Leaching of ^{137}Cs in cemented waste forms in Perú

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Abstract

The leaching rate of ^{137}Cs from the chemical treatment process in cement matrix has been studied. The solidification matrix was a standard Portland cement mixed with 5–10% of bentonite clay. The cement specimens were prepared from construction cement which is basically a standard Portland cement. The mixtures were cast into 50 mm diameter cylindrical molds with a height of 50 mm, which were then sealed and cured for 28 days prior to the leaching experiments. The leaching rates from the cement matrix for ^{137}Cs : 2 E–05 (cm/d), after 155 days were measured. The results presented in this paper are part of the results obtained in a 2 years concrete testing work which will be used for conditioning of sludge from the chemical precipitation of decontamination waste.

Keywords. Portland cement; radioactive waste; leaching rate; waste management.

1. Introduction

The current management of low and intermediate liquid wastes produced in Peru is a significant task for Peruvian Institute of Nuclear Energy for the treatment and conditioning of radioactive liquid wastes. Some kind of liquid radioactive wastes are expected by the operation of the nuclear research reactor according to its safety analysis. Effluents contaminated with long-lived radioisotopes are produced by the regeneration of the ion exchange resins. They will be treated by precipitation of the main radionuclides in a precipitation plant. A important radionuclide like ^{137}Cs will be present in the sludge of the precipitation as a result of the reaction taking place during chemical treatment steps. The sludge are inmobilized with cement so it is important make the research of the leaching of ^{137}Cs in the cement matrix..

Solidification of concentrates in a matrix is believed to be necessary for final storage , because in the safety assessment of the storage site, flooding of water and accordingly potential contamination of the groundwater due to leaching of the waste, can not be completely excluded when simple mild steel drums are used for packaging.

Although cement has several unfavourable 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. It is generally assumed that the cement leachability of ^{137}Cs and other radionuclides can be reduced by adding minerals like bentonite. **T**he leaching rate of ^{137}Cs from the chemical treatment process in cement matrix has been studied.

2. Materials and methods

The cement specimens were prepared from construction cement which is basically a standard Portland cement (64,18% CaO , 21,86 % SiO2; 4,81% Al_2O_3; 3,23% Fe_2O_3; 0,65 % K_2O; 0,15% Na_2O; 2,41% SO_3 and 0,96% MgO) . The cement was mixed with 100 g of water with ^{137}Cs and bentonite. The mixtures were cast into 50 mm diameter cylindrical molds with a height of 50 mm, which were then sealed and cured for 28 days prior to the leaching experiments .

More than 80 different formulations of mortar form were examined to optimize their sorption properties. In this paper we discuss three representative formulations. The chemical composition of the cement and additives are shown in Table I.

Table I Chemical composition of cement matrix

Samples		M1	M2	M3
Mixture ratio	Cement (g)	293	263	263
	Water (ml)	117	105	105
	Clay (10%) (g)	--	41	--
	Bentonite (10%) (g)	--	--	41
	Activity Cs-137 (MBq)	80	71	71
	Activity Co-60 (MBq)	107	96	96

3. Experimental

Samples for leachability determination were prepared according to the ISO 6961 standard procedure. Leachant was exchanged and analysed for radioactivity after 1, 3, 7 , 10, 14, 21 days, and thereafter every week for 1 month and from there on every month, until 155th day. After each leaching period the radioactivity in the leachant was measured using NaI (Tl) detector and spectrometry system and software. The volume of the leachant in every leaching period was 1100 ml.

4. Mathematical treatment of data

The results are expressed by the leaching rate; R_n(cm/d), as :

$$Rn = \frac{(An \times V)}{(Ao \times S \times Tn)} \qquad (1)$$

where : R_n is the leaching rate, A_n is the amount of the radionuclide leached in the n-th leaching interval (Bq), A_o is the initial amount of radionuclide in specimen (Bq), V is the volumen of specimen (cm^3), S is the geometric surface area of the specimen immersed in leachant (cm^2) and t is the time of the n-th leaching interval (d).

The cumulative fraction leached ,F_n , is calculated from

$$Fn = \frac{\sum An}{Ao} \qquad (2)$$

where : F_n is the cumulative fraction leached, A_n is the amount of the radionuclide leached in the n-th leaching interval (Bq) and A_o is the initial amount of radionuclide in specimen (Bq).

5. Results and discusion

The results of the leaching tests of immobilized simulated sludge produced in chemical treatment are given as the leaching rate, *R*n (cm/d) and the cumulative fraction leached after

155 days. Table III shows that the values are better in the case of the cement matrix with 10 % bentonite.

The results of the specimen measurements can be seen in Table III. The variation of leaching rate and the cumulative fraction leached of ^{137}Cs with time is shown in Fig. 1 and 2, respectively.

The results show that leachability is lower in the case of cemented waste form with 10 % of bentonite. It can be seen that especially, for of ^{137}Cs, show good improvement.

Results presented in this paper are part of the results obtained in 2 years and concrete testing research which will influence the design of a radioactive waste management for a future Peruvian radioactive waste storing centre.

Table II Leaching rate and cumulative fraction leached of ^{137}Cs from various cemented forms

	W/C = 0.40		W/C = 0.40 + 10% clay		W/C = 0.40 + 10% bentonite	
	Cs - 137		Cs - 137		Cs - 137	
Day	Rn (cm/d)	Fn	Rn (cm/d)	Fn	Rn (cm/d)	Fn
1	5.21E-02	6.47E-02	3.69E-02	4.59E-02	2.57E-02	3.20E-02
3	1.36E-02	9.85E-02	8.80E-03	6.78E-02	4.43E-03	4.30E-02
7	5.00E-03	1.24E-01	3.50E-03	8.52E-02	2.34E-03	5.46E-02
10	5.90E-03	1.46E-01	1.10E-03	8.92E-02	1.14E-03	5.89E-02
14	4.30E-03	1.67E-01	8.00E-04	9.29E-02	4.40E-04	6.10E-02
21	2.30E-03	1.87E-01	4.00E-04	9.64E-02	2.00E-04	6.28E-02
28	2.20E-03	2.06E-01	3.00E-04	9.93E-02	2.10E-04	6.46E-02
35	8.00E-04	2.13E-01	3.00E-04	1.02E-01	1.10E-04	6.55E-02
65	1.60E-04	2.19E-01	6.00E-05	1.04E-01	2.00E-05	6.63E-02
95	1.40E-04	2.24E-01	6.00E-05	1.06E-01	2.00E-05	6.70E-02
126	1.30E-04	2.29E-01	5.00E-05	1.08E-01		
155	1.20E-04	2.33E-01	4.00E-05	1.09E-01		

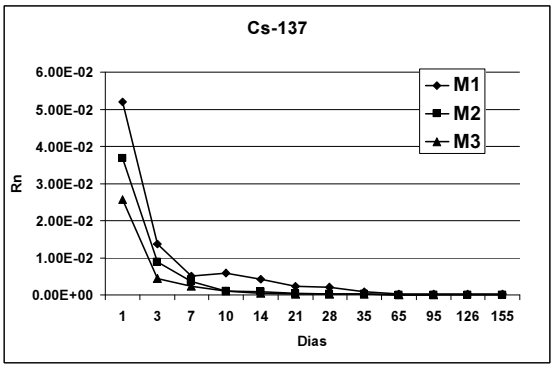

Fig. 1 Leaching rate (Rn) of ^{137}Cs from various cemented waste forms

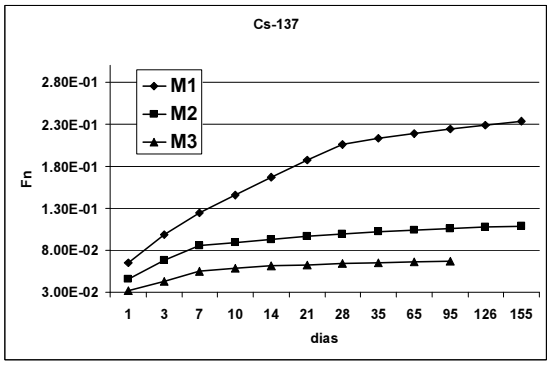

Fig. 1 Cumulative fraction leached of of ^{137}Cs from various cemented waste forms

5. Conclusion

Considering the immobilizing process for treated waste; cementation method was selected. The leachability of a particular type of sludge has been studied. The results revealed that cemented waste forms of sludge with ^{137}Cs exhibited a low leaching rate.

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