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THE MIGRATION OF THE RADIONUCLIDE Am-241 IN UNSATURATED SOIL FROM SALIGNY AREA*

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Abstract. The functioning of the Cernavodă Nuclear Power Plant will generate low and medium active waste which will be contaminated with long-life fission products (U, Pu, Np, Am), radioactive carbon (C-14) and tritium (H-3), which through their radiochemical characteristics and their influence upon the environment and people, request special attention regarding their storage and disposal. Based on the geological and mineralogical researches regarding the location of a repository for low and medium active waste, Saligny area near the Cernavodă Nuclear Power Plant was chosen. The repository will be located in loess, seated on sedimentary formations with insertions of clay patches. The main target of the research is to obtain some experimental data necessary for the evaluation of the migration of the radionuclide Am-241 (resulted from Cernavodă Nuclear Power Plant) in unsaturated soils in Saligny area, which might be the host of the Final Repository for Low and Medium Active Waste. From the analysis of the test data obtained in the laboratory for the determination of the migration parameters of the radionuclide Am-241 in the material of the geological formation of Saligny area it results that there is a direct correlation between the values of these parameters and the basic mineralogical component – clay – of the soil sample.

Key words: soil, groundwater, radionuclide, migration.

1. INTRODUCTION

The research shows the main characteristics of the soil, of the groundwater and the physical and chemical properties of the radionuclides from the radioactive waste, that must be studied to obtain basic data for the evaluation of the long–term security of Final Repository For Low and Medium Active Waste. The main characteristics of the three elements (soil, groundwater, radionuclide) are shown taken into account a repository security evaluation [1, 9, 10].

The main physical and chemical characteristics of the soil influencing the migration of the radionuclides are: granulometry, mineral component, density, porosity, permeability, natural humidity and the pH [2].

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A good soil for the location of a repository for radioactive waste must have high capacity for ionic exchanges, small porosity and lowest permeability. In some cases of radionuclides (Sr, Cs) the contents of natural carbonate can be beneficial leads to a great inconvenient for actinides, which form soluble composites with carbonated ions [3].

The physical and chemical characteristics of the groundwater that influence the migration of radionuclides in geological patches are: temperature, hardness, pH, electroconductibility, content of dissolved salts and the concentration of inorganic dissolved ions [4].

The main parameters of transport that influence the migration velocity of Am-241 in soil are the pH of groundwater, the chemical species of radionuclide and the mineral composition of unsaturated soils [5, 6].

2. EXPERIMENTAL TECHNIQUES

For the evaluation of the process taking place between dissolved radionuclides and the surface of the hosting rock, in experimental researches made in laboratory two techniques are used: static ("in batch") and dynamic (on column).

The static technique consists in contacting a solution in which the radionuclide (radionuclides) of interest is dissolved and an illustrative sample of hosting-rock preliminary processed. In these conditions only the physical and chemical processes are evaluated that compete to the radionuclides retention (sorption) on the constitutive minerals surface of the rocks [5, 6].

These processes can be:

- physic sorption, a quick and irreversible process influenced by solution's pH;
- chemical sorption, an electrostatic attraction process, when the radionuclides are in cation form and the surface of the solid is loaded with negative electric charge;
- chemical substitution, when a component from the structure of the solid phase is replaced by a component of the liquid phase.

This process of radionuclides sorption on a solid surface, is numerically evaluated through the distribution coefficient K_d that represents the ratio between the concentration of the radionuclide retained on the solid phase and its concentration in the solution [7].

The dynamic technique consists in passing a solution, loaded with the interested radionuclide (radionuclides), with a certain velocity through a column loaded with geological material, previously treated. This method has the inconvenient that the slitting phenomenon of the solution flowing through the column can occur, the result being a short time of contact between the two phases, obtaining the retaining coefficients lower than those obtained in the static technique, where the contact period (reaction) is longer [5, 6].

Both techniques have inconvenients and they give extreme values in comparison with the real conditions existing in the geological material, because the geological material used in the test is disturbed and does not reflect the physical and chemical characteristics of the horizon from the boring kernel sampled on.

Using undisturbed boring kernel to determine the distribution coefficients of the radionuclides is a technique that gives closer results in comparison with the results which can be obtained in the field, because thus the physical, lithological and structure characteristics of the geologic patch are generally kept.

3. THE EXPERIMENTAL STUDY OF THE MIGRATION OF THE RADIONUCLIDE AM-241

To evaluate the distribution and retardation coefficients, the migration velocity and the migration time for Am-241 laboratory tests were made in static system ("in batch" technique). The tests were made in certain working conditions, taking into account the physical and chemical characteristics influence of the three elements that interferes: soil, groundwater and the radionuclide [8].

The migration of Am-241 in the unsaturated soils intercepted by Saligny Drilling-15 has been experimentally studied using as standard source for Am-241 a solution of $AmCl_3$ with an measured activity Λ_{et} = 15.341 μCi / l and an pH = 3.

The algorithm of the experimental study was the following:

1. For each horizon, the horizon water is prepared by contacting the representative boring kernel with demineralised water, the ratio between the volume of the liquid phase and the mass of the solid phase being V/M = 2/1 [ml/g]. The contacting time was 30 days.

2. After expiry of 30 days of contacting period, for each horizon the liquid phase and the solid phase of the contacting sample is separated by filtering and the liquid phase is retained, representing the horizon water.

3. Each horizon water is contacted by solution of $AmCl_3$, obtaining a dilution of 1:10.

4. The pH of the dilution is corrected, using a solution of NaOH with a concentration 1 M, to obtain a pH = 7.1–8.6 coresponding to the pH values of the horizon waters.

5. For each horizon the dilution is divided into two equals volumes representing the blind sample and the contacting sample.

6. For each horizon, by α spectrometry, the activity Λ_0 of the blind sample is measured.

7. For each horizon the contacting sample is contacted with soil using "in batch" method, the ratio between the volume of the liquid phase and the mass of the solid phase being V/M = 10/1 [ml/g]. The contacting time was 30 days.

8. After expiry of 30 days of contacting period, for each horizon the liquid phase and the solid phase of the contacting sample is separated by filtering and the liquid phase is retained.

9. For each horizon, by α spectrometry, the activity Λ of the contacting sample is measured.

10. The mathematical parameters that define the migration of the Am-241 in the unsaturated soil are calculated using the following relations:

$$K_d = \frac{\Lambda_0 - \Lambda}{\Lambda} \cdot \frac{V}{M}$$

where:

K_d [ml/g] – the distribution coefficient;
Λ_0 [Bq] – the primary activity of the radionuclide in the liquid phase;
Λ [Bq] – the final activity of the radionuclide in the liquid phase;
V [ml] – the volume of the liquid phase;
M [g] – the mass of the solid phase.

$$R = 1 + \frac{\rho}{\eta} \cdot \mathrm{K}_d$$

where:

R – the retardation factor ;
ρ [g/cm^3] – the density of the geological patch;
η [cm^3/cm^3] – the porosity of the geological patch;
K_d [ml/g] – the distribution coefficient.

$$v_a = \frac{k \cdot i}{\eta}$$

where:

v_a [cm/year] – the migration velocity of the groundwater;
k [cm/year] – the permeability of the geological patch;
i [cm/cm] – the hydraulic gradient of the geological patch;
η [cm^3/cm^3] – the porosity of the geological patch.

$$t_a = \frac{d}{v_a}$$

where:

t_a [years] – the migration time of the groundwater;
d [cm] – the distance between the contaminating source of the geological patch and the measuring point;
v_a [cm/year] – the migration velocity of the groundwater.

In practice, for the calculation of the migration time of the groundwater t_a, the distance between the contaminating source of geological patch and the measuring point is considered $d = 1$ cm, therefore the last formula becomes:

$$t_a = \frac{1}{v_a}$$

$$v = \frac{v_a}{R}$$

where:

v [cm/year] – the migration velocity of the radionuclide;
v_a [cm/year] – the migration velocity of the groundwater;
R – the retardation factor.

$$t = t_a \cdot R$$

where:

t [years] – the migration time of the radionuclide;
t_a [years] – the migration time of the groundwater;
R – the retardation factor.

4. TEST RESULTS

The values of the parameters characterising the migration of the radionuclide Am-241 in the unsaturated soils intercepted by Saligny Drilling-15, are presented in the Table 1.

In the case of soils intercepted by Saligny Drilling-15, the pH of the groundwaters on horizons is very litle changed, being in the range of low acidity. In this situation, all chemical species of the radionuclide Am-241 (all the states of oxidation AmIII, AmIV, AmV and AmVI) have a good sorption in the clay patches intercepted by the drilling [6]. Because of that, the mineral composition of unsaturated soils remains the main parameter of transport that influences the migration of Am-241 in soil.

The main factor the Am-241 migration in the soils intercepted by Saligny Drilling-15 depends on the content in clay minerals of the soils, especially of

Table 1

The test results with Am-241

Horizon	**Type of soil**	**Depth** [m]	K_d **[ml/g]**	R	V_a **[cm/year]**	$t_a \times 10^3$ **[years]**	V **[cm/year]**	$t \times 10^3$ **[years]**
A	dusty loess	3.4÷15.0	4.012	16.695	769.207	1.300	46.074	21.704
B	clayish loess	15.0÷32.1	4.249	22.438	409.633	2.441	18.256	54.771
C	red and brown clay	32.1÷40.0	4.893	25.934	128.446	7.785	4.953	201.896
D	clayish and sandy deposits	40.0÷69.5	8.901	57.979	11.280	88.652	0.195	5139.838

smectit (montmorillonit), that have the greatest capacity of cation exchange, so it best retains the radionuclide.

The content of clay minerals increases with depth, the greatest concentration being in the depth interval of 34÷43 m, where brown clay with the greatest content of smectit appears. This depth interval corresponds to the lower patch of C-horizon and to the higher patch of D-horizon. In this way we explain the growth with the depth of the distribution coefficient K_d values and of the retardation coefficient R. These can be observed in Fig. 1 and Fig. 2.

The migration velocity *V* and the migration time *t* of Am-241 vary in direct proportion, respectively in inverse proportion, with the content of clay minerals of each horizon. It is remarkable that for D-horizon there are the smallest value for migration velocity *V* and the greatest value for migration time t, because of the very high content of clay minerals in this horizon, especially smectit that indicates this horizon as the geological medium with the best retention capacity for radionuclide. These can be observed in Fig. 3 and Fig. 4.

5. CONCLUSIONS

From the analysis of the test data the following conclusions arise:

- The main factor the Am-241 migration in unsaturated soils intercepted by Saligny Drilling-15 depends on the content of clay minerals, especially smectit, as the main parameter of transport that characterises the retention capacity for radionuclide in the geological material.

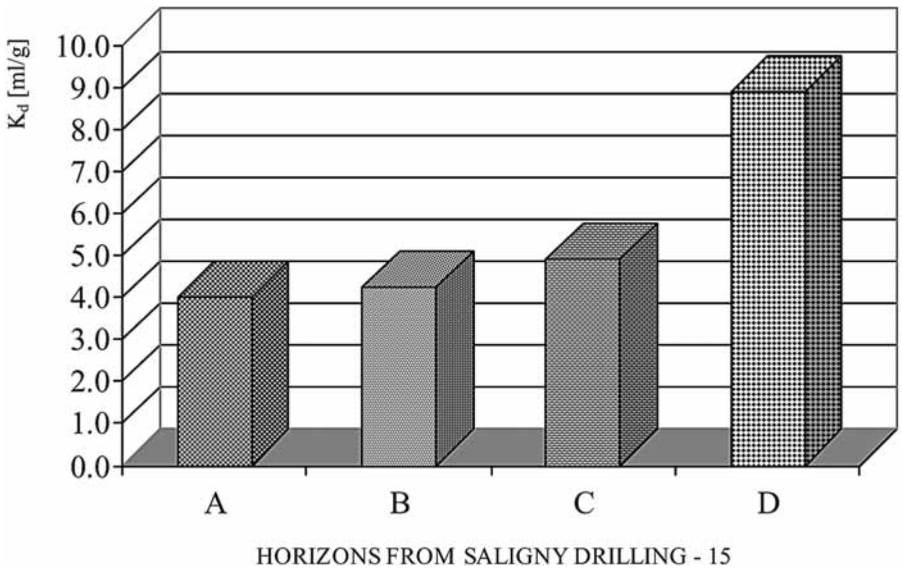

Fig. 1 – The values of the distribution coefficient K_d for Am-241.

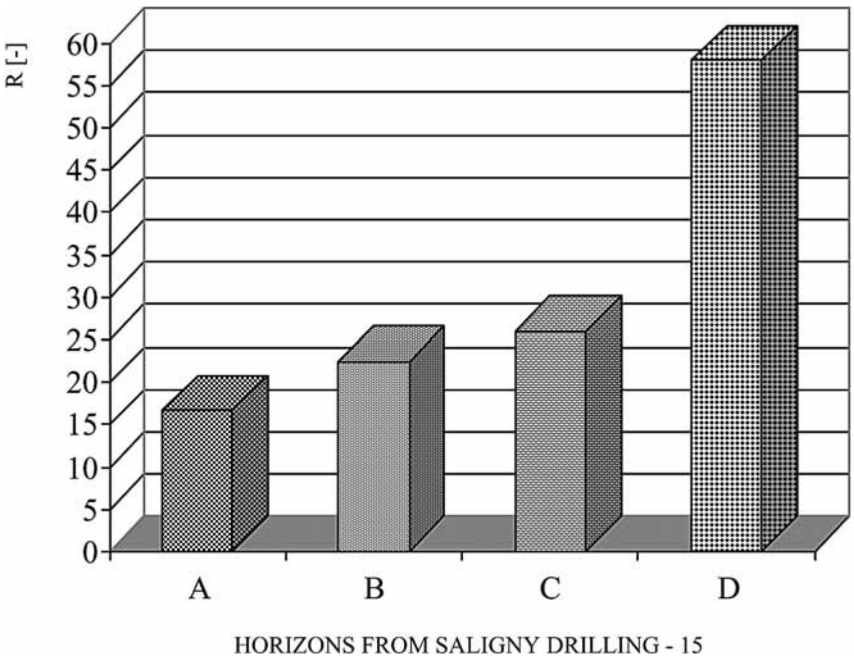

Fig. 2 – The values of the retardation coefficient *R* for Am-241.

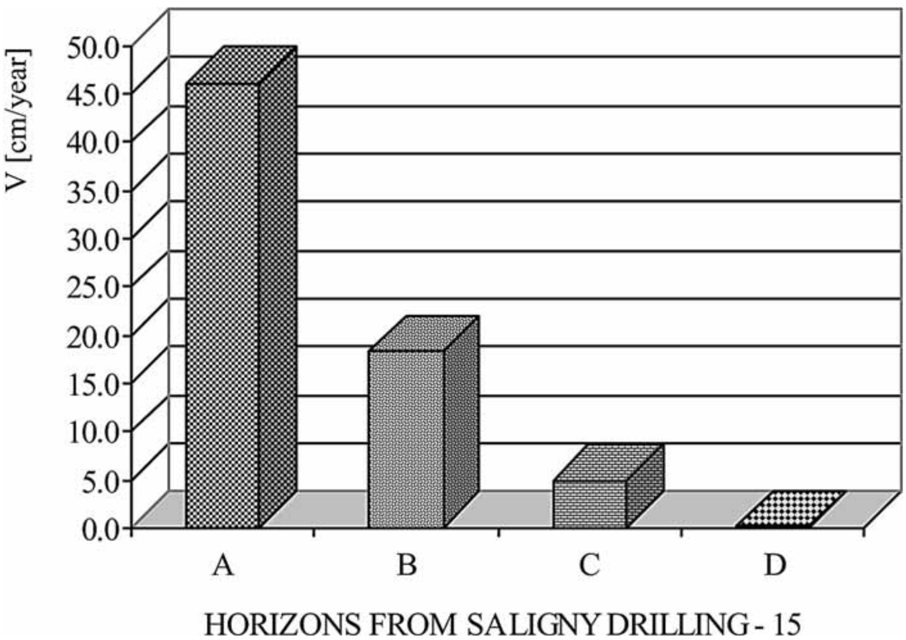

Fig. 3 – The values of the migration velocity *V* for Am-241.

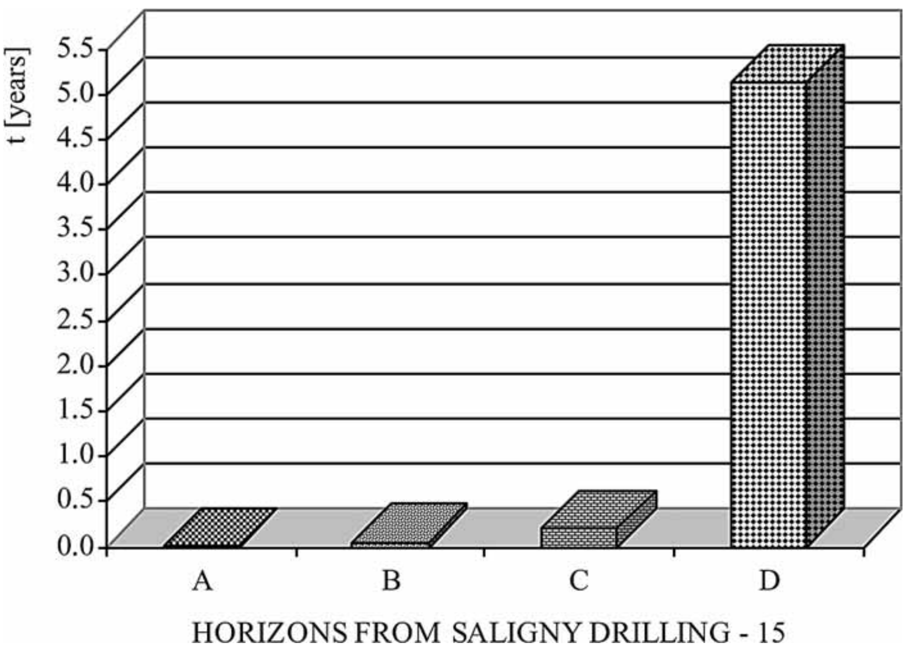

Fig. 4 – The values of the migration time t for Am-241.

- The parameters of migration of Am-241 are ordered as follows:
  – $K_d(A) < K_d(B) < K_d(C) < K_d(D)$
  – $R(A) < R(B) < R(C) < R(D)$
  – $V(D) << V(C) < V(B) < V(A)$
  – $t(A) < t(B) < t(C) << t(D)$.

The analysis of the resulted migration parameters for the soils intercepted by Saligny Drilling-15 indicates a proper potential of the repository location in Saligny area to constitute a natural obstacle against radionuclides migration.

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