31.5
Clayey loess – a1	2	1780	0.26	6.31
Clayey loess upper Ib	8	1570	0.14	6.31
Clayey loess Iab2	2	1720	0.25	6.31
Clayey loess lower Ib	6	1690	0.25	6.31
Red clay	8	1760	0.32	1.58
Pre-quatarnary clay-1	5	1760	0.31	2.84
Sand lenses	5	1560	0.34	2.84
Pre-quatarnary clay-2	6	1760	0.31	2.84
Aquifer				
Berriasian-Valanginian limestones	15	1800	0.30	315

In the normal evolution scenarios of the Saligny site it is assumed an infiltration rate of about 20 mm/year for the natural ground [4]. In the first 300 years after the repository closure, the repository 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 about 10% from the natural infiltration rate and, after 500 years, the infiltration rate in the disposal structures is assumed to be as it is in the natural ground.

The initial activity of radio-nuclides considered in the evaluation are  $^{137}\text{Cs}$  (8.9E+14Bq),  $^{90}\text{Sr}$  (2.7 E+14Bq),  $^{63}\text{Ni}$  (9.2 E+11Bq),  $^{59}\text{Ni}$  (6.4 E+09Bq) and  $^{94}\text{Nb}$  (1.5E+10Bq), based on the information reproduced in reference [4].

Table 2

Distribution coefficients of the radionuclides considered in present work [ $\text{m}^3/\text{kg}$ ]

Element	Concrete	Silty loess	Clayey Loess	Red Clay	Pre- quaternary Clay	Sand lenses	Aquifer
Cs	0.02	0.774	1.131	4.131	2.366	0	1
Ni	0.1	0.6	0.6	0.6	0.6	0.4	1
Nb	0.5	1	1	1	1	0.55	1
<b>Sr</b>	0.001	0.006	0.011	0.012	0.012	0	0

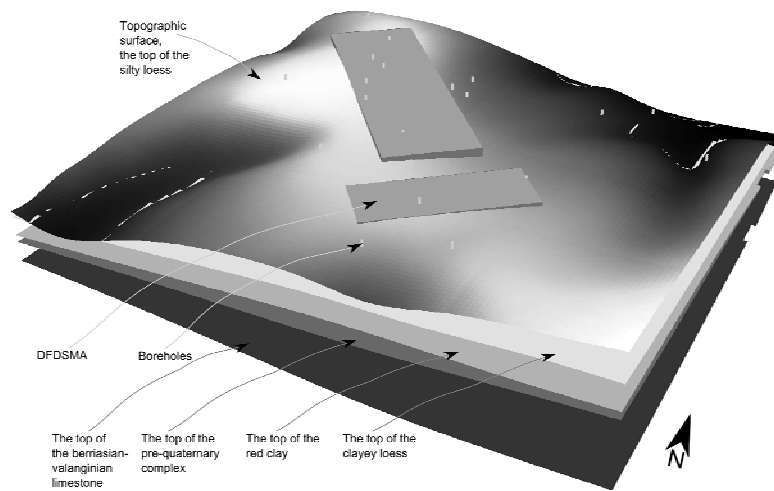


Fig. 1 – Conceptual model of the Saligny disposal system.

#### 4. CONCEPTUAL AND MATHEMATICAL MODEL

The conceptual model of Saligny repository has been developed using the interaction matrix method in accordance with the evaluation requirements of the AMBER computer code. The disposal system was split into a series of assumed homogenous compartments and the transfer processes between the compartments were expressed as transfer coefficients that represent the fraction of the activity in a particular compartment transferred from that compartment to another one, per unit time. The conceptual model of the Saligny disposal facility considered in the assessment within 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, and they are mentioned as DFDSMA in figure. The lower ones are geological layers of Saligny site which consist of silty loess mentioned in the figure as topographic surface, clayey loess, Quaternary red clay, Pre-quaternary complex and berriasian-

valanginian limestone. The unsaturated zone is represented by the silty and clayey loess, the Quaternary red clay and the upper part of the Pre-quaternary complex, while the saturated zone includes the lowest compartments of Pre-quaternary complex as well as the berriasian-valanginian limestone.

The mathematical model developed in order to assess the distribution of 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 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  $i$  compartment, the rate at which the inventory of radio-nuclides in a compartment changes in time is given by:

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

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  $y^{-1}$ ].  $\lambda_N$  is the decay constant for radionuclide  $N$  [ $y^{-1}$ ] and  $\lambda_{ji}$  and  $\lambda_{ij}$  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 version 5 of AMBER computer code. The dispersion was considered by the discretisation of the disposal facility compartments.

Radioactive decay is represented through the decay rate ( $\lambda$ , in  $y^{-1}$ ), which is given by the equation  $\lambda = \ln 2 / t_{1/2}$ , where  $t_{1/2}$  is the half-life of the radionuclide ( $y$ ).

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 (2):

$$R = 1 + \frac{\rho \cdot k_d}{\vartheta_w} \quad (2)$$

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

For the unsaturated transfers, the advective transfer rate of radio-nuclides ( $\lambda$  flow, in  $y^{-1}$ ) is given by equation (3):

$$\lambda_{flow} = \frac{q}{L \cdot \vartheta_w \cdot R}, \quad (3)$$

where  $q$  represents the annual flow rate through the compartment [ $\text{m}\cdot\text{y}^{-1}$ ],  $L$  represents the length of the compartment in the direction of the water flow (m),  $\vartheta_w$  represents the water-filled porosity of the compartment (dimensionless), and  $R$  represents the retardation of the compartment (dimensionless). The parameter  $q$  [ $\text{m}\cdot\text{y}^{-1}$ ] represents the infiltration rate of water through the repository compartments and geosphere (unsaturated zone). The infiltration rate,  $q$  [ $\text{m}\cdot\text{y}^{-1}$ ], is experimentally determined and it is a site specific parameter.

For the saturated zone, the water infiltration rate is given by equation (4):

$$q = K \frac{\partial H}{\partial x}, \quad (4)$$

where  $K$  represents the hydraulic conductivity of the compartment [ $\text{m}\cdot\text{y}^{-1}$ ],  $\partial H/\partial x$  represents the hydraulic gradient (dimensionless) and  $x$  is flow direction of infiltration water.

## 5. RESULTS AND DISCUSSIONS

In the present paper, the transfer time is defined as the time period necessary for a radionuclide to transit a disposal system compartment. The transfer time cannot 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 last moment when the radionuclide leaves the compartment. Due to this, it is not possible to measure or to calculate that period of time for each radionuclide, and, because the radionuclide migration process is considered as a continuous one, the transfer time is calculated based on the variation of radionuclide concentration in the disposal system compartments, taking into account the moments when the radionuclide concentration reaches the peak values in two adjacent compartments.

As examples, to illustrate the transfer time calculation, the time variation of the  $^{137}\text{Cs}$  concentration in the lower part of the foundation ground and silty loess, as well as the time variation of the  $^{14}\text{C}$  concentration in the silty loess and clay loess are presented in the Figs. 2 and 3.

Based on the results presented in Fig. 2, it is possible to calculate the moments when the radionuclide concentration in each compartment reaches minimum initial values, the peak values and the minimum final values. Thus, the transfer time can be defined as the difference between the moments when the peak values are reached, for each compartment presented in Fig. 1.

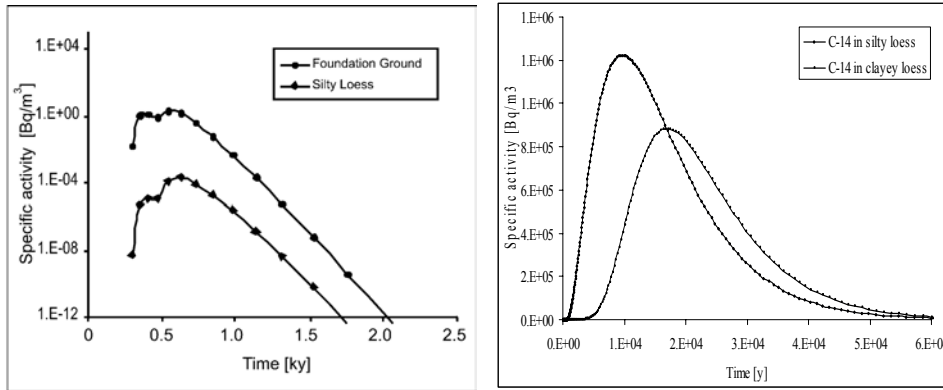


Fig. 2 – Time variation of  $^{137}\text{Cs}$  and  $^{14}\text{C}$  specific activity in different geological layers.

Figures 3, 4, 5 and 6 present the transfer times ( $x$  axis, years) for the  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ ,  $^{59}\text{Ni}$ ,  $^{63}\text{Ni}$  and  $^{94}\text{Nb}$  in the geological layers of Saligny site, calculated in the assumptions of the normal evolution scenario of the Saligny repository. The depth under the repository foundation represents  $y$  axis (metres).

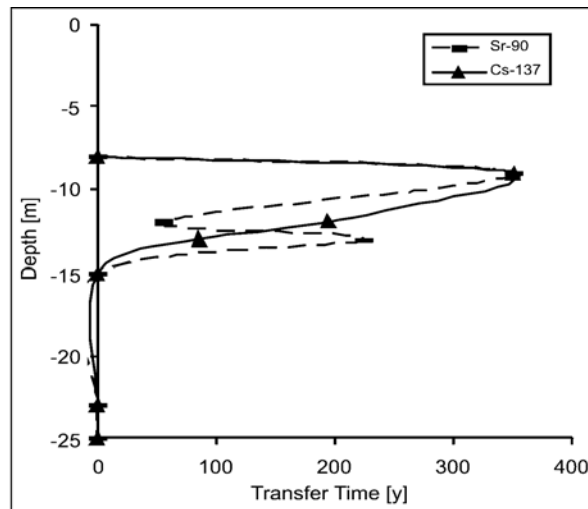


Fig. 3 – The transfer time functions of  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  vs. depth.

Due to the short lived radionuclides  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ , the contaminant plume is extended only few meters under the repository foundation. Then, their transfer time can be calculated only for the upper part of the Saligny site layers. As is presented in Fig. 3, their highest transfer time is of 351 years corresponding to the silty loess. Comparing the transfer time values obtained for the first two geological layers under the waste form with the half-life period of these isotopes, which is

30 years and 29.1 years respectively, it can be observed that these layers have a barrier function for  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ .

The transfer time values for  $^{59}\text{Ni}$  isotope vary between  $3.16\text{E}+03$  years and  $2.5\text{E}+05$  years. Comparing the transfer time values obtained for each layer with the half-life of this isotope, which is  $7.54\text{E}+04$  years, it can be observed that the geological layers of Saligny site have a barrier function for  $^{59}\text{Ni}$ . The total transfer time is of  $1.1\text{E}+06$  years, two magnitude order higher than the half life of this isotope, thus emphasizing the role of geological layers of Saligny site to retard the migration of  $^{59}\text{Ni}$ .

The transfer time values for the isotope  $^{63}\text{Ni}$  isotope vary between 150 years and 630 years. By comparing the transfer time values obtained for each layer with the half-life period of this isotope, which is 96 years, it can be noticed that the geological layers of Saligny site have a barrier function for  $^{63}\text{Ni}$ . The highest value of transfer time is noted in the slab foundation emphasizing the affinity of  $^{63}\text{Ni}$  on cemented based materials. The total transfer time is of 2730 years, about 30 times higher than half life, thus emphasizing the role of geological layers of Saligny site to retard the migration of  $^{63}\text{Ni}$  as well as to allow the natural decay in first 15 meters deep under the repository. Actually, the distribution coefficient is dependent only on the chemical element, subsequently, the two isotopes of nickel having the same behavior in the model compartments. However, as a consequence of the different half life of the two isotopes, they will reach the peak value in a certain compartment at distinct time moments. Therefore, we can note the different form of the transfer time variation of the two isotopes of nickel, as presented in the Figs. 4 and 5.

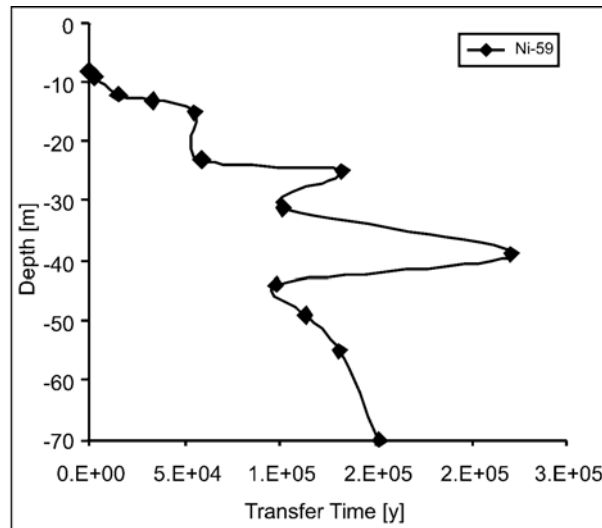


Fig. 4 – The transfer time functions of  $^{59}\text{Ni}$  vs. depth.

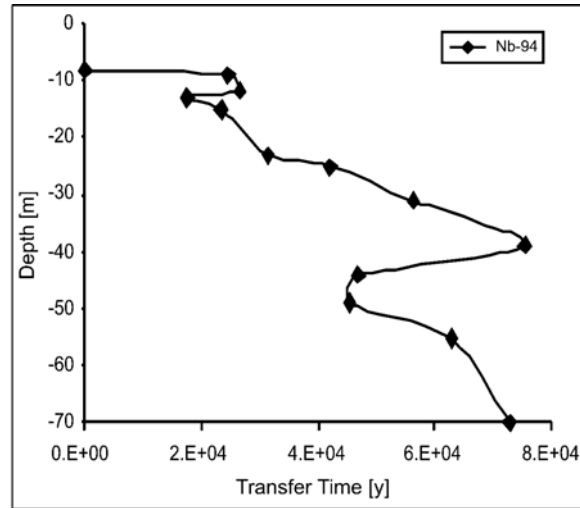


Fig. 5 – The transfer time functions of  $^{63}\text{Ni}$  vs. depth.

The transfer time values for  $^{94}\text{Nb}$  vary between  $1.7\text{E}+04$  years and  $7.5\text{E}+04$  years. Comparing the transfer time values obtained for each layer with the half-life period of this isotope, which is of  $2.03\text{E}+04$  years, it can be observed that the geological layers of Saligny site have a barrier function for  $^{94}\text{Nb}$ . The highest value of transfer time is noted in the red clay. The total transfer time is  $5.02\text{E}+5$  years, about 25 times higher than half life, emphasizing the role of geological layers of Saligny site to retard the migration of  $^{94}\text{Nb}$  as well as to allow the natural decay.

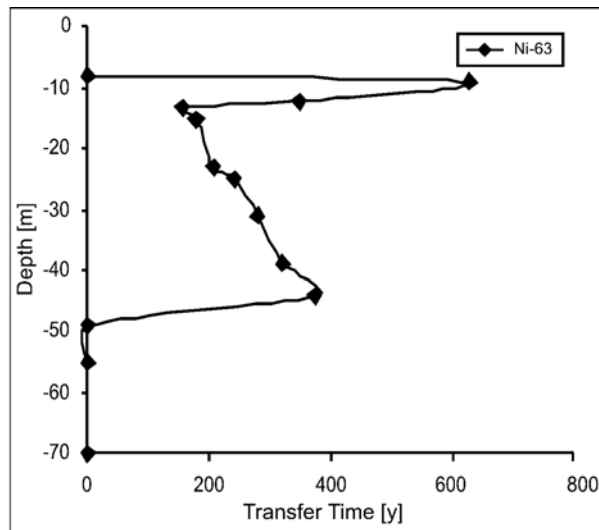


Fig. 6 – The transfer time functions of  $^{94}\text{Nb}$  vs. depth.

## 6. CONCLUSIONS

The transfer time values for  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^{63}\text{Ni}$ ,  $^{59}\text{Ni}$  and  $^{94}\text{Nb}$  were evaluated as complementary safety indicators for the Saligny site. Comparing the transfer time values obtained for each layer with the half-life period of these isotopes, it can be observed that the Saligny site geological layers have a barrier function. Moreover, comparing the total transfer time for each radionuclide through the geological layers of Saligny site with its half-life period, we can note the role of geological layers of Saligny site to retard the migration of the analyzed isotopes.

## REFERENCES

1. C. Bucur, M. Olteanu, *Determination of Diffusion and Distribution Coefficients used in the Radionuclide Transport Program*, SIEN, Bucharest, October, 2003.
2. O. Niculae, D. Dogaru, G. Jinescu, O.G. Dului, *Influence of the site properties on the long term evolution scenarios for Saligny Near Surface Radioactive Waste Repository*, National Conference of Physics, 10–13 September, 2008, Magurele, Romania.
3. D. Dogaru, O. Niculae, Ghe. Jinescu, O.G. Dului, *The influence of the chemical properties of the geological formations on the migration parameters of the radionuclides from Saligny repository*, Scientific Study & Research – Chemistry & Chemical Engineering, Biotechnology, Food Industry, **9**, pp. 153–168, 2008.
4. \*\*\* *Safety Report for Siting a near Surface Repository at Saligny site*, Internal Report no. 1547/2007, SITON, Magurele, 2007.
5. C. Bucur, I. Anghel, S.D. Ware, M. Pavelescu, *The importance of geochemical characterization of repository host horizons for radioactive waste disposal: Saligny repository site for L/ILW, Romania*, The 9<sup>th</sup> International Conference on Radioactive Waste Management and Environmental Remediation, Oxford, September 2003.
6. Jin Beak Park, Joo, Wan Park, Eun, Young Lee, Chang, Lak Kim, *Experiences from the source, term analysis of a Low and Intermediate Level Radwaste Disposal Facility*, WM'03 Conference, February 23–27, Tucson, Arizona, USA, 2003.
7. M. Stoica, *Ostracode purbeckiene din Dobrogea de Sud*, Ph.D. Thesis, University of Bucharest, Faculty of Geology and Geophysics. Bucharest, 2003.
8. D. Dogaru, O. Niculae, M. Terente, G. Jinescu, O.G. Dului, *Estimation of the transfer time of radionuclides in geological layers of Saligny site*, NUCLEAR 2009 Conference, May 27–29, 2009, Pitesti, Romania.
9. IAEA-TECDOC, *1372 Safety indicators for the safety assessment of radioactive waste disposal*, IAEA, Vienna, 2003.
10. IAEA-TECDOC, *Improvement of Safety Assessment Methodologies for Near Surface Disposal Facilities*, Volume I: “Review and Enhancement of Safety Assessment Approaches and tools”, Volume II: “Test Cases”, Vienna, 2004.
11. W. Ashton, J. Sumerling, *Biosphere Database for Assessments of radioactive waste Disposals*, UK Department of the Environment Report DoE/RW/88/083, 1988.