THE ARGONNE SUPERGEL FOR CBRN DECONTAMINATION

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ABSTRACT

- The Argonne SuperGel was developed between 2003 and 2015 to fill a gap in our nation's capability to quickly decontaminate important structures following a radiological or nuclear release event. Specifically, the decontamination technology was developed to minimize damage to monuments, high valued structures, and critical infrastructure while reducing environmental and health impacts. An important criterion during its development, common reagents were employed that could be easily acquired in order to minimize the timeline for its deployment. Its current formulation uses off-the-shelf super-absorbing hydrogels common to the food and agricultural industry and common salts. Over the years, two formulations of the Argonne SuperGel have been developed to specifically target radioactive cesium contaminations and then, more generically, actinide and fission product contaminations. A biodegradable derivative of phosphoric acid is used in small amounts to promote the removal of insoluble actinide species.
- Since its development, we have had the opportunity to test the SuperGel in the removal of legacy contaminations in hot cell facilities and former glovebox facilities at Argonne. This has provided a unique opportunity to evaluate the SuperGel on a range of contaminants outside the original specifications for its use.
- Concept to Pilot-Scale demonstration in 18 months
  - Long chemical shelf life, common reagents, cost effective
  - Spray and vacuum technology is off-the-shelf
- Waste is compatible with low level radioactive waste regulations

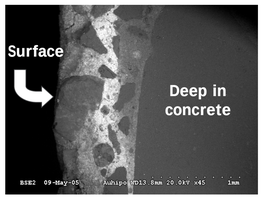
METHODS

- For development studies
  - Building materials were contaminated with Cs-137 or Am-241 solution and allowed to age.
  - Salt-based wash solutions were mixed with *aggregate or crushed materials* to determine the sorption kinetics and equilibrium.
  - Promising wash solutions from were used to reconstitute the hydrogel to produce formulations of the Argonne SuperGel for *coupon* testing.
  - Coupons were contaminated with Cs-137 or Am-241 and aged prior to being held in contact with the SuperGel.
  - After SuperGel removal, coupons were counted to determine Decontamination Factors (DFs).

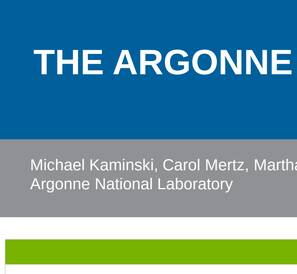
- Blg. 350 trials
  - Applied and removed SuperGel by trowel, sprayed with water and wiped clean of residue.
  - For painted surface and a couple bare concrete, a control was performed where the area was wiped with a dry towel first, measured for total radiation, sprayed with water and wiped, and measured for radiation to check for loose contamination.

METHODS (CONT.)

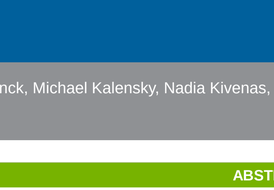
- The gel were prepared from an anionic polyacrylamide/polyacrylate (referred to as "PAM/30%PAA", Hydrosource Green Canteens, Castle International, USA).
  - The anionic gel was prepared at a cross-linked-to-linear ratio of 99/1 [poly(acrylamide) containing 30% anionic charge as the acrylate]
- Various sequestering agents were added to the gel as a dry powder during the gel preparation at 10% by mass and included crystalline silicotitanate (CST, IONSIV, Universal Oil Products), monosodium titanate (MST, 10% suspension, Optima Chemical Group, LLC), cellulose acetate (CA, Aldrich), and others.
- Solutions of ammonium chloride (NH_4Cl, 99.5 + %, A.C.S. reagent, Sigma-Aldrich), potassium chloride (KCl, Analytical AR Reagent, Mallinckrodt), sodium carbonate (Na_2CO_3, anhydrous, granular AR ACS Primary Standard, Mallinckrodt), and etidronic acid (hydroxyethane diphosphonic acid HEDPA, Aldrich) were prepared with deionized H_2O (~18 MΩ•cm resistivity at 25°C).
- For gamma analysis, all monolithic samples were wrapped in plastic prior to movement and analyzed on an ORTEC high-purity germanium detector (HPGe).
  - For Cs-137, the samples were counted on a platform set at 20 cm from the detector face.
  - For Am-241, the samples were placed against the detector face.
  - Each coupon was analyzed for at least 180 seconds of live-time (Cs-137 at 662 keV and 59.5 keV of Am-241).



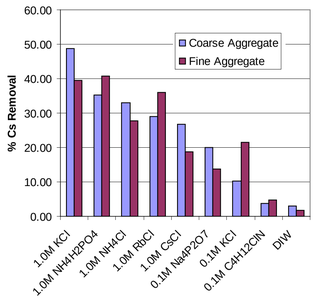
Concrete aggregate fraction less than 0.25 mm at typical of many U.S. based concretes. Particles are predominantly quartz (which can be different colors) although many minerals are present.

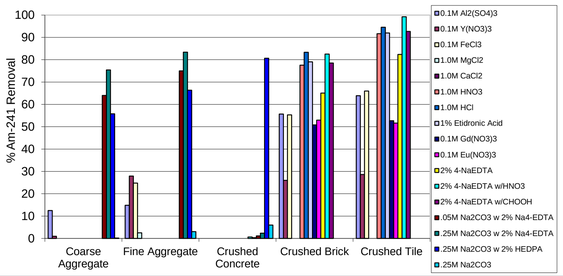


Electron micrographs showing the cross sections of concrete (top) and brick (bottom) test pieces and contaminant penetration. Gold/cesium is represented as white in the image. Field of view in bottom image is ~2 cm.



Large and medium size grains of concrete aggregate from tropical climates. Note the complex, macro-porous texture of the two coral fragments. Calcite, aragonite, and quartz minerals predominate.





- Decontamination from concrete aggregate (fine and coarse) (left, top) using various wash solutions for Cs-137.
- Data shows that potassium chloride and ammonium dihydrogen phosphate can remove cesium effectively from concrete, brick, and tile (not shown).
- Cesium does not Inherently bind strongly to brick material, tile (not shown), or cement (not shown).
- Am decontamination (bottom) required a different approach with carbonate ions and degradable chelators (HEDPA).

DECONTAMINATION OF PLUTONIUM FROM FORMER PU FUEL FABRICATION FACILITY AT ARGONNE

Original Pu fuel fabrication facility Blg. 350 operated from 1959 until 1973 at Argonne National Laboratory.

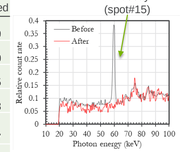
New Brunswick Laboratory operated the Blg. 350 for its mission and used the area for receipt and storage of materials.

Argonne SuperGel was applied (left) for a 2-3 hr dwell time and then removed (middle) and repeated, if necessary. The final area (right) was counted by HPGe and Ludlum α/β probe.

The high bay area has since been converted for safeguards programs, but Pu hotspots (15) remained from legacy Pu work.

Spot #	Description	DF (α)	$A_{\alpha,i}$	$A_{\beta,i}$	% α removed
9	Bare concrete	**1.4**	29	223	27.59
2	Bare concrete	**3.6**	362	327	72.10
11	Painted concrete	**64.5**	387	292	98.45
4	Painted concrete	**10.5**	63	267	90.48
6	Painted + bare concrete	**108**	108	252	99.07
15	Bare concrete	**1080**	1080	455	99.91

Am-241 from Pu decay (spot#15)



DF's were as high as 1080, and 13 of the 15 spots were cleaned to free release standards (~20 cpm alpha).

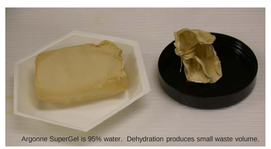
PILOT SCALE TESTING ON WALL-TYPE TEST PLATFORM

- Tests conducted by Batelle at Idaho National Laboratory
- Concrete coupons (15 cm × 15 cm, and 4 cm thick) were prepared in a single batch from Type II Portland cement (Burns Brothers Redi-Mix, Idaho Falls)
- Am-243 applied by aerosol solution onto surface (~50nCi/coupon)
- SuperGel (99/1 cross linked/linear polymer) reconstituted in 0.5M K_2CO_3, 0.025 M HEDPA at 100% hydration
- Samples were aged for two weeks
- SuperGel applied by brush
- Samples counted in front of intrinsic high purity germanium detector (Canberra LEGe Model GL 2825R/S, Meriden, CT
- Vacuumed using Shop-Vac (4.5 HP)
- Average removal was 67% in single application of SuperGel

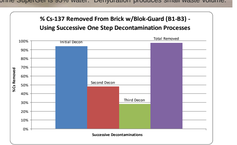
	Pre-Decon Activity (nCi/Coupon)	Post-Decon Activity (nCi/Coupon)	%R
	52	10	80%
	56	21	62%
	51	18	65%
	42	16	62%
Avg	**50**	**17**	**67%**
SD	6	5	9%

CONCLUSIONS AND NEXT STEPS

- NH_4^+ and K^+ ions work best in wash solution for cesium decontamination while carbonate/HEDPA works well for Americium
- Tile can be decontaminated of Cs and Am much more easily
- Brick porosity a serious problem
  - Applying a sealer dramatically improves DFs (graph at bottom right)
- Conducted field trial in the K-wing spent fuel hot cell in Blg. 205 at Argonne to remove contamination from corroded waste barrels and transfer trolley lines
- Conducted field trials in alpha gamma hot cell facility at Argonne to remove loose contamination from corroding steel floor
- Field trials show that the SuperGel is effective on legacy contaminations and not just "fresh" contamination for which it was designed.
- The capacity for water uptake has led to interest in using the SuperGel to facilitate ***chemical and biological agent decontamination*** to increase the dwell time of solution-based decon agents or to soak the de-activating solutions.



Argonne SuperGel is 95% water. Dehydration produces small waste volume.



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