

**Effects of Aging and Decontamination Pressure on the Penetration of Radionuclides into Concrete  
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**ABSTRACT**

Radionuclide penetration into concrete was investigated as function of time and pressure of an applied water spray. Depth profiles of three radionuclides into a concrete surface over the time periods of 20-24 hours, 9 days, and 90 days were found by grinding the surface. The profiles were starkly different between the radionuclides, as Eu-152 deposited close to the surface and did not penetrate further over time while the depth of Cs-137 increased with time. Such differences suggest that response time during a remediation effort is more important for Cs-137 decontamination compared to Eu-152. The depth profiles were also used to assess the water pressure that was needed to remove contamination. Static tests, where the surface was soaked in 0.5M KCl, accessed up to 80  $\mu\text{m}$  into the surface, while ablation through high pressure washing removed approximately 10-20  $\mu\text{m}$  of the surface. Similar grind profiles were then collected on decontaminated coupons, displaying that high pressure washing drove Sr-85 into the surface, decreasing the removal of the nuclide. As such, radionuclide specific decontamination may be required because of the varied response times and pressures effects for each radionuclide.

**INTRODUCTION**

The release of radiological material from a nuclear incident (meltdown, radiological dispersal device, or improvised nuclear device) has the potential to cause extensive radiological contamination requiring rapid decontamination. Rapid decontamination by readily accessible means (e.g., fire housing with salt additives[1-3], high pressure washing[4, 5]) can reduce dose to first responders [2] and reopen critical infrastructures before radionuclides become fixed within porous structural materials such as concrete.[6] Once fixed, more destructive decontamination means (e.g., scabbling and demolition) are often the only methods able to adequately reduce radioactivity from contaminated structures.[6] Rapid decontamination techniques may also influence fixation of radionuclides within building materials through increased penetration, raising concerns in regards to final decontamination methods. Final decontamination, where dose level requirements would be much lower than those for the first responders and critical personnel in the immediate aftermath, may be required before the repopulation of affected areas. Therefore, understanding the effect of rapid decontamination techniques on the penetration of radionuclides into concrete will greatly aid the development of decontamination protocols for the immediate aftermath and later on in the recovery effort of a nuclear incident.

This study aims to understand the effect of the rapid decontamination methods of high pressure washing and washing with salt additives on the penetration of radionuclides into concrete. To this end, we first identified how penetration profiles of Cs-137, Sr-85, and Eu-152 evolve over time. These three radionuclides were chosen to span a number of important ion characteristics and serve as surrogates for commonly observed nuclides after a radiological release (Sr-85 for Sr-90, Eu-152 for other lanthanides and Am-241).[7] The depth profiles enabled us to infer the depth within the surface that each decontamination method is able to access. The depth profiles after decontamination suggested that salt additives and high pressures may cause Cs-137 and Sr-85 to penetrate further into concrete, respectively, raising concerns for final decontamination and indicating that radionuclide specific response may be required.

## **EXPERIMENTAL**

### **Coupon preparation**

Concrete coupons were prepared using Quikrete® standard 4000 psi concrete mix. The concrete mix was prepared using the least amount of water possible, resulting in low porosity and the strongest concrete. To prepare standard concrete coupons, 83 mL of deionized water was added per kg of concrete mix, which was mixed for three minutes and allowed to rest. The wet concrete was mixed again for another three minutes at which time it was ready for use. The wet mixture was scooped into 1.5-in. tall x 1.25-in. diameter pieces of PVC pipe and pressed to remove voids in the concrete. The top surface was smoothed with a spatula to create a layer of cement on top of the concrete in an attempt to represent unaged concrete. The coupons were removed from the mold after 10 to 15 days and were ready for use after 40 days of curing.

### **Coupon contamination**

Concrete coupons were spiked with 200  $\mu\text{L}$  of solution containing 10,000 CPM  $\text{mL}^{-1}$  of Cs-137, Sr-85, and Eu-152 and allowed to dry for two hours in a fume hood. After drying the coupons were carefully wrapped in plastic wrap, bagged, and counted on an Ortec Detective for 5 minutes. Contaminated surfaces were placed (within bags) at the surface of the detector for maximum efficiency. Cs-137 and Sr-85, counts were determined at 661 and 514 keV, respectively, while Eu-152 was counted at both 121 and 344 keV. The samples were then placed with the contaminated surface facing up and aged for 20-24 hours, 9 days ( $\approx 200$  hours), and 90 days ( $\approx 2000$  hours) within the bags.

### **Decontamination testing**

Three methods of decontaminating coupons were tested: static, flow, and high pressure washing. Each of these methods are detailed elsewhere.[1, 4] In brief, static tests consisted of placing the coupon contaminated side down within a petri dish suspended on 4 glass beads. The beads ensured that the entire surface was available to the decontamination solution. The solution was then spiked into the dish so that at least 5 mm of the coupon was below the solution surface. The coupon was exposed to the solution for one hour before it was removed, dabbed lightly dry with a wipe, and recounted. In flow tests, 500 mL of solution was flowed over the contaminated surface of the coupon at  $100 \text{ mL min}^{-1}$ . [2] For flow and static tests, 0.5 M KCl was used as a chemical means to desorb radionuclides from the concrete coupons. High pressure tests were performed within a chamber where a motor brought the contaminated coupon through the spray path generated by a 200 psi, 1.2 gpm high pressure washer (Ryobi) at  $5 \text{ mm sec}^{-1}$ . [4] A  $40^\circ$  degree spray nozzle was located 15 cm from the surface, resulting in a pressure of 80" water (2.9 psi) at

the coupon surface as measured with a Pitot probe. Then, coupons were dabbed dry with a wipe and recounted.

### Depth profiles: Grind tests

The penetration of radionuclides into the coupons was then determined by using a sand paper grinding procedure adapted from Barescut, et al.[8, 9] Strips of sand paper, 2 inches wide ( $\approx 5$  cm) and 4.5 inches long (11.5 cm) were cut from 320 grit, silicon carbide abrasive sand paper (3M). The coupon was then placed contaminated side down and ground back and forth 10 times leaving  $\frac{1}{4}$  of an inch between the coupon and the edge of the sand paper. The coupon was removed and the sand paper was carefully rolled, secured with transparent adhesive tape (Scotch), and placed in the gamma counting tube. Sand paper rolls were counted for 1 hr. on a Wiz<sup>2</sup> Automatic Gamma Counter (Perkin Elmer). The mass removed from the coupon was considered to be the average of the difference in weights before and after the grind of both the sand paper and the coupon. This mass was converted to a depth removed using the density of concrete since the small depth removed could not be measured due to the heterogeneous underside of the coupon. This grind procedure was performed 20 times for each coupon, after which the coupon was re-bagged and counted on the Detective.

### Profile normalizations

Depth-activity profiles were normalized to percent activity to remove scatter and allow for more substantive comparisons between the profiles. The fraction activity removed of each radionuclide at each depth ( $f_{act,i}$ ) was determined using the activity of grind  $i$  in counts per minute ( $CPM_i$ ) divided the total CPM within all grinds and the fraction of each radionuclide removed from the coupon ( $f_{removed}$ ):

$$f_{act,i} = \frac{CPM_i}{\sum CPM / f_{removed}} \quad (1)$$

From this, the percent of radioactivity remaining within the coupon at depth  $i$  ( $f_{remain,i}$ ) can be calculated:

$$f_{remain,i} = \left[ 1 - \sum_1^i f_{act,i} \right] \times 100 \quad (2)$$

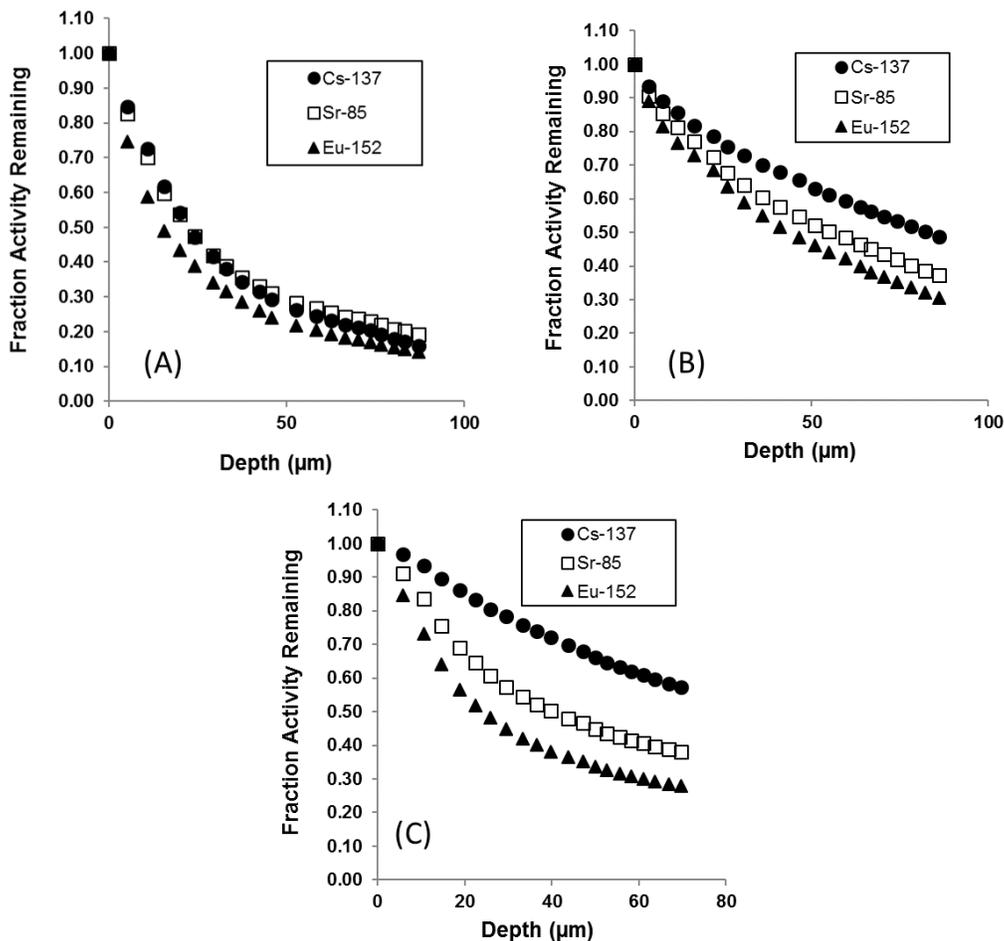
## RESULTS/DICUSSION

### Grind profiles with time

We began with coupons that were not decontaminated to establish a baseline of radionuclide penetration into concrete. Each radionuclide had different penetration behavior over time, highlighting each ions' chemical properties. The limited solubility of trivalent Eu-152 caused the radionuclide to precipitate near the surface and not penetrate deeply into the coupon surface. The penetration depth of Eu-152 did not drastically increase from 20 hours to 90 days (Fig 1). As a nuclide penetrates concrete within a solution, the pH of the solution increases due to the carbonates and other salts bound within the cement fraction of concrete. The increase in pH combined with sorption sites on the aggregate minerals increases the affinity of Eu-152 for the solid phase. Eu-152 then strongly binds through mostly irreversible interactions preventing it from penetrating further. Conversely, the penetration of highly soluble Cs-137 increases over time (Fig.1). Monovalent Cs-137 is not effected by the increased pH within the concrete

pores. Therefore, the only retention mechanism for Cs-137 on concrete is reversible sorption to negative charge sites found on the aggregate minerals. Accordingly, Cs-137 retention drastically increases over time (Fig. 1). Sr-85, which is less soluble than Cs-137 but more so than Eu-152, displays penetration depths somewhat in between the other radionuclides (Fig 1.) Overall, the depth of penetration followed Cs-137 > Sr-85 > Eu-152 which directly follows ion solubility.

The penetration profiles also offer some information on the amount of the surface that requires removal during final decontamination. For instance, if a reduction of 50% in Cs-137 activity is required to return dose to acceptable levels, 15-20  $\mu\text{m}$  of the surface would need to be removed after 20-24 hours. As time increases, 70 – 80  $\mu\text{m}$  of the surface would need to be removed after 9 days and over 120  $\mu\text{m}$  after 90 days (extrapolated). On the other hand, only 20-30  $\mu\text{m}$  of the surface would need to be removed at all time periods for the same percent removal of Eu-152. Such a difference in surface removals indicates that nuclide specific decontamination methods may be prudent especially in areas where activity is dominated by a single radionuclide.



**Figure 1.** Percent of the radionuclide beyond each depth of concrete for Cs-137 (black circles), Sr-85 (white squares) and Eu-152 (black triangles) after aging for (A) 20 hours, (B) 9 days, and (C) 90 days.

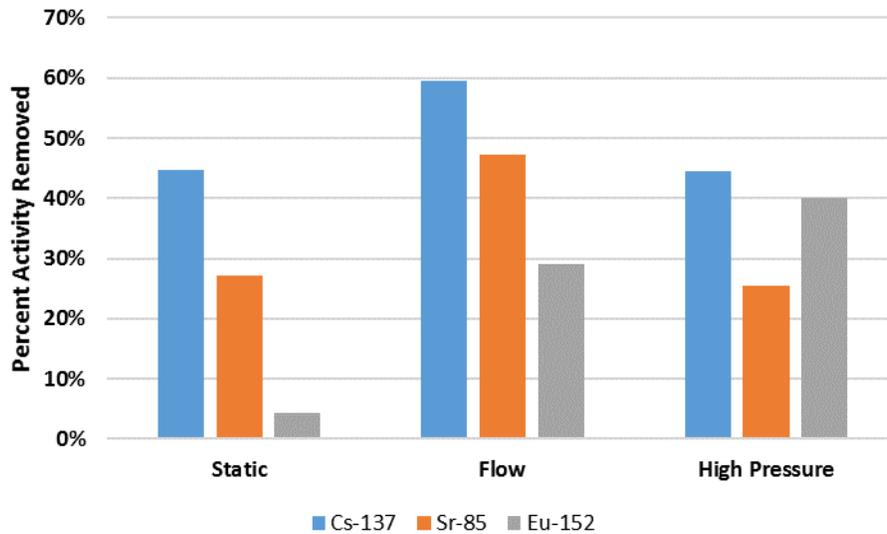
## Depth accessed by decontamination methods

Using the depth profiles, we could then assess the depth that each of the decontamination methods is able to access into the concrete coupons. Static removals after a 24 penetration time were 43%, 27%, and 3% for Cs-137, Sr-85 and Eu-152, respectively. (Fig. 2) These relative ranks follow the expected affinity of each radionuclide for concrete and also correspond to the ability of each radionuclide to penetrate (Cs>Sr>Eu). The removals indicate that a least 10-15  $\mu\text{m}$  into the surface was accessed during the static tests, though this depth is likely much deeper since chemical desorption is the only removal mechanism in the no flow environment.

We can better estimate the depth accessed using a best case scenario of chemical desorption of Cs from concrete aggregate. Sorption to concrete aggregate is the dominant retention mechanism of Cs within concrete. In such a case, removals of Cs-137 were around 40-50% at equilibrium depending on the mineralogy of the aggregate [10] and at least 80 to 100% of the contamination on the coupon would need to be accessed to reach the observed 43% removal from the coupon. Therefore, the depth accessed by the 0.5M KCl solution would increase to at least 80  $\mu\text{m}$ . With that being said, competition between the radionuclides for sorption sites and non-linear desorption isotherms may also increase the removals closer to the surface while decreased activity due to shielding/distance from the driving of radionuclides further into the surface cannot be discredited.

For flow tests chemical desorption is still the dominant mechanism of removal. However the flow decreases the effect of diffusion kinetics through turbulent mixing and increases the affinity of the ions for the aqueous phase by changes in the chemical potential (higher volume used). As such, the removal of all the radionuclides were larger in flow tests compared to static tests (Fig. 2). The increased removals corresponded to an increase of depth accessed by the decontamination solution. The depth accessed was at least 25-30  $\mu\text{m}$  and would need to be over 100  $\mu\text{m}$  into the surface if the maximum desorption is achieved. However, the changes in chemical potential and mixing likely reduce the total depth needed since removal percentages are expected to be higher than for tests without a physical mixing component.

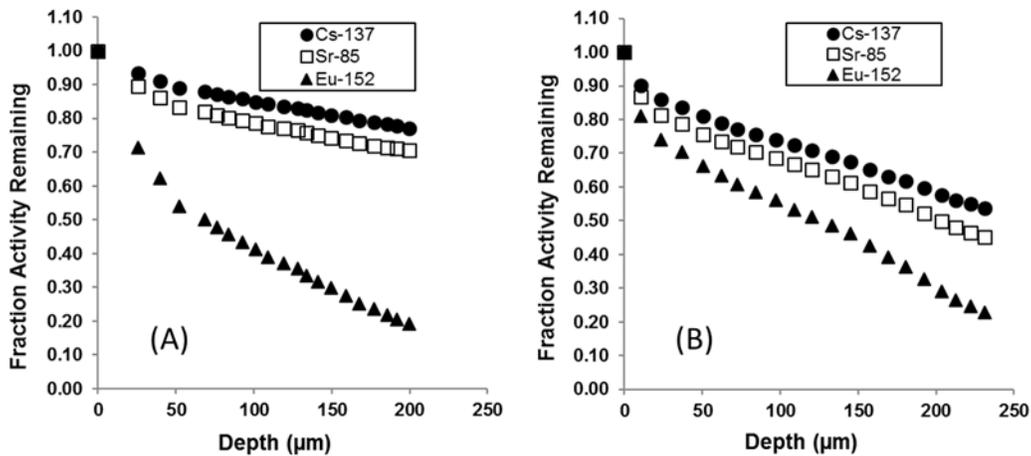
During high pressure decontamination, we changed the decontamination solution to tap water so that the main removal mechanism would be ablation of the surface. Removals using the high pressure system were drastically increased for Eu-152 due to its high affinity for the surface, while Sr-85 and Cs-137 removals were similar to those seen in the static experiments (Fig. 2). Eu-152 resided closer to the surface making removal by ablation more likely. These removals indicate that approximately 10-20  $\mu\text{m}$  of the surface is ablated, removing the radionuclides along with surface particles. The surface ablation should remove a similar amount of the three radionuclides since all three have similar depth profiles after 24 hours (Fig. 1A). Therefore, the lower removals of Sr-85 compared to Cs-137 and Eu-152 indicate that another phenomena is likely present, which led us to investigate the possibility of the high pressure system driving Sr-85 into the concrete.



**Figure 2.** Removals of Cs-137 (blue), Sr-85 (orange), and Eu-152 (grey) during static, flow, and high pressure decontamination tests.

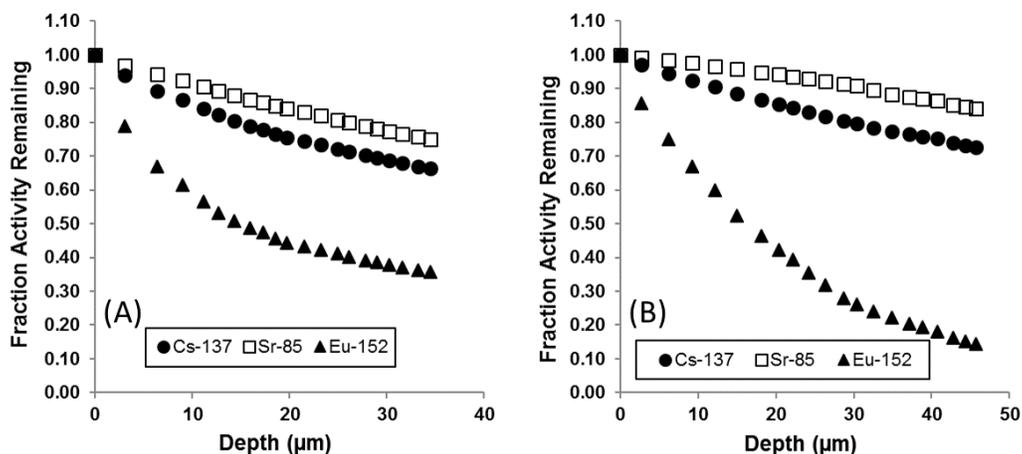
### Depth profiles following decontamination

Depth profiles of radionuclide within decontaminated coupons highlight the effect of decontamination pressure on radionuclide penetration. After decontamination, we aged the coupons for 90 days before grinding to magnify the difference between the radionuclide as seen for the 90 days aged coupons without decontamination. In both flow and static decontaminated coupons there was no change seen in the relative amount of penetration when comparing the radionuclides, as Cs-137 penetrated furthest followed by Sr-85 and Eu-152 (Fig. 3). For static tests, Cs-137 and Sr-85 seem to penetrate further into the surface compared to coupons that were not decontaminated (Fig. 1C vs. Fig. 3A). The addition of water and salt coupled with the longer contact time after the initial contamination likely desorbs/dissolves Cs-137 and Sr-85 allowing the radionuclide to penetrate further into the coupon. Sr-85, which is thought to sorb to concrete by precipitation than sorption, is more readily dissolved increasing penetration making its depth profile similar to Cs-137. Eu-152 does not display these effects, likely due to its low solubility and irreversible interactions. Flow tests did not cause increased penetration of radionuclides into concrete as seen in static tests. The lower contact time in flow tests decreased the amount of decontamination solution that penetrated the surface.



**Figure 3.** Radionuclide depth profiles after decontamination for static tests (left) and flow tests (right) using 0.5 M KCl.

Radionuclide depth profiles were distinctly different after decontamination by high pressure washing compared to non-decontaminated coupons and coupons decontaminated by static and flow tests. Most apparent of the changes in depth profile was that Sr-85 penetrated further into the surface compared to Cs-137 (Fig 4). We observed that Sr-85 removals were lower than those of Cs-137 and Eu-152 during pressure washing even though each removal should be similar due to the matching depth profiles. The underlying reason for the difference now becomes more apparent, as Sr-85 is forced into the coupon further than the other radionuclides during high pressure washing, decreasing removals. The retention mechanism of Sr-85 is likely precipitation causing it to not be bound to the mineral phase but rather precipitated on the outside of the solid matrix. Such a weak bond to the solid phase makes the nuclide more susceptible to removal from the solid by the high pressure and forced deeper into the surface rather than being removed with the solid material. We performed a second replicate to confirm that this change in penetration was reproducible and found good agreement (Fig. 4).



**Figure 4.** Radionuclide depth profiles after decontamination using high pressure tap water in two replicate coupons.

## CONCLUSIONS

The response time and wash pressure have distinct effects on the penetration of radionuclides into concrete. Over time, the depth of penetration followed Cs-137 > Sr-85 > Eu-152 which directly follows ion solubility. Such depth profiles indicate that response time is radionuclide dependent as 15-20 µm of the surface would need to be removed to reduce Cs-137 activity by 50% after 20 hours while, after 9 days, 70 – 80 µm of the surface would need to be removed. On the other hand, only 20-30 µm of the surface would need to be removed to reduce Eu-152 levels by 50% at any of the time periods studied (20 hrs. to 90 days). Therefore, the response time is more critical for Cs-137 than Eu-152 removals.

The use of high pressure washing to decontaminate concrete surfaces has similar, if not increased, removals for the radionuclides Cs-137, Sr-85 and Eu-152 compared to static and flow tests with the added benefits of decreased contact time and water use. However, high pressure washing drives Sr-85 into concrete raising concern on the method's effectiveness in removing soluble radionuclides that do not have strong interactions with mineral phases (Sr-90, Na-24, Co-60, etc.). This ineffectiveness should be considered when selecting a decontamination method after an incident as nuclide specific pressure may be required. Overall, creating radionuclide specific response times and decontamination means may increase effectiveness of dose reduction and final decontamination following a nuclear event.

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