IMMOBILIZATION OF ^{137}Cs IN CONCRETE

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Abstract

To assess the safety for disposal of radioactive waste-cement-bentonite composition, the leaching of ^{137}Cs 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. Results presented in this paper are examples of results obtained in a 10 year mortar and concrete testing project, which will influence the design of the engineered trenches system for a future central radioactive waste disposal center.

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.[1-7] 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 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 radio-waste 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]

$$D\frac{\partial^2 A}{\partial X^2} - V_V\frac{\partial A}{\partial X} - \left(1 + \frac{1-f}{f}\rho_T k_D\right)\frac{\partial A}{\partial t} = 0 \quad (1) \quad \text{or} \quad D\frac{\partial^2 A}{\partial X^2} - V_V\frac{\partial A}{\partial X} - K_F\frac{\partial A}{\partial t} = 0 \quad (1')$$

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 - 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)
- t - time variable (d)

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

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

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

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

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 it is defined as:

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

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 (Bq); (Table 1)
- 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)

Experimental

Materials and mortar composition

Concrete samples were made of: Portland cement PC-20-Z-45 Mpa, Sand, fraction 0-2 mm, granulate, fraction 2-4, 4-8, and 8-15 mm, Water, Additive Superfluidal. More than 150 different formulations of concrete formulations were examined to optimize their mechanical and sorption properties. In this paper we discuss four representative formulations. Concrete compositions are shown in Table 1.

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

	Portland cement	Sand 0-2 mm	Aggregate 2-4 mm	Aggregate 4-8 mm	Aggregate 8-15 mm	Water	Admixture
Sample 1	440	672	85	463	724	150	8
Sample 2	420	750	83	417	734	140	8
Sample 3	400	858	95	536	496	140	8
Sample 4	400	626	69	576	715	140	8

Results and discussion

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 2 gives ^{137}Cs, leach coefficients in different concrete samples.

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.

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, ^{137}Cs	4.62×10^{-5}	5.62×10^{-6}	4.73×10^{-6}	8.44×10^{-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.

Coeff.	Formula			
	M_1	M_2	M_3	M_4
K_F, ^{137}Cs	41.6	87.5	98.4	65.5
k_d, ^{137}Cs	1.6-3.8	6.8-16.7	6.9-16.9	6.7-16.3

Conclusions

The analysis of the results presented in Table 2 and Table 3 shows that the values of retardation factors and coefficients of radionuclides ^{137}Cs, 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 engineered trenches 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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References

1. R. H. Burns, *Atom. Energy Rev.* **1971**, *9*, 547–552.
2. I. Hashimoto, K. B. Deshpande, S. H. Thomas, *I&EC Fundamentals* **1964**, *3*, 213–217.
3. E. D. Hespe, *Atom. Energy Rev.* **1971**, *9*, 195–207.
4. A. H. Lu, *Health Physics* **1978**, *34*, 39–43.
5. N. Moriyama, S. Dojiri, H. Matsuzuru, *Health Physics* **1977**, *32*, 549–552.
6. I. Plecas, Lj. Mihajlovic, A. Kostadinovic, *Radioactive Waste Management and Nuclear Fuel Cycle* **1985**, *6*, 161–175.
7. I. Plecas, J. Drljaca, A. Peric, A. Kostadinovic, S. Glodic, *Radioactive Waste Management and the Nuclear Fuel Cycle* **1990**, *14*, 195–205.

Povzetek

Za oceno varnosti odlaganja utrjenih radioaktivnih odpadkov v betonskih zmeseh cementa in bentonita smo študirali izluževanje ^{137}Cs iz teh kompozitov v tekočinskem okolju. Izluževalni preizkusi so bili usklajeni s priporočeno metodo IAEA. Na osnovi poenostavljenega modela migracije radionuklidov v trdni snovi smo določili zadrževalne faktorje K_F in koeficiente porazdelitve k_d. Raziskave parametrov imobilizacije ^{137}Cs v betonu so del 10 letnega projekta preizkušanja betonskih in maltnih materijalov za imobilzacijo odpadnih radioaktivnih snovi in so med drugim podlaga za načrtovanje varnostnega sistema inženirskih pregrad v bodočem centralnem odlagališču radioaktivnih odpadkov.