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COMPARATIVE LEACHING STUDIES OF ^{60}Co FROM SPENT RADIOACTIVE ION - EXCHANGE RESIN INCORPORATED IN CEMENT

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ABSTRACT

Leaching of ^{60}Co from ion - exchange resin incorporated in cement using two methods based on theoretical equation has been developed. These were: Method I, diffusion equation derived for a plane source model and Method II, empirical model employing a polynomial equation (1, 2, 3). Results presented in this paper are examples of data obtained in a cement testing project which will influence the design of the future radioactive waste storage centre.

Introduction

The intermediate level radioactive wastes produced as a result of the operation of reprocessing plants and nuclear power station are currently being considered for immobilization in a number of possible matrices, including cement, bitumen and plastics. In the storage or disposal of these solidified wastes, some radioactive products may be leached into contacting water and this water might enter into the environment. Therefore, an estimation of the quantities of radionuclides that can be leached from waste composites is essential in radioactive waste management.

At our laboratory, a promising composite for solidification of radioactive wastes has been developed by incorporation of spent ion exchange resins in cement. Leaching of ^{60}Co was studied using the method recommended by the IAEA (1). Transport phenomena involved in the leaching of a radioactive material from a composite matrix into surrounding water were investigated using two methods based on theoretical equations. These were: Methods I, diffusion equation derived for a plane source model, and Method II, empirical method employing a polynomial equation (2, 3). These two methods are compared with respect to their applicability to the ^{60}Co leaching data.

Experimental

Leaching test

A cation mix bed spent ion - exchange resin was contaminated with ^{60}Co by a batch method to give a radioactivity of $7.4 \cdot 10^7$ Bq per ortho cylinder shape sample, (diameter 5 cm) (3, 4, 5, 6).
Two commercial cements were used: Portland, PC - 55 MPa normal type by Yugoslavia industrial standard, and Portland blast - furnace slag cement, PC - 20Z 45 MPa. The chemical composition of these cements is summarized in Table 1.

TABLE 1.
Chemical Composition of Cement (wt%)

Chemical component	PC - 55 MPa	PC - 20Z 45 MPa
SiO_2	21.6	28.5
Al_2O_3	5.5	11.7
Fe_2O_3	3.2	2.3
CaO	66.8	52.9
MgO	1.5	3.3
Insoluble residue	0.6	0.7
Ignition loss	0.8	0.6

Samples of cement composites were prepared by mixing each cement (100 g) with water (20 g) and wet mix bed ion-exchange resin (27 g). After sufficient mixing, both pastes were poured into a cylindrical vessel. The curing time of the specimens was 28 days.

Leaching tests were carried out using the method recommended by Hespe (1).

Each specimen was immersed in a leaching vessel containing 250 ml of tap water. At present time intervals, leachant was removed for radioactivity measurements by "EG&G ORTEC" spectrometry system and software, with the low-level background.

Mathematical Treatment of Data

Method I, diffusion equation based on a plane source model

In this model, the fraction f leached at time is given by

$$f = \frac{a}{A} = \frac{2S\sqrt{Dt}}{V\sqrt{\pi}} \tag{1}$$

where a[Bq] is the radioactivity leached during the leaching renewal period, A[Bq] is the radioactivity initially in the specimen, D[cm^2/day] is the diffusion coefficient, and S[cm^2] and V[cm^3] are the surface area and volume of a specimen, respectively. The value of D can be calculated from the slope m of the linear relation of f and $t^{1/2}$, i. e.

$$m = \frac{2S\sqrt{D}}{V\sqrt{\pi}} \tag{2}$$

or

$$D = \frac{\pi m^2 V^2}{4S^2} \tag{3}$$

The amount of a radionuclide leached from the waste composites over a given period can be predicted from equation (1).

Method II polynomial equation

The orthogonal polynomial is one of the most useful empirical equations. Its general form is:

$$y(x) = \sum_{i=1}^{n} A_i \Phi_i(x_i) \tag{4}$$

where A_i is the parameter to be determined , and Φ_i is the function of x_i.
Here, $\Phi_i(x_i)$ is taken as $t_i^{1/2}$, and the leaching fraction is given by :

$$f = \sum_{i=1}^{n} A_i t_i^{\frac{1}{2}} \tag{5}$$

To simplify the mathematical treatment, a third degree polynomial of the form

$$f = A_1 t^{\frac{1}{2}} + A_2 t + A_3 t^{\frac{3}{2}} \tag{6}$$

was fitted to the leaching data.

Results

Results are presented in FIG 1. and FIG 2, which show the fraction of ^{60}Co leached from cement composites as a function of the square root of leaching period, $t^{1/2}(d^{1/2})$, and their linear and polynomial approximation.

In the data for both cement composite, linearity between f and $t^{1/2}$ is not observed throughout the time tested; however, there are two different linearities before and after a leaching time of about 20 days.

The leaching process can be described by:

$$f = m t^{\frac{1}{2}} + b \tag{7}$$

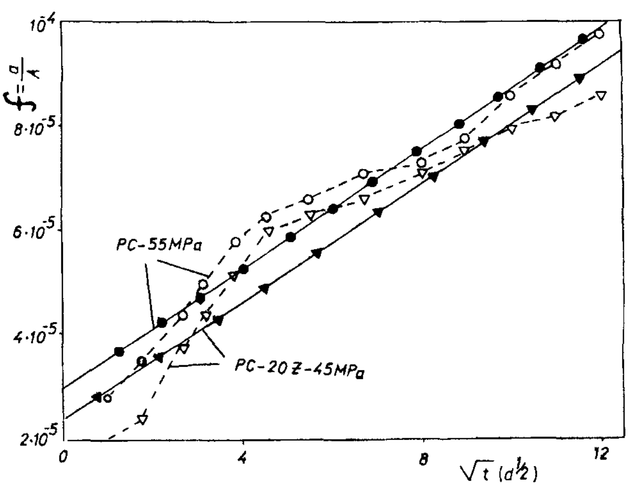

FIG 1
Plot of f vs $t^{1/2}$ for leaching of ^{60}Co from spent ion - exchange resin incorporated in two type of cement, using linear aproximation

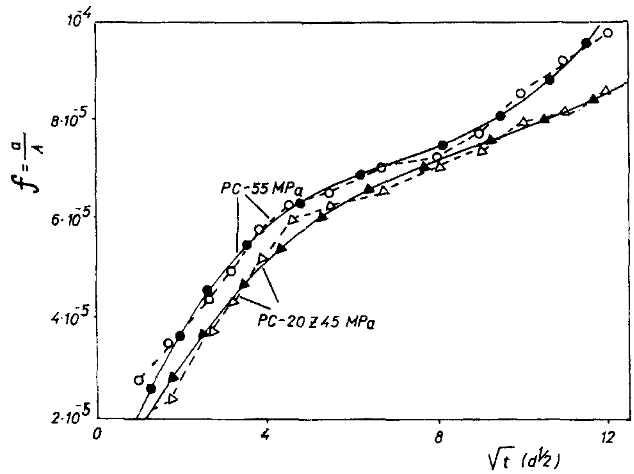

FIG 2
Plot of f vs $t^{1/2}$ for leaching of ^{60}Co from spent ion - exchange resin incorporated in two type of cement, using polynomial equation (--)

where m is the coefficient defined by equation (2), and b is the intercept of f-axis at t = 0, which corresponds to the contribution of initial leaching process. From the application of Method I to the leaching data we obtained for PC-55 MPa:

$$f_{55} = 5.76 \cdot 10^{-6} \sqrt{t} + 2.97 \cdot 10^{-5} \tag{8}$$

and for PC-20Z 45 MPa:

$$f_{45} = 5.68 \cdot 10^{-6} \sqrt{t} + 2.4 \cdot 10^{-5} \tag{9}$$

The diffusion coefficients predicted by Method I are $D_{55} = 1.79 \cdot 10^{-11} [\frac{cm^2}{d}]$ and $D_{45} = 1.74 \cdot 10^{-11} [\frac{cm^2}{d}]$.

Using the method of least squares, Method II yields:

$$f_{55} = 2.428 \cdot 10^{-5}\sqrt{t} - 2.941 \cdot 10^{-6}\, t + 1.345 \cdot 10^{-7}\sqrt{t^3} \quad (e^2 = 7.154 \cdot 10^{-11}) \tag{10}$$

and

$$f_{45} = 1.910 \cdot 10^{-5}\sqrt{t} - 1.780 \cdot 10^{-6}\, t + 6.556 \cdot 10^{-8}\sqrt{t^3} \quad (e^2 = 4.752 \cdot 10^{-11}) \tag{11}$$

Conclusion

Though method I, can't describe the whole leaching process, it is very convenient to simulate the leaching in the longer period. For both cement, Method II gives very good approximation during the whole period tested. Analyzing the results presented in equations (8) - (11) and FIG 1. and FIG. 2, it is noticed that calculated values are quite good, and similar to the literature data (2,3).

Results presented in this paper will influence the design of the our future engineered trenches disposal system for radioactive waste.

Acknowledgements

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