

EFFECT OF CURING TIME ON THE FRACTION OF ^{137}Cs FROM CEMENT MATRIX

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Abstract

To assess the safety of disposal of radioactive waste material in concrete, curing conditions and time of leaching radionuclides ^{137}Cs have been studied. Leaching tests in concrete were carried out in accordance with a method recommended by IAEA.(1) Curing conditions and curing time prior to commencing the leaching test are critically important in leach studies since the extent of hydration of the cement materials determines how much hydration product develops and whether it is available to block the pore network, thereby reducing leaching. Results presented in this paper are examples of results obtained in a 20-year concrete testing project which will influence the design of the engineer trenches system for future central Serbian radioactive waste storing center.

Introduction

Cement has been successfully used in radioactive waste management to immobilize a wide range of waste including filter sludges, ion-exchange resins and evaporator concentrates produced in nuclear power plants. In any assessment of cement-immobilized radwaste composites, leach of radionuclides (^{60}Co , ^{137}Cs , ^{54}Mn ...) is an important factor, for it must be assumed that the composite material will eventually come into contact with leachant solutions.(2,3,4) 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 concrete is a mixture of cement, sand, stone aggregate 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 curing and setting; to a large extent it will determine the porosity of the resulting material.

Materials, methods and experimental conditions

Concrete samples were made of: Portland cement, PC-45 MPa, Sand, (fraction: 0-2 mm) Agregate, (fraction: 2-4 mm; 4-8 mm and 8-15 mm), Water attested according to Yugoslav standard, Initial activity per sample $A_0 = 7 \cdot 10^6$ of ^{137}Cs .

In this paper we discuss three representative formulation of concrete composition shown in Table I.

Table I. Representative formulation of concrete composition as grams, for 1000 cm³ of concrete

	Portland cement	Sand 0-2 mm	Aggregate 2-4 mm	Aggregate 4-8 mm	Aggregate 8-15 mm	Water ml
Sample 1	400	822	91	595	476	140
Sample 2	400	692	75	423	794	140
Sample 3	400	662	73	317	933	140

After sufficient mixing, the wet concrete with initial activity was poured into a six cylindrical vessel (diameter and height 5 cm). The drying-curing time of the specimens was 30, 60 and 90 days. The leaching test was carried out according to the method recommended by the IAEA.(1) The specimen taken from a cylindrical vessel immediately before the test was immersed in the leaching vessel containing 250 ml of tap water. At each present time interval, the leachant was removed and leached radioactivity was measured using a "EG&G-ORTEC spectrometry system and software."

Results

The effective diffusion coefficients, calculated from equation (1) are shown on Table II., Table III. and Table IV.

$$D = \frac{\pi m^2 V^2}{4S^2} \text{ (cm}^2/\text{s)} \quad (1)$$

where: $D(\text{cm}^2/\text{s})$ is diffusion coefficient, $S(\text{cm}^2)$ is exposed surface area of specimens, $V(\text{cm}^3)$ is volume of specimens, $m(d^{-1/2})$ is gradient of cumulative fraction of radionuclide leached at time $t(\text{s})$, $t(\text{s})$ is time.

Table II., Table III and Table IV, presents "Effect of curing time on the leach rate of ¹³⁷Cs from Portland cement", for different samples.

Table II. Effect of curing time on the leach rate of ^{137}Cs from Sample 1

Cumulative time leached (days)	Diffusion coefficient (cm^2/s)		
	30 days cured	60 days cured	90 days cured
1 - 30	$8.4 \cdot 10^{-10}$	$4.2 \cdot 10^{-10}$	$5.8 \cdot 10^{-11}$
30 - 65	$5.7 \cdot 10^{-10}$	$3.8 \cdot 10^{-10}$	$6.3 \cdot 10^{-11}$
65 - 180	$4.3 \cdot 10^{-10}$	$3.5 \cdot 10^{-11}$	$1.9 \cdot 10^{-13}$

Table III. Effect of curing time on the leach rate of ^{137}Cs from Sample 2

Cumulative time leached (days)	Diffusion coefficient (cm^2/s)		
	30 days cured	60 days cured	90 days cured
1 - 30	$9.5 \cdot 10^{-10}$	$4.8 \cdot 10^{-10}$	$8.9 \cdot 10^{-11}$
30 - 65	$6.7 \cdot 10^{-10}$	$4.2 \cdot 10^{-10}$	$3.8 \cdot 10^{-11}$
65 - 180	$5.9 \cdot 10^{-10}$	$3.8 \cdot 10^{-11}$	$3.5 \cdot 10^{-11}$

Table IV. Effect of curing time on the leach rate of ^{137}Cs from Sample 3

Cumulative time leached (days)	Diffusion coefficient (cm^2/s)		
	30 days cured	60 days cured	90 days cured
1 - 30	$8.1 \cdot 10^{-10}$	$3.6 \cdot 10^{-10}$	$2.5 \cdot 10^{-11}$
30 - 65	$5.2 \cdot 10^{-10}$	$4.2 \cdot 10^{-10}$	$2.0 \cdot 10^{-11}$
65 - 180	$3.8 \cdot 10^{-11}$	$3.6 \cdot 10^{-11}$	$7.8 \cdot 10^{-12}$

Conclusion

All results exhibit the same general characteristics. An enhanced initial period of leaching occurs during the first 25-30 days or so, followed by a distinct reduction in the leach rate which is broadly maintained up to the long period of leaching. The leach behavior of concrete materials can be explained as a combination of two processes; surface wash-off, which is not diffusion controlled, followed by a static diffusion stage. Enhanced initial period of leaching can be explained in terms of a rapid equilibrium being established between spaces present in the surface pores of the Portland cement and ions in solution in the leachant; hence the term wash-off. It is this second stage which is controlled by diffusion and which dominates the long-term leaching behavior of the material. Under these circumstances the effect of increased curing time on the diffusion coefficient becomes apparent. For example, the diffusion coefficient at 90 days curing is almost 2 orders of magnitude lower than that obtained after 30 days curing. Results presented in this paper are examples of results obtained in a 10-year concrete testing project which will influence the design of the engineer trenches system for future central Serbian radioactive waste storing center.

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