RADIONUCLIDE DIFFUSION IN GEOLOGICAL MEDIA

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Radionuclides can be transported through geological media by advection and diffusion, diffusion being the dominant transport mechanism in the very low permeable media. This paper performs an evaluation of the diffusion coefficients for Cs-137 and Co-60 (the main radionuclides in the L/ILW) in Saligny site specific horizons.

Key words: radioactive waste, diffusion, radionuclide, permeability, porosity, loess, clay, multivariate regression.

INTRODUCTION

The Saligny Site, situated near the Cernavoda Nuclear Power Plant in Romania, has been chosen as a repository site for placement of future Low and Intermediate Level Radioactive Waste (L/ILW). Its proximity to Cernavoda NPP, dry climate conditions, low water infiltration rates, and favourable geochemical properties exhibited by subsurface geologic horizons, are characteristics that make Saligny a suitable site for L/IL radioactive waste disposal in Romania.

The LIL wastes generated in a nuclear power plant contain a variety of radionuclides; almost all of the disposed activity is found in relatively short-lived radionuclides, including Cs-137, Co-60, Sr-90, H-3, and Fe-55. Diffusion coefficients were determined for two of these radionuclides, Cs-137 and Co-60.

Lithological investigations of the subsurface geologic horizons for the Saligny site indicate the presence of a large loess formation deposited in the quaternary geologic era. Fig. 1 provides the stratigraphy of the Saligny site [1].

The large loess formation is divided into a silty loess (horizon A) and a clayey loess (horizon B) deposited over a red quaternary clay formation (horizon C). A thin layer of consolidated loess was found inside of A and B horizons. Below the red clay horizon is a pre-quaternary clay formation (horizon D), a very heterogeneous horizon comprised of discontinuous lenses of sand, silt, clay, gravel, and carbonates, that lays on the Barremian limestone formation (horizon Br).

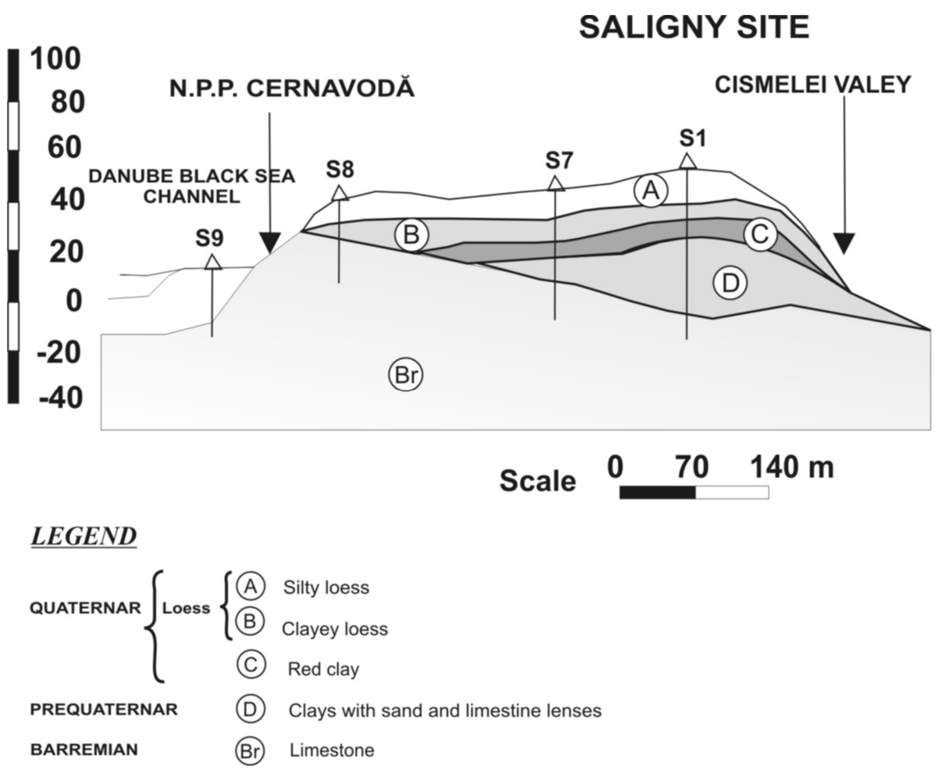

Fig. 1. Stratigraphy of the Saligny Site.

The first three horizons (A, B, and C), in which the saturation is between 0.2 and 0.8, belong to unsaturated zone. The saturation in upper part of D horizon is smaller than 1, but in the lower part the send lances are filled with water (saturation 1). From this reason, the lower part of the pre-quaternary clay formation is considered as the first saturated zone.

The water table is 40 to 45m below the surface part. The main aquifer is found in the horizon E, being connected to the neighboring Danube River.

Table 1 presents the main hydrological parameters for the studied soil samples [2].

Table 1

Hydrological Parameters for the Studied Geological Horizons

Geological Unit	**Average density, ρ kg/m^3**	**Average permeability, k [m^2]**	**Average porosity, n [%]**
Loess	1540	7.72E-12	42.33
Clayey loess	1631	4.98E-12	38.79
Consolidated loess	1783	7.52E-13	36.10
Red clay	1766	4.98E-13	32.82
Quaternary clay	1760	9.05E-13	33.31

RADIONUCLIDE TRANSPORT BY DIFFUSION

Contaminants dissolved in groundwater are transported by two mechanisms: diffusion and advection. By diffusion, contaminants move in the direction of the concentration gradient, meaning from the areas of higher concentration to lower concentration. Diffusion will occur as long as a concentration gradient exists, even if the groundwater is not flowing. This process may be the major factor in mass transport in geological materials of very low permeability.

Contaminants are also transported by advection also known as convection. This occurs as the flowing groundwater carries the dissolved solutes with it. At the scale of a few pore diameters, groundwater will move parallel to the flow path with different rates due to differences in the pore size. This causes the contaminant plume to spread along the direction of the flow path, process called longitudinal dispersion. Because the flow path diverges around mineral grains, the contaminant plume will also spread laterally, process known as traverse dispersion. At the laboratory column scale, the movement of a contaminant through a uniform porous media can be described by advection-dispersion equation, which account for advection, diffusion, and pore-scale dispersion.

The mass of fluid diffusing is proportional with concentration gradient (Fick's first low - empirical); in one dimension, Fick's first low is:

$$J_x = -D\frac{\partial C}{\partial x} \tag{1}$$

where: J – mass flux of contaminant per unit area per unit time, [kg/m^2·s]
D – diffusion coefficient, [m^2/s]
C – contaminant concentration, [kg/m^3]

The Fick’s second low is applicable for systems where the contaminant concentrations are changing with time; in one dimension, the Fick’s second low is:

$$\frac{\partial C}{\partial t} = D\left(\frac{\partial^2 C}{\partial x^2}\right) \tag{2}$$

The Fick’s lows refer only to diffusion in homogeneous medium such as water; when dealing with diffusion through a fluid in a heterogeneous system such as groundwater in a porous rock, the Fick’s low have to be modified to take account of the fact that the water occupies only a fraction of the total volume occupied by the rock. Diffusion in porous media is affected in different ways by the geometry of porous structure and by the contaminant interaction with the pore walls. To account for these effects, an effective diffusion coefficient, D^*, must be used to describe the contaminant diffusion in porous media [3].

$$D^* = \omega D \tag{3}$$

$$\omega = n_e/\tau^2 \tag{4}$$

where: ω – is a coefficient that is related to the tortuosity
n_e – effective porosity (open and interconnected pores)
τ – tortuosity – is a measure of the effect of the shape of the flow path followed by water molecules in a porous media

For the contaminants that are sorbed on mineral surfaces of the porous media, the net rate of diffusion will be smaller than for species with no sorption; for the contaminant with sorption, the apparent diffusion coefficient is defined by:

$$D_a = \frac{D^*}{R} \tag{5}$$

The transient diffusion transport of radionuclide in a porous medium is described by Fick’s second low:

$$\frac{\partial C}{\partial t} = \frac{D^*}{R}\left(\frac{\partial^2 C}{\partial x^2}\right) \tag{6}$$

where: C = contaminant concentration, [kg/m^3]
D =diffusion coefficient, [m^2/s]
R = retardation factor (1 for nonsorbing solutes),
x = distance, [m]
t = time, [s]

DIFFUSION EXPERIMENTS

Diffusion experiments on soils sampled from the main horizons characteristic to the Saligny site were performed.

The diffusion coefficients in soil were determined using an experimental setup made up of two stainless steel cups one filled with a soil in equilibrium with a tracer solution mix and the other with a soil sample saturated only with Saligny simulated water. The average well water chemical composition at Saligny, measured over a period of 4 years, showed that the dominant cations are Na^+ (150.4 mg/L) and Ca^{2+} (68.8 mg/L) while the anions are SO_4^{2-}(212.3 mg/L), Cl^-(240.9 mg/L), HCO^{3-} (351.1 mg/L) [4].

The concentration of ^{137}Cs in the cup containing the soil in equilibrium with tracer was 1.2E+08 Bq/m^3 while the concentration of ^{60}Co was 4.9E+06Bq/m^3. The two cells were put in contact for a period of time, after which each cell was taken apart and the soil plugs were sliced. Concentration of the two radionuclides was determined in each slice and a normalized concentration profile as a function of distance was obtained [5, 6].

Diffusion coefficients are obtained by solving the one-dimensional diffusive equation (6) for the measured radionuclide concentration profile over time.

The classical analytical solution to partial differential equation (6) with simple boundary condition is well discussed in the literature [7]. For a porous medium of length L, the applicable initial and boundary conditions are:

$$\text{I.C:} \quad c(x,0) = c_0 \text{ for } 0 < x < x_0$$

$$c(x,0) = 0 \text{ for } x_0 < x < L \qquad (7)$$

$$\text{B.C.} \quad \frac{\partial c}{\partial x} = 0 \text{, at } x = 0 \text{ and } x = L$$

With the initial and boundary condition given in the equation (7), an analytical solution to the equation (6) is [7]:

$$\frac{C}{C_0} = \frac{x_0}{L} + \frac{2}{\pi}\sum_{i=1}^{\infty}\left[\exp\left(-\frac{\pi^2 \cdot i^2 \cdot D_a \cdot t}{L}\right)/i\right]\cos\frac{i\pi x}{L}\sin\frac{i\pi x_0}{L} \qquad (8)$$

where: C – radionuclide concentration at distance x, Bq/kg

C_0 – initial radionuclide concentration in contaminated soil sample

x_0 – interface between contaminated and non-contaminated soil samples, m

L – diffusion cell length, m

This equation was used for experimental determination of cesium and cobalt diffusion coefficients in the main horizons of Saligny site.

The radionuclides concentration in each slice was measured using a gamma multi-channel spectrometer with Ge(Li) detector. With the normalized concentration profile, a computer program is used to calculate, according to equation (8), the apparent diffusion coefficient for each radionuclide. The computer program uses the least squares method in order to determine the value for the apparent diffusion coefficient that minimizes the residual sum of squares.

A schematic representation of the diffusion cell used in the experiments is presented in Fig. 2 [8].

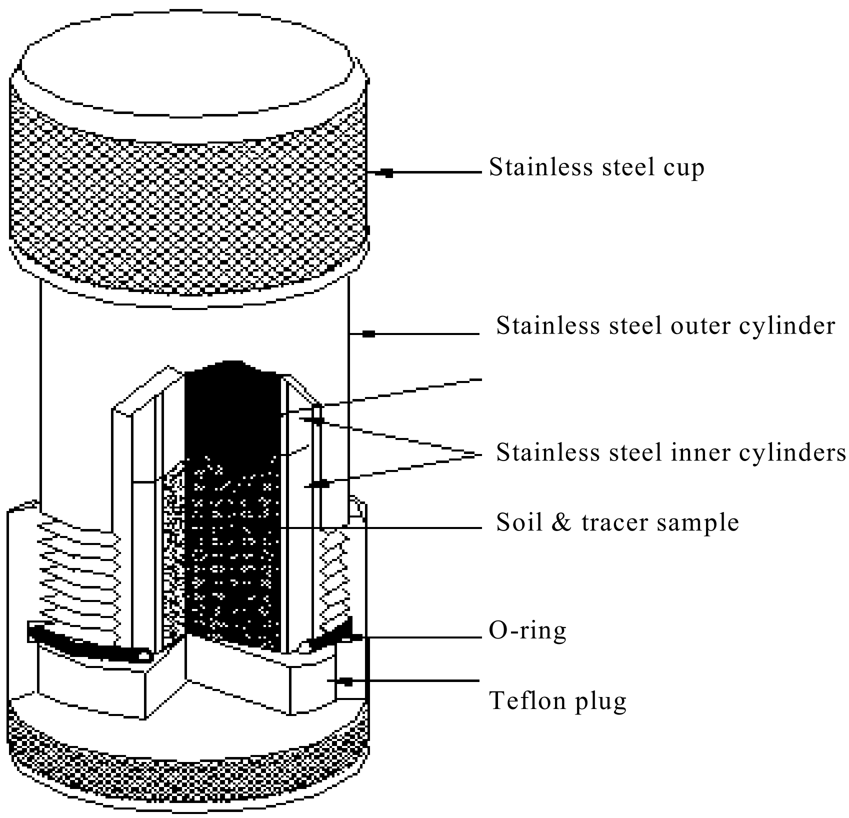

Fig. 2. Diffusion closed-cell.

RESULTS AND DISCUSSION

The experimental values for apparent diffusion coefficients for the two radionuclides are given in the Table 2.

Table 2

Diffusion coefficients for Cs-137 and Co-60 in the geological media studied

Geological Unit	**D_a [m^2/s]**	
	Cs-137	**Co-60**
Loess	1.82E-09	2.84E-09
Consolidated loess	9.72E-10	1.55E-09
Clayey loess	1.51E-09	2.34E-09
Red clay	6.36E-10	1.12E-09
Quaternary clay	7.43E-10	1.16E-09

Fig. 3 provides the variation of the diffusion coefficients with the type of the geological media. The experimental data show that radionuclides diffuse slower in the samples with low permeability and low porosity. Diffusion coefficients for both radionuclides have higher values in the samples more permeable, characterized by a larger porosity, than in less permeable samples.

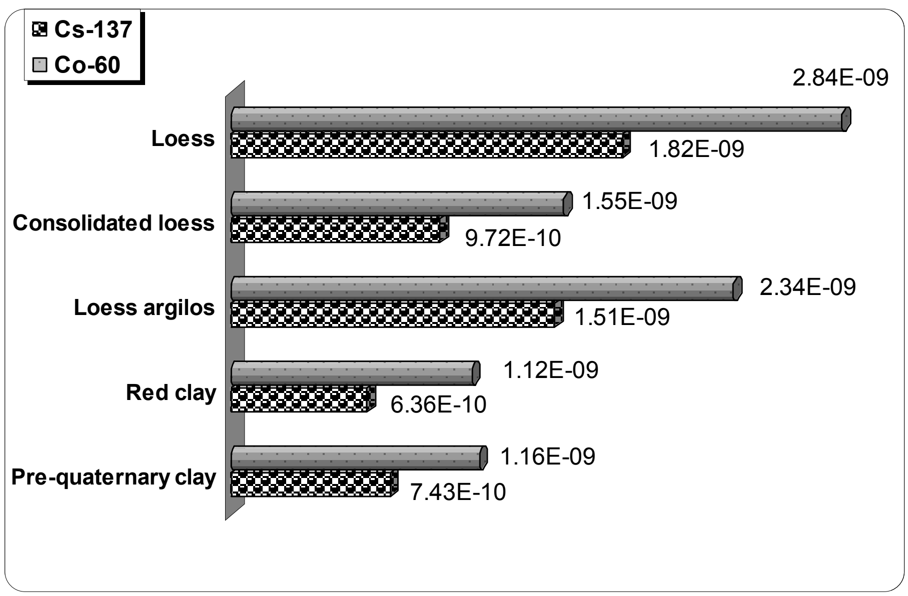

Fig. 3. Variation of the diffusion coefficients with the type of geological media.

Figs. 4 and 5 present the dependency of the diffusion coefficients on permeability and porosity respectively.

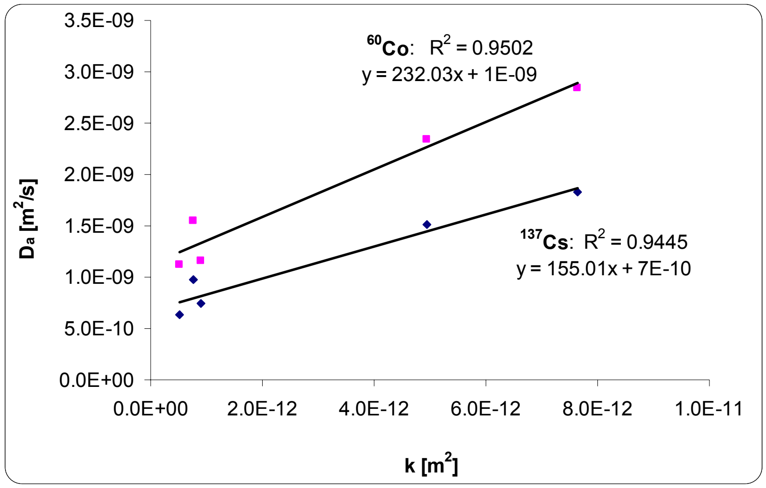

Fig. 4. Correlation between diffusion coefficients and permeability.

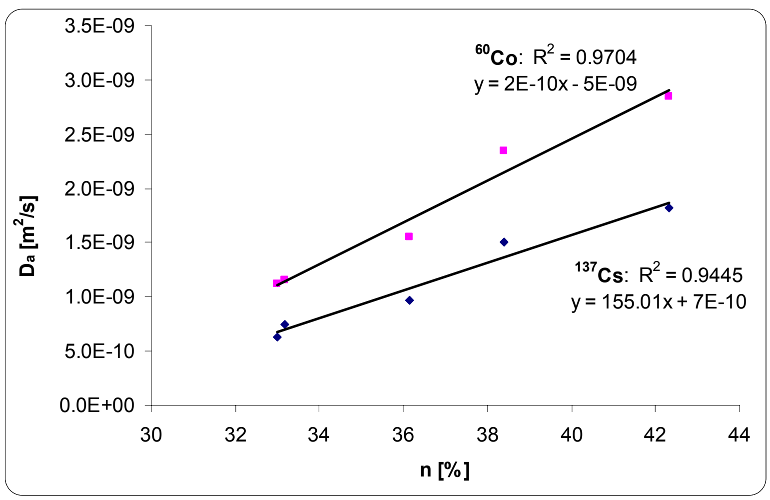

Fig. 5. Correlation between diffusion coefficients and porosity.

Good linear correlations between diffusion coefficients and permeability of the rock (correlation coefficient 0.94) and between diffusion coefficients and porosity (correlation coefficient 0.96) were observed.

A multivariable linear regression was used to determine the influence of the permeability and porosity of the rock on the diffusion coefficient. By multiple variable regression, an equation between independent variables and one dependent variable can be found:

$$y = m_1 \cdot x_1 + m_2 \cdot x_2 + \ldots + m_n x_n + b \quad (9)$$

where: y – is dependent variable
x_i – are independent variables
m_i – are regression coefficients
b – a constant

The multivariable regression was conducted on the diffusion coefficient results by using the measured values for the permeability and porosity as independent variables and diffusion coefficient estimates as dependent variables. Table 3 presents the results of the multivariable linear regression and its associated statistics. Equations found by regression between dependent and independent variables are:

For Cs-137:

$$D_a = (65.70 \pm 19.12) \cdot k + (7.76E\text{-}11 \pm 1.56E\text{-}11) \cdot n - 1.9E\text{-}09 \pm 5.2E\text{-}10 \quad (10)$$

For Co-60:

$$D_a = (92.98 \pm 26.53) \cdot k + (1.21E\text{-}10 \pm 2.17E\text{-}11) \cdot n - 2.90E\text{-}09 \pm 7.23E\text{-}10 \quad (11)$$

Table 3

Regression coefficients and statistics associated with regression

Nuclide	**$m_1 \pm se_1$**	**$m_2 \pm se_2$**	**$b \pm se_b$**	**se_y**	**R^2**	**F**	**df**
^{137}Cs	65.70± 19.12	7.76E-11± 1.56E-11	-1.9E-09± 5.2E-10	7.07E-11	0.981	307.49	12
^{60}Co	92.98± 26.53	1.21E-10± 2.17E-11	-2.9E-09± 7.23E-10	9.82E-11	0.984	359.26	12

In Table 3, se_1, se_2 are the standard error values for the coefficients m_1, m_2; se_b is the standard error value for the constant b; se_y is the standard error for the y estimate; R^2 is the coefficient of determination, df represents the degrees of freedom; F is F statistic, or the F-observed value. The F statistic is used to determine whether the observed relationship between the dependent and independent variables occurs by chance. The F-values must exceed some critical value for the regression to be useful in predicting the dependent variable. For df=12, the critical value for 95% confidence level is 3.89, much lower than F-observed values (307.49, for ^{137}Cs and 359.26 for ^{60}Co), that is the results of regression and correlation are indeed true and not the consequence of chance.

CONCLUSIONS

Very low diffusion coefficients were obtained for both radionuclides in all geological media studied.

This suggests that in the porous media characteristic to Saligny site, the dominant transport mechanism is advection.

From the multivariate regression, applied on diffusion coefficient values as dependent variable and permeability and porosity of the rock as independent variables, a significant calculated F-value was obtained; this indicates that the results of regression and correlation are indeed true and not the consequence of chance.

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