Letter to Editor

Leaching Study on the Process of Solidification of Radionuclide ^{60}Co in Concrete

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Abstract

To assess the safety for disposal of radioactive waste-cement 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 the 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. Results presented in this paper are examples of results obtained in a 30-year mortar and concrete testing project, which will influence the design of the engineered trench system for a future central radioactive waste disposal center.

Keywords: concrete, leaching, immobilization, radioactive waste, retardation factors, coefficients of distribution

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 [1-6]. 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;
– 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 containment of dry or liquid wastes. At present there is also widespread interest in the use of near-surface concrete trench systems for the disposal of radwaste materials. Typical concrete is a mixture of cement, sand, granulate and water in various proportions, that together determine the structural properties and tightness of the poured 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. 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.

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].

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

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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, K_f is velocity of radionuclide per velocity of the conveying fluid
D – diffusion coefficient (cm^2/d) or (cm^2/s)
A – concentration in liquid (mol/l) or (Bq)
X – length (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), = distribution factor of a radionuclide between soil and water (ml/g)
t – time (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].

Determining the Effective Coefficients of Diffusion

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

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

where:
D – Difussion coefficient (cm^2/d) or (cm^2/s);
m – ($\Sigma A_n/A_o$) x ($1/\sqrt{\Sigma t}$), slope of the straight line ($d^{-1/2}$);
A_o – initial sample activity at time zero (Bq); (Table I)
A_n – activity leached out of sample after leaching time t, (Bq);
t – time (d);
V – sample volume (cm^3);
S – sample surface (cm^2).

Experimental

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
- Water,
- Additive, Superfluidal);

More than 150 different formulations of concrete were examined to optimize their mechanical and sorptive properties. In this paper we discuss four representative formulations. Concrete compositions are shown in Table 1.

Results and Discusion

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

Using equations (2) and (3), retardation factors, K_F (=) 1, and distribution coefficients, k_d (ml/g) are calculated. Table 2 gives ^{60}Co leach coefficients in different concrete samples.

Table 1. Representative formulation of concrete composition as grams, for 1000 cm^3 of concrete.

	Portland cement	Sand 0-2 mm	Aggregate 2-4 mm	Aggregate 4-8 mm	Aggregate 8-15 mm	Water	Aditive
Sample 1	400	672	85	463	724	150	8
Sample 2	400	692	75	423	794	150	8
Sample 3	400	822	91	595	476	150	8
Sample 4	400	662	73	317	933	150	8

Initial sample activity of ^{60}Co, $A_0 = 8 \cdot 10^7$ Bq/sample

Table 2. Leach coefficients D_e (cm^2/d) in different grout samples after 60 days, using Eq.(4).

Leach coeff.	Formula			
	Sample 1	Sample 2	Sample 3	Sample 4
D_e, ^{60}Co	$5.22 \cdot 10^{-5}$	$4.72 \cdot 10^{-6}$	$3.24 \cdot 10^{-6}$	$6.14 \cdot 10^{-6}$

Table 3. Retardation factor K_F and coefficients of distribution k_d (ml/g), after 60 days, ρ_T=2.5 (g/cm^3), f=0.15-0.30.

Coeffic.	Formula			
	Sample 1	Sample 2	Sample 3	Sample 4
K_F, ^{60}Co	51.2	93.5	95.4	75.5
k_d, ^{60}Co	1-3.5	6-16.5	6-16.5	6-14

Table 3 gives the results of retardation factors, K_F and coefficients of distribution k_d (ml/g), for four mortar formulations for each radionuclide, during 60 days.

Conclusions

The analysis of the results presented in Tables 2 and 3 shows that the values of retardation factors and coefficients of radionuclides ^{60}Co are similar to the literature data, and prove that the one-dimensional model can be used for calculating parameters of the migration process. The system of concrete trenches engineered as a 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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