THE EFFECT OF AUTOGENOUS HEALING ON THE PENETRATION OF CESIUM-137 THROUGH CRACKS IN CEMENTITIOUS BARRIER MATERIALS

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ABSTRACT

The experiments performed in this research were designed to learn more about the phenomenon of autogenous healing of cracks in cementitious materials used as barrier media in low level radioactive waste management. A novel method of producing cracks in cementitious materials was developed which allowed the accurate measurement of changes in flow rate of a radioactive solution penetrating the cracks undergoing autogenous healing. Three mix designs were studied: (1) 100% cement, (2) 70% cement/30% microsilica and (3) 70% cement/30% fly ash.

Replicate samples were studied using tap water containing Cs-137. In order to study the effect of carbonate on autogenous healing, other replicate samples of the three mixes were studied using tap water containing 10 ppm of added carbonate. All samples tested exhibited a substantial decrease in flow rate with time, indicating that autogenous healing was occurring. For the tap water studies the samples containing fly ash underwent autogenous healing at a rate faster than the 100% cement mix and much faster than the microsilica mix. When carbonate was added to the tap water it caused a definite enhancement in the autogenous healing process in all three mix designs. The results of the Cs-137 analysis of water which had penetrated the cracks indicated that there was no preferential retention of Cs-137 in the cracks under going autogenous healing for any of the situations studied.

INTRODUCTION

Cementitious materials are widely used in a variety of forms as barrier media in radioactive waste disposal facilities. The cracks that can form in cementitious barriers provide a route for water to enter and/or escape from the facility, possibly releasing radioactive material. It is well known that under certain conditions cracks can repair themselves, a phenomenon known as autogenous healing. It has been found that the presence of water is essential for healing. This healing is postulated to be due to the formation of calcium carbonate crystals from calcium hydroxide in the mortar reacting with dissolved carbondioxide or carbonates in the water environment (1). The phenomenon of autogenous healing, though well known for many years, has not been studied to any extent with regard to its effect on the penetration of radioactivity through cracks in cementitious barrier materials used in radioactive waste management, particularly with regard to the effect that the carbonate content of the impinging water has upon the rate of autogenous healing.

This study included the development of a novel method of creating single cracks in experimental cementitious cylinders. The method allows similar cracks to be created in replicate samples. The production of these experimental cracks permits anaccurate measurement of the effect of autogenous healing on the rate of penetration of a Cs-137 solution through the cracks. Also, the effect on autogenous healing caused by the addition of carbonate to the solution was evaluated. And, a measurement of Cs-137 concentration in the penetrating solution was made in order to see if any preferential absorption of Cs-137 by the cementitious material was occurring.

METHODS AND MATERIALS

The autogenous healing effect was studied with five replicate samples of three mix designs using tap water containing Cs-137. In order to study the effect of carbonate on autogenous healing, five other replicate samples of each of the three mixes were studied using tap water containing 10 ppm of added carbonate.

Three mix designs were studied in this experiment: (1) 100% cement, sand and water, (2) 70% cement/30% microsilica by volume, sand and water, and (3) 70% cement/30% fly ash by volume, sand and water. A water to cement ratio of 0.4 to 1 by weight and a cement to sand ratio of 1 to 2 by volume was used in all mix designs. All mixing was carried out with a small electric mixing bowl using the standard procedure specified in ASTM C305-65 (2).

The mortar was initially cast in rubber cylindrical molds 6.0-cm inner diameter by 7.0 cm high. A wooden wedge was placed in the moist mortar at the top of one of the ends. After a sufficient time of curing, the mortar cylinder was removed from the rubber mold and the wedge was removed from the mortar. The specimen was then cracked into two pieces by tapping on a steel chisel placed in the groove formed by the wooden wedge. The two pieces of mortar were then carefully reunited in their original positions and installed into a tightly clamped rubber cylinder. A 6.0-cm o.d. PVC pipe (Fig. 1) was clamped to the rubber cylinder and acted as an upper reservoir for the Cs-137 solution. The pipe was capable of holding approximately 300 ml of solution. Initial flow rates through the cracks of replicate specimens were essentially equalized by adjusting the tightness of the clamps. Equalizing the initial flow rates was felt to be desirable in order to minimize any dependence of

autogenous healing on initial flow rates. An inverted PVC cap was connected to the bottom side of the rubber tube in order to collect the Cs-137 solution which had penetrated through the crack. The solution then flowed through a small tube into a graduated cylinder which allowed an accurate determination of flow rate as a function of time.

RESULTS AND DISCUSSION

The results were analyzed for statistical differences using a two factor (fixed effects) model. Figures 2, 3 and 4 show the results for the three mixes tested using tap water and tap water with carbonate added. For the 100% cement mix, shown in Fig. 2, there was a rapid decrease in flow rate during the first few days. This is felt to be more a result of the settling of loose particles than autogenous healing. The gradual decrease of flow rate thereafter, for both types of solution, was probably due to autogenous healing. The statistical analysis indicated that after 15 days there was a significant difference between the tap water only and the added carbonate results. This implied that the addition of carbonate, over and above the carbonate normally present in the tap water, did enhance autogenous healing. Figure 3 gives the results for the 70% cement/30% microsilica mix. The statistical analysis indicated that after the first day there was a significant difference between the tap water and the added carbonate solution flow rate results. Apparently adding carbonate causes a major enhancement of autogenous healing when microsilica is present. Figure 4 shows the results for the 70% cement/30% fly ash mix. In this case it appears from the figure that the addition of carbonate enhanced autogenous healing. However, the statistical analysis did not reveal any significant differences between the tap water and carbonate added flow rates. Apparently, for the fly ash mix there is sufficient carbonate in tap water to give the maximum effect on autogenous healing.

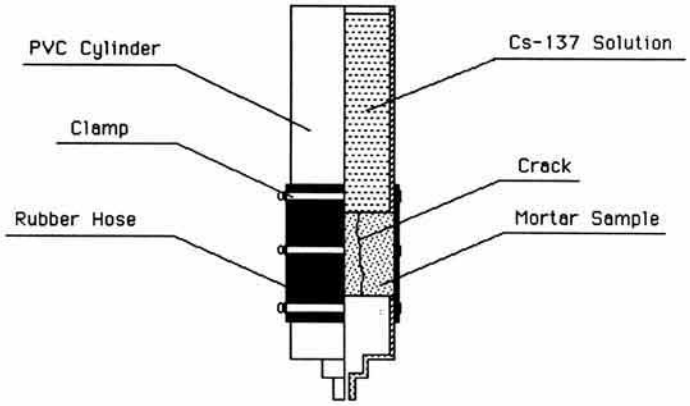

Fig. 1. Experimental apparatus for studying the flow rate of Cs-137 solution through cracks undergoing autogenous healing.

Figures 5 and 6 show the results in another way. Figure 5 compares the reduction in flow rate for the three mix designs using tap water only. The statistical analysis indicated that the 100% cement was different from the 70% cement/30% microsilica for all days, with the 100% cement mix healing faster. The analysis also indicated that the 100% cement was different from the 70% cement/30% fly ash after 7 days, with the fly ash mix healing faster. And, the analysis for the 70% cement/30% fly ash versus the 70% cement/30% microsilica indicated a difference for all days with the fly ash healing faster. Figure 6 compares the reduc-

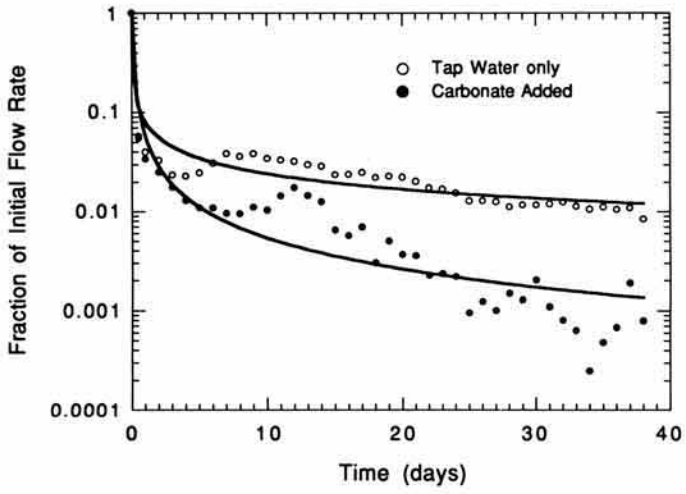

Fig. 2. Reduction of the flow rate of Cs-137 solution through cracks undergoing autogenous healing in the 100% cement mix design for tap water and carbonate added to tap water.

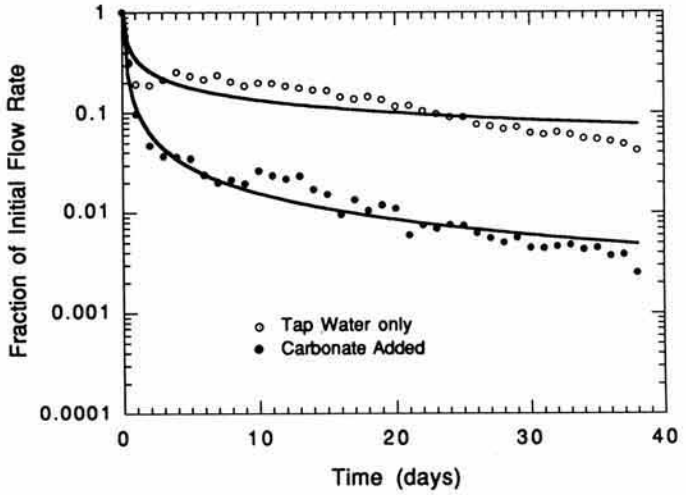

Fig. 3. Reduction of theflow rate of Cs-137 solution through cracks undergoing autogenous healing in the 70% cement plus 30% microsilica mix design for tap water and carbonate added to tap water.

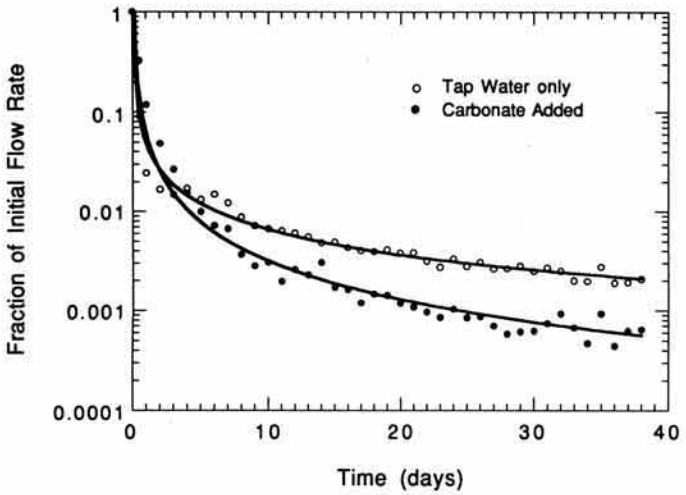

Fig. 4. Reduction of the flow rate of Cs-137 solution through cracks undergoing autogenous healing in the 70% cement plus 30% fly ash mix design for tap water and carbonate added to tap water.

tion in flow rate for the three mix designs for the case where carbonate was added to the solution. Although the flow rates appear to have the same relative relationships as in Fig.5, the statistical test indicated that there were no differences in the flow rates between any of the mix designs. Hence, it appears that the addition of carbonate enhanced autogenous healing in all three mix designs to a common level.

Figure 7 shows, as a function of elapsed time, the centration of Cs-137 in the solution which has penetrated

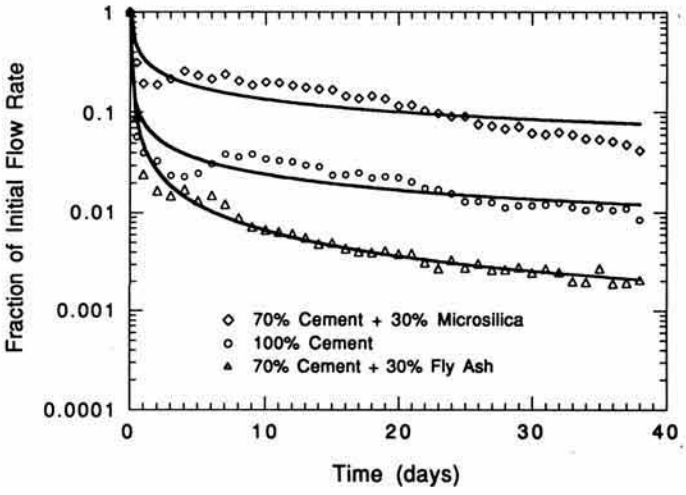

Fig. 5. Comparison of the reduction in flow rate of the tap water only Cs-137 solution for the three mix designs.

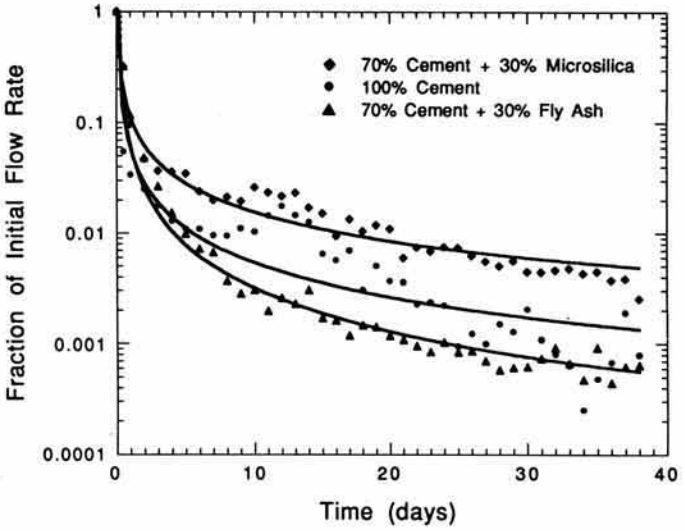

Fig. 6. Comparison of the reduction in flow rate of the tap water plus carbonate Cs-137 solution for the three mix designs.

the cracks undergoing autogenous healing in the 70% cement/30% microsilica specimens. It could not be proven conclusively that there was a statistically significant difference in Cs-137 concentration at any time between the tap water and carbonate added experiments. Studies on the 100% cement and on the 70% cement/30% fly ash specimens gave the same results.

In summary, it appears that, for the tap water experiments, cement containing fly ash undergoes significant autogenous healing and at a rate faster than the 100% cement and much faster than the microsilica mix. If carbonate is

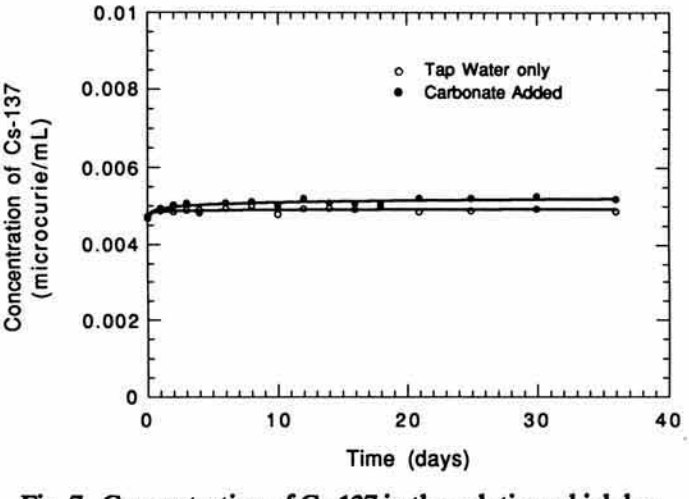

Fig. 7. Concentration of Cs-137 in the solution which has penetrated the cracks undergoing autogenous healing.

added to give a level of about 10 ppm it caused a definite enhancement of the autogenous healing process in all three mix designs, especially for the microsilica mix, to the extent that they all healed at a common rate. And, finally fly ash was shown to undergo autogenous healing to the same extent, whether or not carbonate was added to the tap water. The results of the Cs-137 analysis indicated that there was no preferential adsorption or retention of Cs-137 in the cracks undergoing autogenous healing for any of the situations studied.

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2. ASTM C305-65 (1970). Standard Method for Mechanical Mixing XPB of Hydraulic Cement Pastes and Mortars of Plastic Consistency, American National Standards Institute.