EPA 600/R-13/204 | September 2013 | www.epa.gov/ord



# **Technology Evaluation Report**

Decontamination of Concrete and Granite Contaminated with Americium-243



Office of Research and Development National Homeland Security Research Center

# **Technology Evaluation Report**

Decontamination of Concrete and Granite Contaminated with Americium-243

UNITED STATES ENVIRONMENTAL PROTECTION AGENCY RESEARCH TRIANGLE PARK, NC 27711

## DISCLAIMER

The U.S. Environmental Protection Agency (EPA), through its Office of Research and Development's National Homeland Security Research Center, funded and managed this technology evaluation through Contract No. EP-C-10-001 with Battelle. This report has been peer and administratively reviewed and has been approved for publication as an EPA document. The views expressed in this report are those of the authors and do not necessarily reflect the views or policies of the Agency. Mention of trade names or commercial products does not constitute endorsement or recommendation for use of a specific product.

Questions concerning this document or its application should be addressed to:

John Drake National Homeland Security Research Center Office of Research and Development U.S. Environmental Protection Agency 26 West Martin Luther King Dr. Cincinnati, OH 45268 513-569-7164 drake.john@epa.gov

## ACKNOWLEDGMENTS

Contributions of the following individuals and organizations to the development of this document are gratefully acknowledged.

#### U.S. Environmental Protection Agency (EPA)

John Drake, Office of Research and Development (ORD)/NHSRC Kathy Hall, ORD/NHSRC Emily Snyder, ORD/NHSRC James Mitchell, Region 5 Scott Hudson, Office of Emergency Management (OEM), Consequence Management Advisory Team (CMAT)

#### **Battelle Memorial Institute**

#### U.S. Department of Energy's Idaho National Laboratories

# Contents

DISCLAIMER	. ii
CKNOWLEDGMENTS	iii
ontents	. ii
bbreviations/Acronyms	iv
xecutive Summary	vi
.0 Introduction	. 1
<ul> <li><b>0</b> Technology Description</li></ul>	3
<ul> <li>0 Experimental Details</li></ul>	5 8
<ul> <li>.0 Quality Assurance/Quality Control.</li> <li>4.1 Intrinsic Germanium Detector.</li> <li>4.2 Audits</li> <li>4.3 QA/QC Reporting.</li> </ul>	.11 .12
<ul> <li><b>.0</b> Evaluation Results and Performance Summary</li></ul>	.13
.0 References	19

## Tables

Table 3-1.	Concrete Characterization	5
Table 3-2.	Number of Coupons Included in Technology Evaluation	7
Table 3-3.	Details of Each Testing Time Period	. 10
Table 4-1.	Calibration Results - Difference (keV) from Th-228 Calibration Energies	. 11
Table 4-2.	NIST-Traceable Eu-152 Activity Standard Check	. 12
Table 5-1.	RRII Am-243 Decontamination Efficacy Results	. 14
Table 5-2.	ASG Am-243 Decontamination Efficacy Results	. 15
Table 5-3.	Operational Factors of RRII	. 17
Table 5-4.	Operational Factors of ASG	. 18

# Figures

Figure 3-1. Surface finish of concrete and granite coupons	6
Figure 3-2. Demonstration of contaminant application technique	7
Figure 3-3. Small test stand.	8
Figure 3-4. Rinsing and vacuuming RRII from concrete coupon	8
Figure 3-5. ASG before application, as applied to coupon, and during vacuum removal	

# Abbreviations/Acronyms

Am	americium
ANSI	American National Standards Institute
ASG	Argonne SuperGel
Bq	becquerel(s)
°C	degree(s) Celsius
cm	centimeter(s)
CBRN	chemical, biological, radiological, and nuclear
CMAT	Consequence Management Advisory Team
DARPA	Defense Advanced Research Projects Agency
DF	decontamination factor
DHS	U.S. Department of Homeland Security
DI	de-ionized
EAI	Environmental Alternatives, Inc.
EPA	U.S. Environmental Protection Agency
Eu	europium
h	hour(s)
HSRP	Homeland Security Research Program
ICP-MS	inductively coupled plasma mass-spectrometry
IEEE	Institute of Electrical and Electronics Engineers
INL	Idaho National Laboratory
	kilo electron volts
keV	
mL	milliliter(s)
L	liter(s)
Lpm	liters per minute
m	meter(s)
m <sup>2</sup>	square meter(s)
mm	millimeter(s)
nCi	nanoCurie(s)
NHSRC	National Homeland Security Research Center
NIST	National Institute of Standards and Technology
NPP	nuclear power plant
%R	percent removal
OEM	Office of Emergency Management
ORD	Office of Research and Development
PE	performance evaluation
PPE	personal protective equipment
QA	quality assurance
QAPP	Quality Assurance Project Plan
QC	quality control
QMP	Quality Management Plan
RCT	radiological control technician
RDD	radiological dispersion device
Νυυ	radiological dispersion device

RML	Radiological Measurement Laboratory
RRII	Rad-Release II
RSD	relative standard deviation
Th	thorium
TTEP	Technology Testing and Evaluation Program

### **Executive Summary**

The U.S. Environmental Protection Agency's (EPA's) Homeland Security Research Program (HSRP) is helping to protect human health and the environment from adverse impacts resulting from Chemical, Biological, Radiological and Nuclear (CBRN) contamination whether it results from an intentional act (for instance, terrorism) a criminal act or an unintentional act, (such as a natural disaster or industrial accident). One way HSRP helps to protect human health and the environment is by carrying out performance tests on technologies relevant to homeland security. Through its Technology Testing and Evaluation Program (TTEP), HSRP recently evaluated the performance of Environmental Alternatives, Inc.'s Rad-Release II (RRII) and Argonne National Laboratory's SuperGel (ASG) intended specifically for decontamination of radiological contamination. These technologies were evaluated for their ability to decontaminate surfaces contaminated with radioactive americium from the surface of unpainted concrete and split face granite such as might result from terrorist use of a radiological dispersion device (RDD) or from a Nuclear Power Plant (NPP) accident.

RRII was applied as a liquid with spray bottles and removed with a water rinse and vacuum. ASG was applied as a gel and removed with a vacuum. Prior to the application of each decontamination technology, 15 centimeter (cm)  $\times$  15 cm unpainted concrete and split face granite coupons were contaminated with liquid aerosols of Am-243 and placed in a vertical test stand. Following manufacturer's recommendations, the decontamination technologies were applied to all the coupons on the test stand. Thereafter, the residual activity on the contaminated coupons was measured. Important deployment and operational factors were also documented and reported.

A summary of the evaluation results for RRII and ASG is presented below while a discussion of the observed performance can be found in Section 5 of this report.

**Decontamination Efficacy:** The decontamination efficacy (in terms of percent removal, %R) attained by RRII and ASG was evaluated following contamination of the coupons with approximately 50 nanoCuries (nCi) of Am-243, measured by gamma spectroscopy. For the concrete coupons, the %R was determined to be  $88 \pm 5\%$  for RRII and  $67 \pm 9\%$  for ASG. For the granite coupons, the %R for Am-243 was determined to be  $51 \pm 3\%$  for RRII and  $34 \pm 2\%$  for ASG.

**Deployment and Operational Factors:** Use of RRII included a two-step spray application to each surface material coupon and rinse and removal that involved two 30 minute waiting periods. ASG was a one step application that included vacuum removal after a 90-minute waiting period. Both decontamination technologies seem well suited for rough or jagged surfaces as the spray and gel can reach most areas easily. However, the vacuum removal step could become difficult on rough surfaces. Neither of the surface finishes of the concrete or the granite coupons were visibly affected by either of the decontamination technologies.

## **1.0 Introduction**

The U.S. Environmental Protection Agency's (EPA's) Homeland Security Research Program (HSRP) is helping to protect human health and the environment from adverse effects resulting from intentional and unintentional environmental contamination by chemical, biological, radiological or nuclear (CBRN) materials. With an emphasis on decontamination and consequence management, water infrastructure protection, and threat and consequence assessment, HSRP is working to develop tools and information that will help detect the intentional introduction of CBRN contaminants in indoor and outdoor environments and water systems, the containment of these contaminants, the decontamination of buildings and/or water systems, and the disposal of material resulting from cleanups.

The HSRP, through its Technology Testing and Evaluation Program (TTEP), works in partnership with recognized testing organizations; with stakeholder groups consisting of buyers, vendor organizations, and permitters; and with the participation of individual technology developers in carrying out performance tests on homeland security technologies. The program evaluates the performance of innovative homeland security technologies by developing evaluation plans that are responsive to the needs of stakeholders, conducting tests, collecting and analyzing data, and preparing peer-reviewed reports. All evaluations are conducted in accordance with rigorous quality assurance (QA) protocols to ensure that data of known and high quality are generated and that results are defensible. TTEP provides high-quality information that is useful to decision makers in purchasing or applying the evaluated technologies. TTEP provides potential users with unbiased third-party information that can supplement vendor-provided information. Stakeholder involvement ensures that user needs and perspectives are incorporated into the evaluated.

Through TTEP, the HSRP evaluated the decontamination efficacy of two separate technologies: 1) Environmental Alternatives, Inc.'s (EAI) Rad-Release II (RRII); and 2) Argonne National Laboratory's SuperGel (ASG) for decontamination of radioactive americium (Am)-243 from unpainted concrete and granite. This evaluation was conducted according to a quality assurance project plan (QAPP) entitled, "Evaluation of Chemical Technologies for Decontamination of Cobalt, Strontium, and Americium from Porous Surfaces", Version 1.0 dated May 8, 2012, that was developed according to the requirements of the TTEP Quality Management Plan (QMP) Version 3, January 2008. The following performance characteristics of RRII and ASG were evaluated:

• Decontamination efficacy defined as the extent of radionuclide removal following

application of the two decontamination technologies to concrete and granite coupons to which Am-243 had been applied. Another quantitative parameter evaluated was the extent of cross contamination onto uncontaminated surfaces due to the decontamination procedure.

• Deployment and operational data including rate of surface area decontamination, applicability to irregular surfaces, skilled labor requirement, utilities requirements, extent of portability, shelf life of media, secondary waste management including the estimated amount and characteristics of the spent media, and the cost of using the technologies.

This technology evaluation took place during October 2012 at the U.S. Department of Energy's Idaho National Laboratory (INL).

## 2.0 Technology Description

This report provides results for the evaluation of RRII and ASG. The following is a description of each technology, based on information provided by the vendor. The information provided below was not verified during this evaluation.

#### 2.1 Environmental Alternatives, Inc. Rad-Release II

The RRII decontamination technology is a chemical process that involves the sequential topical application of two solutions (applied in the order directed by EAI). RRII extracts radionuclides, including transuranics, from the substrates. This process was developed to be used in sequence to synergistically remove the contaminants via the migration pathways and pores of the contaminated material.

To maximize the efficacy of the extraction process, the chemistry and application are tailored to the specific substrate, targeted contaminant(s), and surface interferences. RRII Formula 1 contains salts to promote ion exchange and surfactants to remove dirt, oil, grease, and other surface interferences. Broad-target and target-specific chelating agents are blended into the solution to sequester and encapsulate the contaminants, keeping them in suspension until they are removed by the subsequent rinse. RRII Formula 2 is designed as a caustic solution containing salts to promote ion exchange, ionic and nonionic surfactants, and additional sequestering agents, also utilized to encapsulate the contaminants and keep them in suspension until they are removed by the subsequent rinse.

RRII can be applied as either an atomized spray or foam (active ingredients do not change). According to the manufacturer, foam deployment of the solution is most appropriate for large scale applications while the spray application (as used during this evaluation) is well suited to smaller applications and applications where waste minimization is a critical factor. Several options are available to facilitate the removal step including vacuuming (as used in this evaluation), simple wiping with absorbent laboratory wipes or rags for small surfaces, use of a clay overlay technique to wick out RRII and contamination over time followed by removal of the clay at a later date, or use of an absorbent polymer that is sprayed over the chemically treated surface to leach or wick out the contaminant-laden solutions and bind them. The sequence of application, dwell, rinse, and removal of the decontamination solution constitutes a single iteration. This procedure may be repeated, as needed, until the desired contaminant removal levels are achieved. More information is available at <u>www.eai-inc.com</u> [accessed 4/1/2013].

## 2.2 Argonne SuperGel

ASG is a system of super-absorbing polymers containing solid sequestering agents dissolved in a nonhazardous ionic wash solution. The resulting hydrogel is applied to a contaminated surface and provides exchangeable ions to the substrate to promote the desorption of radionuclides. The solid sequestering agent provides strong sorption of the target radionuclides within the gel. After removing the radionuclide-laden hydrogel by conventional wet vacuum, the contaminated hydrogel can be dehydrated or incinerated to minimize waste volume without loss of volatilized contaminants. To summarize, ASG provides for:

- *In situ* dissolution of bound contaminants without dissolving or corroding contaminated structural components
- Controlled extraction of water and dissolved radionuclides from the surface and pore/microcrack structures into a super-absorbing hydrogel
- Rapid stabilization of the solubilized radionuclides with high-affinity and high-specificity sequestering agents immobilized in the hydrogel layer, and
- Low toxicity reagents and low volume radioactive waste.

The superabsorbing polymers consist of an anionic mixture of polyacrylamide and polyacrylate in both linear and cross-linked form. The solid sequestering agents are mixed into the dry polymer (10% by mass). The ionic wash solution is composed of a single component salt at 1 mole/liter (L) concentration (no strong acid or base is used). The reconstituted hydrogel (19-20 gram ionic wash solution per gram of dry polymer mix) can be applied by hand for small areas or sprayed on for larger applications. The hydrogel is allowed to react with the contaminated surface for at least 60-90 minutes to maximize the ionic exchange of radionuclides and diffusion/absorption into the hydrogel. The hydrogel is designed to adhere to vertical surfaces without slipping and maintain hydration in direct sunlight for more than an hour. Because no component of the hydrogel is hazardous, there are no special precautions required for ASG disposal until it is used to decontaminate surfaces contaminated with radionuclides (contaminated ASG may need to be disposed of as low level radioactive waste).

Conventional wet-vacuum technology is sufficient to remove the hydrogel from the contaminated surface. For small-scale applications, the head of a standard wet vacuum is adequate, while for larger scale smooth-surface applications, a squeegee attachment is recommended.

## **3.0 Experimental Details**

#### 3.1 Experimental Preparation

#### 3.1.1 Test Coupons

Concrete coupons were prepared in a single batch of concrete made from Type II Portland cement. The company (Burns Brothers Redi-Mix, Idaho Falls, ID) from which the concrete for this evaluation was obtained provided the data shown in Table 3-1 describing the cement clinker used in the concrete mix. The ASTM C150<sup>1</sup> requirement for Type II Portland cement specifies that tricalcium aluminate content be less than 8% of the overall cement clinker. As shown in Table 3-1 the cement clinker used for the concrete coupons was 4.5% tricalcium aluminate. Because the only difference between Type I and II Portland cements is the maximum allowable tricalcium aluminate content, and the maximum for Type I is 15%, the cement used during this evaluation meets the specifications for both Type I and II Portland cements.

<b>Cement Constituent</b>	Percent of Mixture
Tricalcium Silicate	57.6
Dicalcium Silicate	21.1
Tricalcium Aluminate	4.5
Tetracalcium Aluminoferrite	8.7
Minor Constituents	8.1

 Table 3-1.
 Concrete Characterization

To make the concrete coupons, the wet concrete was poured into 0.9 meter (m) square plywood forms (approximately 4 centimeters [cm] deep) with the surface exposed. The surface was then "floated" to allow the smaller aggregate and cement paste to float to the top (the surface used for this evaluation), and then cured for 21 days. Following curing, the 4 cm-thick squares were cut to the desired concrete coupon size of approximately 15 cm  $\times$  15 cm. The coupons had a surface finish that was consistent across all the coupons. This concrete was judged to be representative of exterior concrete commonly found in urban environments in the United States as shown by INL under a previous U.S. Department of Defense, Defense Advanced Research Projects Agency (DARPA) and U.S. Department of Homeland Security (DHS) project<sup>2</sup>.

The granite coupons were provided by INL and were approximately  $16 \text{ cm} \times 16 \text{ cm}$  and 4 cm thick. These coupons consisted of a Milford Pink Granite (Fletcher Granite Co., Westford, Massachusetts) that is pinkish gray with areas of black and white. The surface finish of the granite coupons was that of a split-face granite, a rugged, uneven finish produced by splitting

granite with shims, wedges, or hydraulics. This type of granite has been used in the U.S. National Archives Building, the Smithsonian, and the U.S. Department of the Interior Building in Washington, DC. Figure 3-1 shows the surface texture of both the concrete and granite coupons.

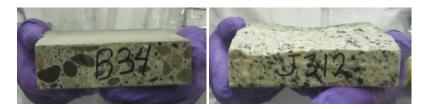


Figure 3-1. Surface finish of concrete and granite coupons.

#### 3.1.2 Coupon Contamination

Am-241 is one of the radionuclides of concern as potentially attractive for use in an RDD. The experimental methods traditionally used by NHSRC for decontamination efficacy evaluations involve measuring contamination using the gamma signature of the radionuclide of concern. However, Am-241 does not exhibit a gamma signature of sufficient strength to allow accurate measurement at the levels representative of those expected for an urban radiological dispersion device (RDD) scenario. However, Am-243, which does have a significant gamma signature, is chemically similar to Am-241, and so was selected as the isotope for this experiment. This allowed for measurement by gamma spectroscopy resulting in a more accurate measurement of the level of contamination than would use of a chemical method, such as inductively coupled plasma-mass spectrometry (ICP-MS), which would have required a contaminant concentration significantly higher than would be realistic. In addition, the high contaminant concentration required by the use of Am-241 would have resulted in a health and safety concern that would have been prohibitive. Table 3-2 describes the number of coupons used in this evaluation. All of the coupons were contaminated with 2.5 milliliters (mL) of unbuffered, slightly acidic aqueous solution containing approximately 20 nanoCurie (nCi)/mL Am-243 which corresponds to an activity level of approximately 50 nCi per coupon ( $\pm$  5 nCi). In the case of an actual RDD, event dry contaminated particles would be expected to settle over a wide area of a city. Application of the contaminant in an aqueous solution was justified because from an experimental standpoint, the ability to apply liquids homogeneously across the surface of the coupons greatly exceeds that capability for dry particles. The aqueous contamination was delivered to each coupon using an aerosolization technique developed by INL under the DARPA/DHS project<sup>2</sup>. Coupons were contaminated approximately two weeks before use.

	Coupons						
Surface Material	Cross- Decon by Decon by contamination Laborator RRII ASG Blanks Blanks						
Concrete	4	4	2	2			
Granite	4	4	2	2			

Table 3-2. Number of Coupons Included in Technology Evaluation

The aerosol delivery device was constructed of two syringes. The plunger and needle were removed from the first syringe and discarded. A compressed air line was then attached to the rear of this syringe. The second syringe containing the contaminant solution was equipped with a 27-gauge needle, which penetrated through the plastic housing near the tip of the first syringe. Compressed air flowing at a rate of approximately 1-2 liters per minute (Lpm) created a turbulent flow through the first syringe. When the contaminant solution in the second syringe was introduced, the contaminant solution became nebulized by the turbulent air flow. A fine aerosol was ejected from the tip of the first syringe, creating a controlled and uniform spray of fine liquid droplets onto the coupon surface. The contaminant spray was applied all the way to the edges of the coupon, which were masked with tape (after having previously been sealed with polyester resin) to ensure that the contaminant was applied only to the working surfaces of the coupons. The photographs in Figure 3-2 show this procedure being performed using a nonradioactive, nonhazardous aqueous dye to demonstrate that 2.5 mL of contaminant solution is effectively distributed across the surface of the coupon.



Figure 3-2. Demonstration of contaminant application technique.

#### 3.1.3 Measurement of Activity on Coupon Surface

Gamma radiation from the surface of each contaminated coupon was measured to quantify contamination levels for 60 minutes both before and after application of the two decontamination technologies using an intrinsic high purity germanium detector (Canberra LEGe Model GL 2825R/S, Meriden, CT). After each coupon was placed in front of the detector face, gamma ray spectra were collected until the average activity level of Am-243 from the surface stabilized to a relative standard deviation (RSD) of less than 2%. The gamma emission energy of 74.66 keV (characteristic of Am-243) was used to identify and measure the activity level of Am-243. To protect against possible interfering radionuclides (not observed during this work), the product of an Am-243 alpha decay (Np-239 with a gamma emission energy of 277.6 keV) was also monitored to confirm the presence of Am-243. Gamma-ray spectra acquired from contaminated

coupons were analyzed using INL's Radiological Measurement Laboratory (RML) data acquisition and spectral analysis programs. Radionuclide activities on each of the coupons were calculated based on efficiency, emission probability, and half-life values. Decay corrections were made based on the date and the duration of the counting period. Full RML gamma counting QA/quality control (QC), as described in the QAPP, was employed and certified results were provided. The minimum detectable level of Am-243 ranged from 0.2 to 0.4 nCi.

#### 3.1.4 Surface Construction Using Test Stand

Because Am-243 is an alpha emitter, there are additional health and safety concerns (compared with use of beta emitters like cesium and cobalt) that were taken into account by the radiological

control technicians (RCTs) and the INL and Portage staff to minimize the possibility of personnel contamination. One control measure was use of a small test stand (Figure 3-3) inside of a radiological hood to hold the coupons. Ten coupons (four concrete, four granite, one concrete blank, and one granite blank) were decontaminated together. The five concrete coupons were placed on the left, with the uncontaminated blank in the lower left position (Figure 3-3). The five granite coupons were placed on the right, with the uncontaminated blank in the lower right position. The blank coupons were included to observe the extent of cross contamination caused by the decontamination activities performed on adjacent coupons.



Figure 3-3. Small test stand.

#### 3.2 Decontamination Technology Procedures

#### 3.2.1 EAI RRII

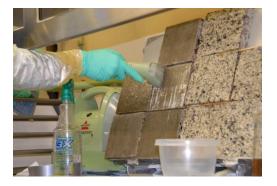


Figure 3-4. Rinsing and vacuuming RRII from concrete coupon

The application of RRII onto the 10 coupons was performed using plastic spray bottles (32 oz. Heavy Duty Spray Bottle, Rubbermaid Professional, Atlanta, GA) as directed by EAI staff via email directions. The coupons were thoroughly wetted with RRII Formula 1 with 3 - 4 sprays. The solution was then worked into the surface of the coupon by scrubbing the entire surface of the coupon once with a scouring pad (Heavy Duty Scouring Pad, 3M Scotch-Brite, St. Paul, MN). During this evaluation, the initial application of RRII Formula 1 took only 10-15 seconds for each coupon. The next step was a 30-minute dwell time for RRII Formula 1 to reside on the surfaces of the coupons. The coupon surfaces were kept damp with 1-2 sprays

of additional RRII Formula 1 approximately every ten minutes. The additional 1-2 sprays of RRII Formula 1 were performed to simulate foam collapse, i.e., the reintroduction of fresh

solutions to the contaminated matrix, as would be observed if RRII were to be deployed as a foam for larger scale applications. After the 30-minute dwell time, the coupon surfaces were thoroughly wetted with a 10% nitric acid rinse solution in deionized (DI) water using another spray bottle. The surface was then vacuumed (vacuum unit "Little Green", Bissell, Grand Rapids, MI) which took about 30 seconds per coupon. The above procedure was repeated using RRII Formula 2. Altogether, the RRII procedure took 79 minutes to complete for ten coupons. Figure 3-4 shows the rinse and vacuuming step of the RRII procedure.

## 3.2.2 ASG

The ASG was prepared by mixing two dry powders with DI water as directed by Argonne staff members via emailed written instruction and phone conversations. The mixture was then stirred with a drill equipped with a mixing tool until the mixture was homogeneous. The manufacturer's instructions called for application with either a plastic spatula/spackling knife, or paintbrush. For this application the ASG was applied approximately six millimeters (mm) thick to the ten coupons using a four inch paintbrush. The specifications of the paint brush were not critical as a perfectly smooth application was not required. A total of two one-liter containers of ASG were applied to the surface of the ten coupons. Altogether, the application and removal of the ASG to the ten coupons required approximately 103 minutes, which included one minute per coupon to apply the gel, a residence time on the surface for 90 minutes, and removal with a wet-vacuum (Little Green, Bissell, Grand Rapids, MI) which required approximately 20 seconds per coupon. Figure 3-5 shows the application and vacuum removal steps for ASG.

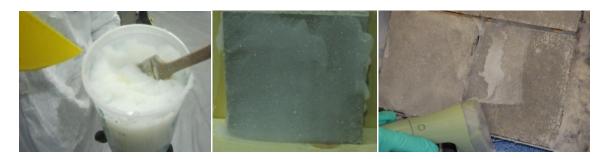


Figure 3-5. ASG before application, as applied to coupon, and during vacuum removal.

## 3.3 Decontamination Conditions

The decontamination technology testing was performed over the course of two days. Table 3-3 presents the number of days between coupon contamination and decontamination, the temperature in degrees Celsius (°C) and the percent relative humidity measured during the evaluation.

Technology	Contaminant	Time Between Coupon Contamination and Decontamination	Temperature During Decontamination (°C)	Relative Humidity During Decontamination (%)
RRII	Am-243	14 days	21.1	16-20
ASG	Am-243	15 days	18.9	16

 Table 3-3. Decontamination Conditions

## 4.0 Quality Assurance/Quality Control

QA/QC procedures were performed in accordance with the QMP and the QAPP for this evaluation.

#### 4.1 Intrinsic Germanium Detector

The germanium detector was calibrated weekly during the evaluation. The calibration was performed in accordance with standardized procedures from the American National Standards Institute (ANSI) and the Institute of Electrical and Electronics Engineers (IEEE).<sup>3</sup> Detector energy was calibrated using thorium (Th)-228 daughter gamma rays at 238.6, 583.2, 860.6, 1620.7, and 2614.5 kilo electron volts (keV). The Am-243 measurements were made using 74.66 keV to identify and quantify Am-243 with confirmation by the 277.6 keV line of the daughter Np-239. Table 4-1 presents the calibration results across the duration of the project, consisting of the difference between the known energy levels and those measured following calibration (rolling average across the six most recent calibrations). These energies were compared to the previous 30 calibrations to confirm that the results were within three standard deviations of the previous calibration results. All the calibrations fell within this requirement.

		Calibration Energy Levels in keV				
Measurement Month	Date Range	Energy 1 238.632	Energy 2 583.191	Energy 3 860.564	Energy 4 1620.735	Energy 5 2614.511
October 2012	10-2-2012 to 11-13-2012	-0.004	0.012	-0.028	-0.222	0.021

Table 4-1. Calibration Results – Difference (keV) from Th-228 Calibration Energies

Gamma ray counting was performed for each coupon, both for initial and final activity levels, until the activity level of Am-243 on the surface had a RSD of less than 2%. This RSD was achieved during the first hour of counting for all the coupons measured during this evaluation. The final activity assigned to each coupon was a compilation of information obtained from all components of the electronic assemblage that comprise the gamma counter, including the raw data and the spectral analysis described in Section 3.1.3. Final spectra and all data that comprise the spectra were sent to a data analyst who independently confirmed the "activity" number arrived at by the spectroscopist. When both the spectroscopist and the data analyst independently arrived at the same value the data were considered certified. This process defined the full gamma counting QA process for certified results.

The background activity of laboratory blank coupons was determined at the start of the experiment by analyzing four arbitrarily selected coupons from the stock of concrete and granite

coupons used for this evaluation. The ambient activity level of these coupons was measured for one hour. No activity was detected above 0.3 nCi for Am-234 on these coupons.

Throughout the evaluation, a second measurement was taken on two coupons to provide duplicate measurements to evaluate the repeatability of the instrument. One of the duplicate measurements was performed after contamination but prior to application of the decontamination technologies, and one was performed after decontamination. Both of the duplicate pairs showed percent difference in activity level of 3% or less, below the acceptable percent difference of 5%.

#### 4.2 Audits

#### 4.2.1 Performance Evaluation Audit

RML performs monthly checks of the accuracy of the Th-228 daughter calibration standards by measuring the activity of a National Institute of Standards and Technology (NIST)-traceable europium (Eu)-152 standard (in units of becquerels, Bq) and comparing the results to the accepted NIST value. Results within 7% of the NIST value are considered to be within acceptable limits. The Eu-152 activity comparison is a routine QC activity performed by INL, but for the purposes of this evaluation, served as the performance evaluation (PE) audit, an audit that confirms the accuracy of the calibration standards used for the instrumentation critical to the results of an evaluation. Table 4-2 gives the results of each of these audits of the detector that was used during this evaluation. All results were within the acceptable difference of 7%.

Date	Eu-152 (keV)	NIST Activity (Bq)	INL RML Result (Bq)	Difference
	Average	124,600	121,600	0.49%
November	122	124,600	118,800	1.45%
2012	779	124,600	120,700	1.77%
-	1408	124,600	121,500	1.35%

 Table 4-2.
 NIST-Traceable Eu-152 Activity Standard Check

#### 4.2.2 Data Quality Audit

At least 10% of the data acquired during the evaluation were audited. The QA Manager traced the data from the initial acquisition, through reduction and statistical analysis, to final reporting, to ensure the integrity of the reported results. All calculations performed on the data undergoing the audit were checked. No significant findings were noted.

#### 4.3 QA/QC Reporting

Each assessment and audit was documented in accordance with the QAPP and the QMP.

## 5.0 Evaluation Results and Performance Summary

#### 5.1 Decontamination Efficacy

The decontamination efficacy was determined for each contaminated coupon in terms of percent removal (%R) and decontamination factor (DF) as defined by the following equations:

 $%R = (1 - A_f / A_o) \times 100\%$  and  $DF = A_o / A_f$ 

where  $A_o$  is the radiological activity from the surface of the coupon before application of the decontamination technologies and  $A_f$  is radiological activity from the surface of the coupon after removal. While the DFs are reported in the following data tables, the narrative describing the results will focus on %R.

#### 5.1.1 RRII Results

Table 5-1 presents the decontamination efficacy, expressed as both %R and DF for RRII when decontaminating Am-243 from concrete and granite coupons. The target activity for each of the contaminated coupons (pre-decontamination) was between 43 nCi and 56 nCi. The overall (both RRII and ASG included) average activity (plus or minus one standard deviation) of the Am-243-contaminated coupons was  $50 \pm 5.5$  nCi, a variability of 11%. The decontamination efficacies of RRII in terms of %R were  $88 \pm 5\%$  for the concrete surfaces and  $51 \pm 3\%$  for the granite surfaces. A paired t-test was performed to determine the likelihood that results for each surface were the same. The %R of Am-243 by RRII from concrete was significantly different (higher) from the %R from granite at the 95% confidence interval (p=0.0022).

As described above in Section 3.1.4, cross contamination blanks were included in the test stand during testing with both contaminants to evaluate the potential for cross contamination due to application of RRII on wall locations above the blank.

Both concrete and granite coupons were used as cross contamination blanks. These coupons were not contaminated, and the pre-decontamination activity measurements indicated extremely low background levels (below the detection limit) of activity. The cross contamination blank coupons were decontaminated using RRII along with the other contaminated coupons. The post-decontamination measurements of activity on these blanks were 1.3 nCi for concrete, and 0.55 nCi for granite. The cross contamination was therefore minimal (1-2% of pre- decontamination activities) but still detectable, and enough to note that the possibility exists of cross contamination to locations previously not contaminated when using RRII in a wide area application.

Surface Material		Pre-Decontamination Activity (nCi/Coupon)	Post-Decontamination Activity (nCi/Coupon)	%R	DF
		56	9.2	84%	6.1
		53	3.2	94%	17
Comonata		55	9.0	84%	6.1
Concrete		54	5.9	89%	9.2
	Avg	55	6.8	88%	9.5
	RSD	1	2.9	5%	4.9
		46	21	54%	2.2
		43	21	51%	2.1
Cuanita		47	22	53%	2.1
Granite		43	23	46%	1.9
	Avg	45	22	51%	2.1
	RSD	2	1	3%	0.1

#### Table 5-1. RRII Am-243 Decontamination Efficacy Results

#### 5.1.2 ASG Results

Table 5-2 presents the decontamination efficacy expressed as both %R and DF for ASG when decontaminating Am-243 from concrete and granite coupons. The target activity for each of the contaminated coupons (pre-decontamination) was between 42 nCi and 56 nCi. The overall (both RRII and ASG included) average activity (plus or minus one standard deviation) of the Am-243-contaminated coupons was  $50 \pm 5.5$  nCi, a variability of 11%. The decontamination efficacies of ASG in terms of %R were  $67 \pm 9\%$  for the concrete surfaces and  $34 \pm 2\%$  for the granite surfaces. A paired t-test was performed to determine the likelihood that results for each surface were the same. The %R of Am-243 by ASG from concrete was significantly different (higher) from the %R from granite at the 95% confidence interval (p=0.0025).

Surface Material		Pre-Decontamination Activity (nCi/Coupon)	Post-Decontamination Activity (nCi/Coupon)	%R	DF
material		<u>52</u>	10	80%	5.0
		56	21	62%	2.6
C (		51	18	65%	2.8
Concrete		42	16	62%	2.6
	Avg	50	17	67%	3.3
	RSD	6	5	9%	1.2
		47	30	36%	1.6
		53	36	32%	1.5
Granite		42	28	33%	1.5
Granite	_	48	32	33%	1.5
	Avg	48	32	34%	1.5
	RSD	5	3	2%	0.04

 Table 5-2. ASG Am-243 Decontamination Efficacy Results

As with the RRII testing, the cross contamination blanks were included in the test stand during testing with both contaminants to evaluate the potential for cross contamination due to application of ASG on wall locations above the blank. Both concrete and granite coupons were used as cross contamination blanks. These coupons had not been contaminated and the pre-decontamination activity measurements indicated extremely low background levels (below the detection limit) of activity. These coupons were decontaminated using ASG along with the contaminated coupons. The post-decontamination measurements of activity on these blanks were below the detection limit for the concrete coupon and 0.36 nCi (detection level for that coupon was 0.2 nCi) for the granite coupon. The cross contamination was therefore minimal (less than 1% of the pre-decontamination activity) during application of ASG.

#### 5.2 Deployment and Operational Factors

Throughout the evaluation, technicians were required to use personal protective equipment (PPE) such as shoulder length gloves because the work was performed in a radiological hood using Am-243. Whenever radiological material is handled, appropriate PPE is required and any waste (e.g., from removal of RRII and ASG from the coupon surfaces) will likely be considered low level radioactive waste and need to be disposed of accordingly. The requirement for this level of PPE was not driven by the use of the decontamination technologies, which do not require use of the extensive PPE, but rather by the presence of Am-243.

#### 5.2.1 RRII

A number of operational factors were documented by the technician who performed the testing with RRII. The application process of RRII was described in Section 3.2.1 and included use of a plastic spray bottle. Application of RRII solutions to each coupon took 10-15 seconds in addition to the recommended dwell time of 30 minutes for each solution. For RRII, the two formulas were applied using the identical procedure which included a 30-minute dwell time for

each. The total elapsed time for the ten coupons decontaminated with RR II was approximately 79 minutes. The application and removal times are applicable only to the experimental scenario using small concrete coupons. According to the manufacturer, if RRII were to be applied to larger surfaces, larger application tools such as larger sprayers or foamers would likely be used which would impact the application rate. In addition, larger vacuum heads would be used for removal. RRII did not cause any visible damage to the surface of the coupons. However, the coupons appeared to have a thin layer of dried residual RRII Formula II remaining on the surface after the final rinse, vacuum removal, and overnight drying. RRII was collected entirely by the wet vacuum and the content of the vacuum canister was solidified in super-absorbing polymer for ease of disposal as a dry granular mixture. Table 5-3 provides some additional detail about certain operational factors for RRII as observed during the use of this experimental setup/test stand with relatively small concrete coupons.

#### 5.2.2 ASG

A number of operational factors were documented by the technician who performed the testing with ASG. Once fully mixed, ASG had the look and consistency of cooked oatmeal but was very "slippery" and tended to slide off a plastic spatula. Therefore the paintbrush was used to apply the ASG (approximately 6 mm thick) to the coupons. However, once on the concrete, ASG adhered rather well. Altogether, the application of ASG took approximately 30 seconds per coupon and removal with a wet vacuum took approximately 50 seconds per coupon. The total elapsed time for the 10 coupons decontaminated with ASG was approximately 100 minutes. If ASG were to be applied to larger surfaces, larger application tools such as large sprayers would likely be used which would impact the application rate. In addition, larger vacuum heads would likely be used for removal. ASG caused no visible damage to the surface of the coupons. Table 5-4 provides some additional detail about certain operational factors for ASG as observed during the use of this experimental setup/test stand with relatively small concrete and granite coupons.

# Table 5-3. Operational Factors for RRII

Parameter	Description/Information
Decontamination rate	Technology Preparation: RRII is provided ready to use. The solutions (Formula 1 and Formula 2) were transferred into spray bottles and applied.
	Application: Using this experimental setup, the initial application of RRII Formula 1 to the coupons took only seconds and then the coupons were kept damp (to simulate the ongoing presence of a foam as would be used during a large-scale application) with reapplication every 10 minutes during the dwell time. Following the 30 minute dwell time, rinsing and vacuuming took approximately 20 seconds per coupon. This
	process was repeated for RRII Formula 2. In all, the application and removal steps took 19 minutes in addition to the two 30 minutes dwell times for RRII. Aside from the dwell times, this process corresponds to a decontamination rate of approximately 0.7 square meters $(m^2)$ /hour (h) for RRII. Estimated volumes used per application of ten coupons (0.2 m <sup>2</sup> ) included 210 milliliters (mL) RRII Formula 1, 210 mL RRII Formula 2, and 180 mL of the rinse solution.
Applicability to irregular surfaces	Application to irregular surfaces would not seem to be problematic, RRII is easily sprayed into hard-to-reach locations. Irregular surfaces may pose a problem for vacuum removal.
Skilled labor	Adequate training would likely include a few minutes of orientation so the technician
requirement	is familiar with the application technique including dwell times and the requirement to keep the surface wet. Larger surfaces may require more complex equipment such as spray or foam application.
Utilities	Electricity for the wet vacuum. Larger surfaces may require more complex equipment
requirement	such as spray or foam application requiring additional utilities.
Extent of portability	At a scale similar to that used for this evaluation, vacuum removal would be the only portability factor. However, for larger scale applications, limiting factors would include the ability to apply RRII at a scale applicable to an urban contamination (area of city blocks or square miles) and then rinse and remove with a vacuum. Portable electrical generation or vacuum capability may be required.
Secondary waste	Approximately 600 mL of liquid was applied per 10 coupons used during this
management	evaluation which corresponds to a waste generation rate of approximately $3 \text{ L/m}^2$ . This generation rate would likely vary for different surface materials depending on how much of the solutions absorb into the surfaces. RRII was collected entirely by the wet vacuum and the content of the vacuum canister was solidified in super-absorbing polymer for ease of disposal as a dry granular mixture.
Surface damage	Concrete and granite surfaces appeared undamaged, however they appeared to have a thin layer of dried residual RRII Formula II remaining on the surface after the final rinse, vacuum removal, and overnight drying.
Cost	RRII solutions are not sold as a stand-alone product but are available only as a decontamination service for which the cost varies greatly from project to project. Typical projects costs are in the approximate range of \$33-\$55/m <sup>2</sup> .

Parameter	Description/Information
Decontamination	Technology Preparation: 15 minutes to measure and mix powder with water.
rate	ASG is able to be used for several days after mixing as long as ASG is kept moist by covering the mixture, as it will dry out if left exposed to air for several days.
	Application: ASG was applied with a paintbrush to each coupon in approximately 30 seconds $(3 \text{ m}^2/\text{h})$ . After a 90-minute dwell time, ASG was removed with a wet vacuum and the surface was wiped with a paper towel at a rate of approximately 50 seconds per coupon $(2 \text{ m}^2/\text{h})$ . Aside from the wait time (which is independent of the surface area), the application and removal rate was approximately 1 m <sup>2</sup> /h. Estimated volumes used per ten coupons included 2 L of ASG. Overall that volume corresponds to a loading of 9 L/m <sup>2</sup> .
Applicability to	Application to irregular surfaces may be problematic as ASG could slide off
irregular surfaces	jagged edges and be hard to apply to hard-to-reach locations. During use on
	the rough split face granite, small amounts of ASG could be seen remaining in
	the crevices after vacuum removal.
Skilled labor	Adequate training would likely include a few minutes of orientation so the
requirement	technician is familiar with the application technique. Larger surfaces may require more complex equipment such as sprayer application.
Utilities	As evaluated here, electricity was required to operate the wet vacuum. Larger
requirement	surfaces may require more complex equipment such as spray application requiring additional utilities.
Extent of portability	At a scale similar to that used for this evaluation, the only limitation on
	portability would be the ability to provide vacuum removal in remote
	locations. However, for larger scale applications, limiting factors would
	include the ability to apply ASG at scale applicable to an urban contamination (area of city blocks or square miles).
Secondary waste	Approximately 4 L of ASG was applied per ten coupons during this
management	evaluation. That volume corresponds to a waste generation rate of
0	approximately 9 $L/m^2$ . ASG was collected entirely by the wet vacuum and the
	content of the vacuum canister was solidified in super-absorbing polymer for
	ease of disposal as a dry granular mixture. The final volume of waste was approximately 4 L.
Surface damage	Concrete and granite surfaces appeared undamaged.
Cost	The material cost is approximately \$0.30/L. This cost corresponds to
	approximately \$2/m <sup>2</sup> if used in a way similar to the process used during this
	evaluation. Labor costs were not calculated.

 Table 5-4. Operational Factors for ASG

#### 6.0 References

- 1. ASTM Standard C 150-07, 2007, "Standard Specification for Portland Cement," ASTM International, West Conshohocken, PA, <u>www.astm.org</u> [accessed 4/1/2013].
- Radionuclide Detection and Decontamination Program, Broad Agency Announcement 03-013, U.S. Department of Defense Advanced Research Projects Agency (DARPA) and the U.S. Department of Homeland Security, classified program.
- 3. Calibration and Use of Germanium Spectrometers for the Measurement of Gamma Emission Rates of Radionuclides, American National Standards Institute. ANSI N42.14-1999. IEEE New York, NY (Rev. 2004).



PRESORTED STANDARD POSTAGE & FEES PAID EPA PERMIT NO. G-35

Office of Research and Development (8101R) Washington, DC 20460

Official Business Penalty for Private Use \$300