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EFFECT OF CURING TIME ON THE FRACTION OF ^{60}CO AND ^{137}Cs LEACHED 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 ^{60}Co and ^{137}Cs have been studied. Leaching tests in concrete were carried out in accordance with a method recommended by IAEA.(1)

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.(2,5)

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.

Materials, methods and experimental conditions

Concrete samples were made of :

- Portland cement, PC-55 MPa
- Sand, fraction: 0-2 mm
- Aggregate, fraction: 2-4 mm; 4-8 mm and 8-15 mm
- Water attested according to Yugoslav standard
- Admixture, super fluidal (super plasticizer)
- Initial activity per sample $A_0 = 7.4.10^6$ of ^{60}Co and ^{127}Cs

In this paper we discuss one representative formulation of concrete composition shown in Table I.(3)

After sufficient mixing, the wet concrete with initial activity was poured into a six cylindrical vessel (diameter and height 5 cm). The drying time of the specimens was 28, 60 and 100 days.

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

Portland cement	Sand 0-2 mn	Aggregate 2-4 mm	Aggregate 4-8 mm	Aggregate 8-15 mm	Water	Admixture
400	692	75	423	794	140	8

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

Figure 1 shows cumulative leach curves of ^{60}Co and ^{137}Cs from Portland Cement at three different curing times. The effective diffusion coefficients, calculated from equation (1) are shown in Table II.

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

where:

D(cm^2/s) is diffusion coefficient

S(cm^2) is exposed surface area of specimens

V(cm^3) is volume of specimens

m($d^{-1/2}$) is gradient of cumulative fraction of radionuclide leached at time t(s)

t(s) is time

Table II

Effect of Curing Time on the Leach Rate of ^{60}Co and ^{137}Cs from Portland Cement

	Diffusion coefficient (cm^2/s)					
Cumulative Time leached (days)	28 days cured	60 days cured	100 days cured	28 days cured	60 days cured	100 days cured
1 - 30	$9.2 \cdot 10^{-10}$	$1.0 \cdot 10^{-10}$	$6.5 \cdot 10^{-11}$	$8.5 \cdot 10^{-10}$	$9.5 \cdot 10^{-11}$	$6.0 \cdot 10^{-11}$
30 - 65	$2.4 \cdot 10^{-11}$	$2.0 \cdot 10^{-10}$	$4.0 \cdot 10^{-12}$	$2.2 \cdot 10^{-10}$	$1.8 \cdot 10^{-10}$	$3.8 \cdot 10^{-12}$
65 - 100	$4.2 \cdot 10^{-11}$	$3.3 \cdot 10^{-11}$	$3.4 \cdot 10^{-13}$	$4.0 \cdot 10^{-11}$	$3.2 \cdot 10^{-11}$	$3.0 \cdot 10^{-13}$

Conclusion

All six profiles in Fig. 1 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

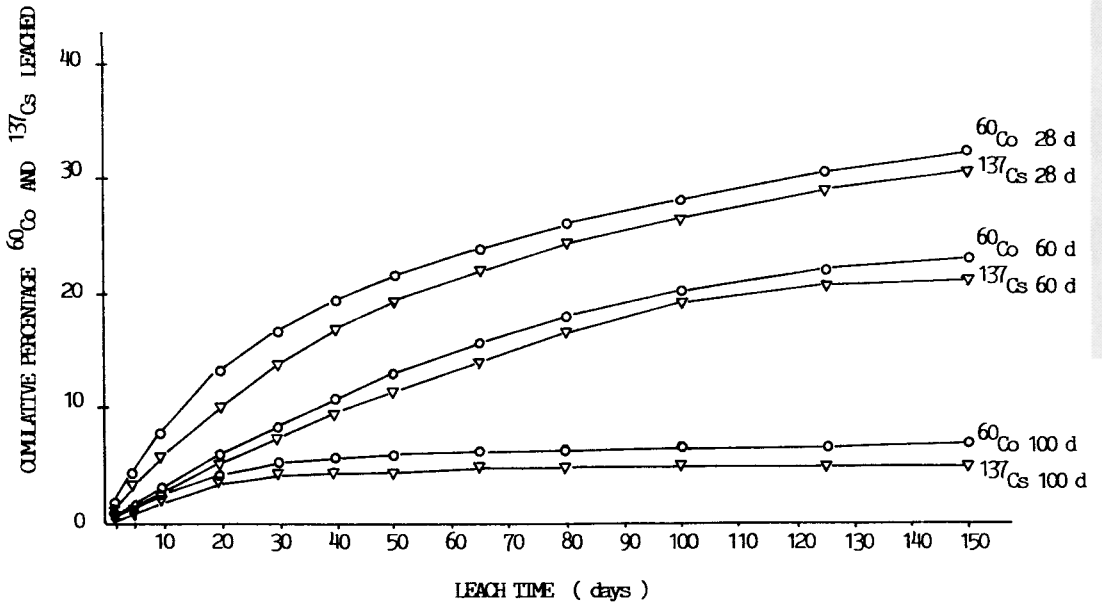

Fig. 1
Cumulative leach curves of ^{60}Co and ^{137}Cs from Portland Cement at three different curing times.

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. When ^{60}Co and ^{137}Cs has been leached from the surface of the waste form, ^{60}Co and specially ^{137}Cs must migrate by longer pathways from the bulk through a depleted surface layer before it can enter the solution.[2] 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 100 days curing is almost 2 orders of magnitude lower than that obtained after 28 days curing.

Results presented in this paper are examples of results obtained in a 5-year concrete testing project which will influence the design of the engineer trenches system for future central Yugoslav radioactive waste storing center.

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