FACTA UNIVERSITATIS
Series: **Working and Living Environmental Protection** Vol. 8, N° 1, 2011, pp. 1 - 5

A LEACHING STUDY IN THE PROCESS OF SOLIDIFICATION OF RADIONUCLIDE ^{137}Cs IN CONCRETE [*]

UDC 691.32:621.642.8:621.039.58:616-089.22

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Abstract. *To assess the safety of the disposal of radioactive waste – of cement composition, the leaching of ^{137}Cs from a waste composite into surrounding fluids has been studied. Leaching tests were carried out in accordance with the method recommended by the IAEA. The determination of retardation factors, KF and the coefficients of distribution, kd, using a simplified mathematical model for analyzing the migration of radionuclides, has been developed. The results presented in this paper are examples of results obtained in a 20 year mortar and concrete testing project, which will influence the design of the engineered trenches system for a future central radioactive waste disposal center.*

Key words: *concrete, leaching, immobilization, radioactive waste*

1. INTRODUCTION

In order to prevent widespread dispersion of radionuclides into the human environment, radioactive waste produced in nuclear facilities has been incorporated in several kinds of matrices [1,2,3,4,5,6,7]. The objective of the immobilization of radioactive waste is to convert waste into forms which are:

- Leach resistant so that the release of radionuclides will be slow even though they may come into contact with flowing water;
- Mechanically, physically and chemically stable for handling transport and disposal.

Concrete is widely used in low-level waste management, both as a means of solidifying waste and for the containment of dry or liquid waste. At present there is also widespread interest in the use of a near-surface concrete trench system for the disposal of ra-

Received November 03, 2011
[*] **Acknowledgements**. Work supported by the Ministry of Science and Technological Development of the Republic of Serbia

dioactive waste material. Typical concrete is a mixture of cement, sand, granulate and water in various proportions, which together determine the structural properties and tightness of the poured material. Cement is a porous, continuously hydrating material whose actual surface area greatly exceeds its geometric surface area. In leaching, the rate of dissolution varies as a function of the chemistry phase and this dissolution exposes or enlarges pores; thus the leaching behavior must be related to pore structure and the composition of the pore solution [6,7,8,9,10]. Although cement has several unfavorable characteristics as a solidifying material, i.e. low volume reduction and relatively high leachability, it possesses many practical advantages: good mechanical characteristics, low cost, easy operation and radiation and thermal stability.

2. Radionuclide Migration Through Porous Materials

The dispersion of radionuclides in porous materials, such as grout or concrete, is described using a one-dimensional differential model [1,5].

$$D\frac{\partial^2 A}{\partial X^2} - V_V\frac{\partial A}{\partial X} - \left(1 + \frac{1-f}{f}\rho_T k_D\right)\frac{\partial A}{\partial t} = 0 \tag{1}$$

or

$$D\frac{\partial^2 A}{\partial X^2} - V_V\frac{\partial A}{\partial X} - K_F\frac{\partial A}{\partial t} = 0 \tag{1'}$$

where:

K_F – retardation factor (=)1
D – diffusion coefficient (cm^2/d) or (cm^2/s)
A – concentration in the liquid (mol/l) or (Bq)
X – length (cm)
V_v – velocity of the leachant fluid (cm/d)
f – porosity (=)1
ρ_T – bulk density (g/cm^3)
k_d – distribution coefficient (ml/g)
t – time variable (d).

Using the Laplace transformation method, Eq.(1') becomes:

$$\frac{A_n}{A_0} = \frac{1}{2}\operatorname{erf} z \left| \sqrt{\frac{V_V X}{4D_e}} \cdot \frac{1-\frac{V_V t}{K_F X}}{\sqrt{\frac{V_V t}{K_F X}}} \right| \tag{2}$$

from which we can calculate the retardation factor, K_F. The coefficient of distribution, k_d, can be calculated:

$$k_d = \frac{(K_F - 1)f}{(1-f)\rho_T} (=) \ \text{(ml/g)} \tag{3}$$

in which: V_v, X, ρ_T, t and A_o are known. A_n and D_e can be determined experimentally using a leaching test procedure [3].

3. DETERMINING THE EFFECTIVE COEFFICIENTS OF DIFFUSION

For the interpretation of the results of the leach tests shown in the following figures and tables, the leach coefficient D, is used, and it is defined as:

$$D = \frac{\pi}{4} m^2 \frac{V^2}{S^2} \quad (cm^2/d) \tag{4}$$

where:

D – leach coefficient (diffusion) (cm^2/d) or (cm^2/s);
m – $(\Sigma A_n/A_o)\cdot(1/\sqrt{\Sigma t})$, slope of the straight line ($d^{-1/2}$);
A_o – initial sample activity at time zero ^{137}Cs (Bq);
A_n – activity leached out of sample after leaching time t, (Bq);
t – duration of leaching renewal period (d); (1,2,3,4,5,6,7,15,30,60,90)
V – sample volume (cm^3);
S – sample surface (cm^2).

4. EXPERIMENTAL PROCEDURE

The concrete samples were made of:

- Portland cement PC-20-Z-45 MPa
- Sand, fraction 0-2 mm
- granulate, fraction 2-4,4-8, and 8-15mm
- 60 (kBq)^{137}Cs in Water,
- Additive, Superfluidal);

More than 150 different formulations of concrete were examined to optimize their mechanical and sorption properties. In this paper we discuss four representative formulations. The composition of the concrete samples are shown in Table 1.

Table 1. The representative formulation of concrete composition in the form of grams, for 1000 cm^3 of concrete. Each sample takes the initial sample activity of 60 (kBq)^{137}Cs in Water

	Portland cement	Sand 0-2 mm	Aggregate 2-4 mm	Aggregate 4-8 mm	Aggregate 8-15 mm	Water	Aditive
Sample 1	400	652	95	443	714	150	8
Sample 2	410	672	85	443	774	160	8
Sample 3	420	802	71	575	496	175	8
Sample 4	430	642	83	307	923	190	8

5. RESULTS AND DISCUSSION

The results were obtained after 90 days. Using equation (4), the coefficients of diffusion are calculated for four experimental samples.

Using equation (2) and (3), the retardation factors, K_F(=)1, and distribution coefficients, k_d(ml/g) were calculated. Table 2 gives ^{137}Cs , leach coefficients in different concrete samples.

Table 2. Leach coefficients $D_e(cm^2/d)$ in different grout samples after 90 days, using Eq.(4)

	Formula			
Leach coeff.	Sample 1	Sample 2	Sample 3	Sample 4
D_e, ^{137}Cs	$5,32\cdot10^{-5}$	$4,62\cdot10^{-6}$	$3,20\cdot10^{-6}$	$6,20\cdot10^{-6}$

Table 3 gives the results of the retardation factors, K_F and the coefficients of distribution, k_d(ml/g), for four mortar formulations for each radionuclide, during 90 days.

Table 3. Retardation factor K_F and the coefficients of distribution k_d(ml/g), after 90 days, ρ_T=2,5 (g/cm^3). f=0,15-0,30

	Formula			
Coeffic.	Sample 1	Sample 2	Sample 3	Sample 4
K_F, ^{137}Cs	51	93	95	75
k_d, ^{137}Cs	1-3	6-16	6-17	6-14

6. CONCLUSIONS

The analysis of the results presented in Table II and Table III shows that the values of the retardation factors and coefficients of radionuclides ^{137}Cs, are similar to those found in the literature, and prove that the one-dimensional model can be used for calculating parameters of the migration process. The system of concrete engineered trenches as the final disposal system for radioactive waste permits secure preservation of radionuclides for more than 300 years in a future disposal system, with multiple safety barriers.

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ANALIZA PROCESA CURENJA U PROCESU SOLIDIFIKACIJE RADIONUKLEIDA ^{137}Cs U BETONU

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Za procenu bezbednosti odlaganja radioaktivnog otpada - cementnog sastava, analizirano je curenje137Cs od kompozita otpada u okolne tečnosti. Testovi su obavljeni u skladu sa metodom preporučene od strane IAEA. Određivanjem faktora retardacije, FK i koeficijenta raspodele, kd, razvijen je pojednostavljeni matematički model za analizu migracija radionuklida. Rezultati prezentovani u ovom radu su primeri rezultata dobijenih tokom 20 godina testiranja maltera i betona, projekta koji će uticati na dizajn sistema projektovanja rovova za buduće centrale radioaktivnog centra za odlaganje otpada.

Ključne reči: *beton, curenja, imobilizacija, radioaktivni otpad*