^{60}Co LEACHING THROUGH MORTAR-RADWASTE MATRIX

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Abstract – *To assess the safety for disposal of radioactive mortar-waste composition, the leaching of ^{60}Co from a waste composite into a surrounding fluid has been studied. Leaching tests were carried out in accordance with a method recommended by IAEA. Determination of retardation factors, K_F and coefficients of distribution, k_d, using a simplified mathematical model for analyzing the migration of radionuclides, has been developed. In our experiment we have achieved the lowest leaching values after 60 days in samples. 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 Serbian radioactive waste storage center.*

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 . The objectives of immobilization of radioactive waste is to convert the 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; and mechanically, physically and chemically stable for handling transport and disposal.

Cement and concrete are widely used in low-level waste management both as a means of solidifying waste and for containment of dry or liquid wastes. At present there is also widespread interest in the use of near-surface concrete trench system for the disposal of radwaste materials. Typical mortar is a mixture of cement, sand and water in various proportions, that together determine the structural properties and tightness of the poured material. Water content is one of the critical parameters and must be carefully controlled during purring and setting; to a large extent it will determine the porosity of the resulting material. Cement is 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 phase chemistry and this dissolution exposes or enlarges pores; thus the leaching behavior must be related to pore structure and the composition of the pore solution[1,2,3].

2. RADIONUCLIDE MIGRATION THROUGH POROUS MATERIALS

The dispersion of radionuclides in porous materials, such as grout or concrete, is described using a one dimensional dimodel [2,4,5,6,7]

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

or

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

where:

K_F - retardation factor (=)1

D - diffusion coefficient (cm^2/d) or (cm^2/s)

A - concentration in liquid (mol/l) or (Bq)

X - mean path length of radionuclide (cm)

V_v - velocity of 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 Laplace transformation method, Eq.(1') becomes:

$$\frac{A_n}{A_0} = \frac{1}{2}\mathrm{erf}\, z\left|\sqrt{\frac{V_V X}{4D_e}}\cdot\frac{1-\frac{V_V t}{K_F X}}{\sqrt{\frac{V_V t}{K_F X}}}\right| \qquad (2)$$

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

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

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 leach tests shown in the following tables, leach coefficient D, is used, and it is defined as:

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

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 (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)

V - sample volume (cm^3);

S - sample surface (cm^2).

4. MATERIALS AND CONCRETE COMPOSITION

Concrete samples were made of:

- Portland cement PC-20-Z-45 MPa ;
- Sand, fraction 0-2 mm
- Water ;
- Additive, Superfluidal;

More then 100 different formulations of mortar formulations were examined to optimize their mechanical and sorption properties. In this paper we discuss four representative formulations. Mortar composition are shown in Table I.

Table I. *Mortar compositions (calculated as gram for 1000 cm^3 of concrete)*

Materials	Formula			
Materials	M_1	M_2	M_3	M_4
Cement (Portland), gr	750	700	650	600
Sand, 0-2 mm	1250	1310	1325	1375
Water, ml	250	235	230	225
Additives, ml	12	13	14	15
Initial activity A_o(kBq)^{60}Co		55,0		

5. RESULTS

The results are obtained after 60 days. Using equation (4), coefficients of diffusion are calculated for four experimental samples.

Using equation (2) and (3) , retardation factors, K_F(=)1, and distribution coefficients, k_d(ml/g) are calculated. Table II gives ^{60}Co, leach coefficients in different mortar samples. Table II gives the results of retardation factors, K_F and coefficients of distribution, k_d(ml/g), for four concrete formulations , during 60 days.

Table II *Leach coefficients D_e(cm^2/s) in different mortar samples after 60 days, using Eq.(4)*

	Formula			
	M_1	M_2	M_3	M_4
Leach coeff. D_e, ^{60}Co	$1{,}03\ 10^{-14}$	$3{,}2\ 10^{-14}$	$5{,}1\ 10^{-13}$	$7{,}1\ 10^{-13}$

Table III *Retardation factor K_F and coeff. of distribution k_d(ml/g), after 60 days, ρ_T=2,5 (g/cm^3). f=0,15-0,30*

	Formula			
	M_1	M_2	M_3	M_4
K_F, ^{60}Co	56,2	34,6	33,7	27,7
k_d, ^{60}Co	2-4	3-5	3-5	6-12

6. CONCLUSION

The analysis of the results presented in Table II and Table III show that the values of retardation factors and coefficients of ^{60}Co radionuclide, are similar to the literature data, and prove that the one-dimensional model can be used for calculating parameters of the migration process. The main conclusion from this experiment is that biggest quantity of water gives higher Retardation factors, K_F, and determine the porosity in samples, and the diffusion coefficient are the critical and major contribution parameters compared to any other mechanisms in analyzing the mortar samples.

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Sadržaj – U radu je prikazano odredjivanje retardacionog koeficijenta, koeficijenta distribucije korišćenjem jednostavnog matematičkog modela za analizu migracije radionuklida.U eksperimentima je odredjivan najniži rezultat izluživanja u uzorcima nakon 60 dana. Rezultati će biti korišćeni pri projektovanju budućeg konačnog odlagališta za Rao niske i srednje radioaktivnosti.

IZLUŽIVANJE ^{60}Co IZ MALTERNOG MATRIKSA

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