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Environmental and Radiological Readiness of Fixatives, Foams, and Intumescent Coatings for D&D Applications – 18553

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ABSTRACT
The development, testing, and characterization of commercial fixatives and foams for their applicability towards fixating contamination, potential structural reinforcement, and fire resiliency in legacy nuclear facility equipment. Much of the equipment, such as gloveboxes, shielded cells, and radiological hoods left over from nuclear processing entails large cavities, and filling these voids after decontamination efforts provides an easy means of shielding workers from excess radiation exposure while immobilizing residual contamination. As such, we look to continue the development of fixatives/intumescent coatings from the previous year and test commercially available polymer-based foam products for use in filling these cavities, providing a secondary level of protection and shielding during the D&D process. During D&D operations of legacy nuclear facilities, the ability to effectively stabilize contamination in large cavities is a significant challenge. For items that must be removed such as gloveboxes and entry hoods, a method of simply filling the volume to stabilize any residual contamination after contamination efforts have concluded would be of great benefit, and further the ALARA culture of the DOE complex by shielding workers in the area prior to the equipment removal. This research is funded as part of the In-Situ Decommissioning and Technology Development and sponsored by DOE- Office of Environmental Management Technology Development.

INTRODUCTION
Nuclear facilities that are moving towards final disposition, face enormous challenges to ensure no holdup material is released to the environment between the time the facility is no longer active through final disposition operations. Few situations are of greater concern during D&D and interim storage activities than the possibility of a fire. As evidenced by the incident at WIPP in February 2014, and others across the DOE and international nuclear complexes, the potential for a release of radioactive contaminants when exposed to fire is ever-present. Workers actively seek to remove as much of the radioactive materials holdup as possible; however, current decontamination methods are not effective in the removal of all contaminated material. As such, for contaminated areas, there is often some residual contamination remaining after material removal and decontamination efforts have concluded. Gloveboxes are a source of hold up contamination that must be addressed, as they are numerous in legacy nuclear facilities. In addition, many of the fixative products in common use during D&D of facilities are highly vulnerable to fire and extreme heat conditions, thereby increasing the risk of a release of the radioactive contaminants, resulting in potential exposure to workers and the public.
The Basis for Interim Operation (BIO) in support of Savannah River Site 235-F Plutonium Fuel Fabrication (PUFF) Facility contains a postulated accident scenario where an earthquake causes a breach of the facility containment structure. This seismic event is also postulated to initiate a large room fire which could propagate and potentially evolve into a full facility fire which engulfs the material at risk (MAR) such as residual Pu-238 and Np-237, causing it to become airborne and released from the building. The responsible site contractor has determined that the unmitigated consequences of this event are greater than 10 rem offsite and 27,000 rem to the collocated worker at 100 meters. This contingency, and others related to fire across the various sites, has prompted a requirement for fire retardant / fire protection technologies that can enhance a facility's overall fire protection posture and mitigate the

release of radioisotopes during these emergencies. In response to the potential hazards present in the SRS 235-F PUFF Facility and other D&D challenges across the DOE complex and around the world, the continued development of fixatives/intumescent coatings and the emergence of fire resistant foams are jointly being advanced by SRNL and FIU.

There has been extensive development of intumescent coatings as a viable technology in fire and explosion protection. Initially developed to protect and insulate various substrates from extreme heat and fire conditions to maintain their structural integrity, research revealed that in certain instances the fire protection was so effective that it protected the primer itself on the substrate. ARC research scientists had firsthand knowledge and experience in the use of intumescent coatings to harden facilities and improve fire protection in support of the U.S. military, and through leveraging a basic layering concept put forth by research scientists at SRNL, explored the feasibility of using the technology to enhance fire resiliency in fixatives used in D&D. Agencies such as the National Aeronautics and Space Administration (NASA) and the Department of Homeland Security (DHS), as well as the oil and gas industries have used this proven, cost effective technology with excellent results in various applications. Capitalizing on technological developments in one area could have direct application to enhancing fire resiliency during D&D activities. Additionally, one method identified by SRNL is the use of two-part epoxy foams capable of filling the interior volume of the glovebox to ensure fixation of any remaining material after decontamination efforts have concluded. SRNL previously characterized five (5) commercially available epoxy foams from the Foam-iT product line (Foam-iT 3, Foam-iT 8, Flex Foam-iT III, Flex Foam-iT 14, and Flex Foam-iT 25) and included the most promising additives for radiological characterization to ensure correlation with the modeled results1-3. For the current work, new foams were investigated including 2-part epoxy based fire retardant foams (FlexFoam-iT 7 FR and FlexFoam-iT 23 FR) and compressed fire-retardant foams (Abesco FireRated FP200 and Great Stuff Fireblock). More foams have since been received by SRNL and are currently under testing.

METHODS

This section outlines the experimental plans for both the fixative/intumescent coatings and the two-part epoxy foams developed and tested by both SRNL and FIU.

Fixative/Intumescent coatings

The test objectives outlined in the final test plan were developed through extensive coordination with SRNL research scientists and SRS 235-F site personnel (i.e.; project managers, safety and fire representatives, etc.), and are designed to advance the testing, evaluation, and possible deployment of intumescent coating (IC) technologies as fire resilient fixatives to mitigate the potential release of radioisotopes during postulated fire scenarios highlighted in the basis for interim operations (BIO) and contingency planning documents in support of D&D activities at SRS 235-F, with a particular emphasis on the 235-F PuFF Facility Cells 6-9. The first main objective of the test plan centered on constructing a to-scale SRS 235-F Hot Cell Test Bed on site at ARC that mirrors the operating environment encountered in an adjoining corner and middle hot cell configuration at the SRS 235-F facility. The second main objective includes an evaluation on the mechanics and processes associated with applying the selected intumescent coatings in the hot cell configurations using: 1) the approved tools as identified in the 235-F Risk Reduction Tooling List, Rev 0, dated 26 January 2015; and 2) alternative application methods, such as airless sprayers, recommended by the IC manufacturer.

<u>Hot Cell Test Bed</u>

In close coordination with SRNL and SRS site personnel, FIU designed and developed a to-scale, combined corner cell and middle cell configuration that mirrors the operating conditions (dimensions, glove ports, surface materials, obstacles/obstructions, etc.) encountered in hot cells 6-9 at the SRS 235-F facility. FIU completed the construction of the hot cell test bed at ARC's Outdoor Technology Testing & Demonstration Facility in May 2017. An ARC Fact Sheet on the hot cell test bed was developed and finalized in June 2017. The dimensions of each cell are 5' wide x 10' long x 7' high and include a 3' raised floor (Figure 1). Pass-through ports and glove ports were sized and positioned in collaboration with SRNL/SRS to closely mimic the actual conditions at the facility. Sheets of 304 stainless steel were installed to represent the surfaces found in the hot cells.

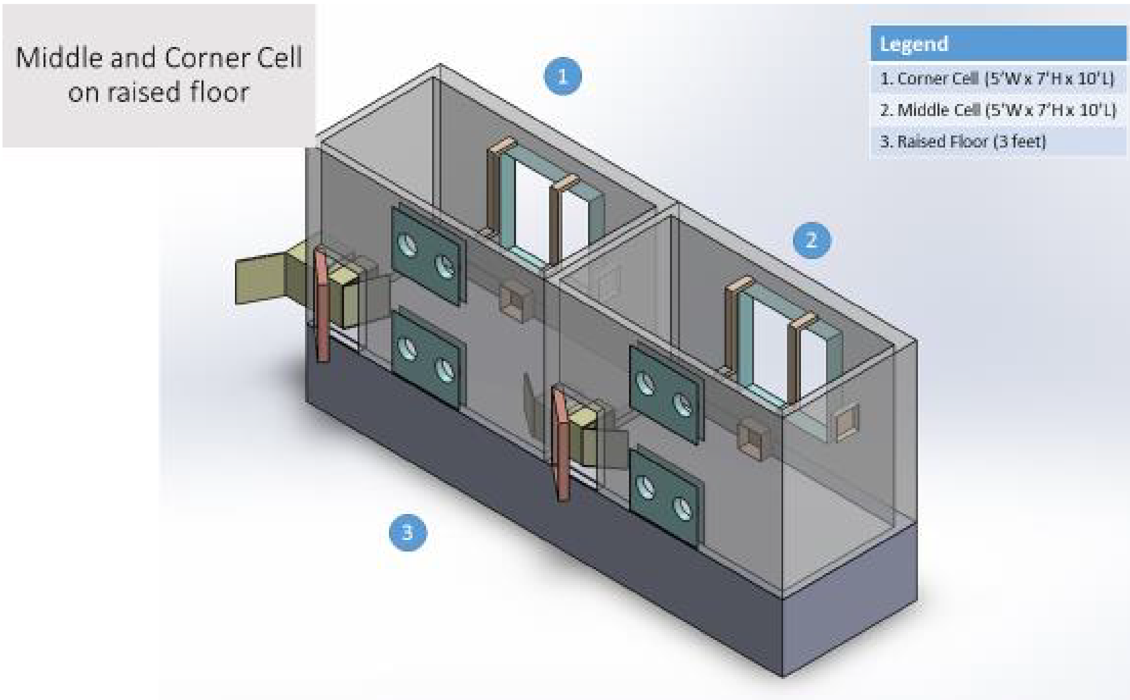

Figure 1. Hot Cell Test Bed

The second primary objective of the test plan began with the application of the intumescent coating using the site approved tools, including an extension pole and a roller brush, cure time monitoring, confirmation of all required tools and materials to fit through the bag in/bag out port and pass-through port, and an evaluation of volume/surface area of intumescent coating required to cover all surfaces to the minimum thickness needed (1/8"). All the required tools and materials easily fit through the bag in/bag out port as well as the pass-through port between hot cells. No significant challenges were encountered in moving materials and supplies associated with the operation from one hot cell to another using the pass-through port. FIU used standard paint trays as well as 2.5-quart plastic containers to hold the intumescent coating. These options were used to mitigate any weight challenges during application. The total volume of product needed to apply a 1/8" thick coating to all of the surfaces would be approximately:

3.9 gal x 2 (floor & ceiling) + 2.8 gal x 2 (side walls) + 5.5 gal x 2 (front/back walls) = 24.4 gal

With the retail cost of the IC being approximately $425 for a 5-gallon container, the approximate cost to coat one hot cell would be $2,125.[1]

[1] These estimates do not include the effect of other hot cell structures such as the bag-in/bag-out and pass-through ports, windows, etc. In addition, the calculations do not consider potential product losses and other variables. The estimated volume of product should therefore include a safety factor multiplier to ensure that enough IC is available.

Application of IC Using Slow Pour Method for Horizontal Surfaces

With 95% of the assayed contamination residing on the floor of the SRS 235-F facility hot cells, and given the composition and characteristics of the intumescent coatings, FIU initially performed a small-scale test of slowly pouring the IC onto a 1' x 1' area within the hot cell testbed to the requisite thickness of 1/8". This method showed significant promise in reducing worker time and potential for disturbing residual contamination during the application of intumescent coating on horizontal / floor surfaces in hot cells. The IC cured within 24-hours of application with a heat index of around 100ºF.

FIU proceeded with a larger-scale test and evaluation of the technique. FIU sectioned off a 5'x5' section of the hot cell to further evaluate the effectiveness of using a simple slow-pour method of applying an intumescent coating to the floor of the hot cell test bed. FIU used the following tools:

1. Container – 2.5-quart-size plastic bucket to hold/transport the IC
2. Gripper - used to maneuver a 2.5-quart-size plastic container of IC
3. Custom wooden T-shaped extension tool - used to spread the IC.

To develop this tool, FIU connected a 13" wooden head to the approved extension handle on the SRS tooling list. FIU poured the IC into a 2.5-quart plastic container outside the hot cell, filling the container with 1 to 1.25 quarts of IC to keep the weight between 2 to 3 pounds to minimize worker fatigue. The container and tools were then passed through the bag in/bag out port. Maneuvering the 2.5-qt container to various locations within the hot cell by manipulating the gripper from the glove-ports was relatively easy and allowed for targeted pouring of the IC. After marking the hot cell border at 1/8" above the floor, FIU poured the IC from the container at a height of about 1 to 3" above the floor to minimize any potential for splatter or disturbance of any residual contamination that may be present in a radioactive environment. FIU then spread and smoothed the IC across the floor area using the wooden T-shaped tool, using the 1/8" markings along the perimeter of the hot cell as a thickness guide. The custom wooden T-shaped extension tool allowed access to all locations within the hot cell test bed from the glove-ports and easily reached the corners. An area of 5' x 5' was coated to 1/8" thickness in approximately 15 minutes once the tools and IC were in the hot cell. Total curing time after the slow pour application was 48 hours under hot and humid weather conditions (temperatures 75°-97° F and humidity 45%-93%). The curing was confirmed by FIU using a basic pressure test based on previous experiments with the material. Using a white paper towel, FIU pressed firmly on the test area, remove the paper towel, and observed the surface for any indentation and/or any discoloration of paper towel. For areas within the hot cell test bed that were beyond arm reach, this test was accomplished using a gripper tool. All horizontal surfaces were cured at 48 hours after the slow pour application method.

The thickness of the intumescent coating was confirmed using a Defelsko PosiTector-6000 FNTS (0-250 mils, Ferrous + Non- Ferrous). This instrument conforms to ISO 2178/2360/2808, ASTM B499/D1186/D1400/D7091/E376/G12, BS3900-C5, SSPC-PA2 and others.

After two applications using the slow pour application method for horizontal surfaces previously described, all areas of the floor coated met the requisite 1/8" thickness requirement for the IC fire rating.

Initial Testing of Handheld Sprayer

While still needing significant additional testing and evaluation, initial proof-of-concept testing with the selected handheld sprayer demonstrated the capacity to spray the IC at various distances. These initial positive results indicate that this type of tool may be an acceptable application method for vertical surfaces and warrants further testing to evaluate this potential. The viscosity of the IC being tested is 120,000 centipoise (CPS). For context, the viscosity of water is 1 CPS and the viscosity of peanut butter is roughly 250,000 CPS.

Epoxy Foam

Sample Preparation

All new samples were fabricated for their expansion profiles in a closed environment. SRNL personnel designed and 3D printed a block with removable sides and an interior volume of 2”x2”x2”. Various liquid volumes of epoxy based foams were tested within this mold as well as sprayed volumes of foams from pressurized canisters. First, the mold was assembled with 5 sides and loaded from the top with the foaming material. The top was then securely placed on the mold and the material allowed to cure for 2 hours. After this time, the mold was disassembled and the material extracted for testing and evaluation.

Following cube evaluation, additive testing was completed using the previously established testing protocols to ensure comparability between sample sets. Preparation of loaded and unloaded foam blanks was performed according to manufacturer direction. All foams were received as two-part epoxies that mixed at a given ratio (either 1:1, 1:2, or 2:1). When loading additives, the larger volume material was placed in the container first and mixed thoroughly with the additive, then the second constituent was added and mixed thoroughly for 10 seconds and allowed to foam and cure. Foaming in all instances for small samples was complete within approximately 5 minutes, though full curing per manufacturer direction persisted for a minimum of 2 hours prior to removal from the mold. The foams chosen all have different mixing profiles and foaming volumes.

Down-Selection of Foam Materials

The properties of interest for the foam materials include density, expansion rate, and additive capacity. High density foams are favorable as they have a higher gamma absorption cross-section, though the other material properties may prove equally as important in determining the most functional material for D&D scenarios. A survey of common foam epoxies was conducted to evaluate their properties (i.e., expansion rate, density). To complete this task, small scale samples were fabricated by mixing the 2-part foam epoxies and canisterized foams as blanks and with additives to evaluate differences in performance for open and closed volume environments. For these materials, each sample was mixed and allowed to cure fully before total volume expansion was calculated. During the curing process, expansion characteristics were monitored.

Radiation Shielding Testing (10 Month Evaluation)

Previously down-selected materials from the initial experiments were re-evaluated for their performance at 10 months using gamma sources of varying energy levels. Testing was conducted using sealed Co-60, Cs-137, and Am-241 sources available at SRNL Building 735-2B. To characterize the radiation shielding efficiency of each foam/additive mixture, a FLIR identiFINDER 2 (Figure 2) was used.

Figure 2: FLIR identiFINDER 2

For the initial testing, the identiFINDER was first used to measure the dose rate of the source on contact. A measurement was then taken of the source at a given standoff greater than the thickness of the foam samples (a static 10 cm) to ensure comparable results are obtained across all samples. An initial dose rate was taken for the undoped foam to allow for a baseline shielding percentage to be obtained for the undoped foams to normalize the effects of the introduced additives. Following this measurement, the foam/additive mixture was placed in front of the indentiFINDER with the source remaining at the standoff (10 cm) with the foam and a dose rate measurement was taken (Figure 3). From this information, percent shielding for each foam and source was calculated.

Figure 3: Radiation measurement setup at 10 cm standoff distance (initial setup)

For the 10-month re-evaluation, the setup shown in Figure 4 was used to better control the distance between the source and the detector. This setup utilized 3D printed parts (green) that mounted the source and detector in a fixed orientation and provided a platform for sample loading on-plane with the detection path. This setup proved to have more reproducibility due to the stationary aspects for source and detector and will be used for all subsequent testing of material.

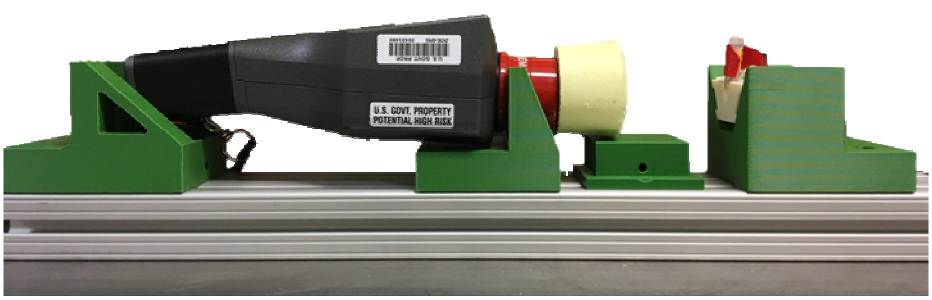

Figure 4: **Radiation measurement setup at 10 cm standoff distance (10-month setup)**

DISCUSSION

Fixative/Intumescent Coatings

Enhancing the fire resiliency of fixative technologies is a completely novel approach used to support D&D activities, and therefore most of the testing to date has been purposefully oriented towards basic proof-of-concept experiments to confirm whether the approach has merit. None of the intumescent coatings on the market today are designed for this specific purpose, and still have significant research and development required to successfully adapt intumescent coatings to D&D activities for fire resiliency and fixative capacity in various nuclear facilities. The preliminary results to date from this particular effort are important in that: 1) they identified a potential vulnerability in fire resiliency (and possibly radiation resistance and environmental durability via testing at SRNL) of the current line of fixatives used in the industry today, and 2) they have been promising in identifying the adaptation of intumescent coatings as a potentially viable approach to enhancing fire resiliency of fixatives and facilities in support of D&D activities.

With 95% of the assayed contamination residing on the floor of the SRS 235-F facility hot cells, and given the composition and characteristics of the intumescent coatings, FIU performed a small-scale test of slowly pouring the IC onto a 1’ x 1’ area within the hot cell testbed. This method showed significant promise in reducing worker time and potential for disturbing residual contamination during the application of intumescent coating on horizontal / floor surfaces in hot cells. The IC cured within 24-hours of application with a heat index of around 100ºF. Consequently, FIU moved forward with a full-scale test and evaluation of the technique. (Figure 5)

Figure 5. Small scale test of a slow pour technique using intumescent coatings (left) and preparing for full-scale demo of slow pour application method for horizontal / floor surfaces in hot cell (right).

The slow pour method for the floor/horizontal surfaces proved very effective during application, significantly reducing time and effort while greatly facilitating application to the requisite thickness level. With an estimated 95% of the contamination residing on the hot cell floors at the SRS 235-F facility and given the composition and characteristics of the IC being tested, the slow pour method may be a viable application method for horizontal surfaces that would expedite application and minimize disturbance of any residual contamination.

Figure 6. Airless sprayer (left) and manual film thickness gauge (right).

Testing of the handheld sprayer showed some initial success as a possible option for applying the IC fixative to vertical/wall surfaces. The sprayer is self-contained, relatively lightweight, battery operated, easily fits through the bag in/bag out and pass-through ports, and appears to be compatible with the viscosity of the FD intumescent coating. FIU conducted a series of comparison tests between roller vs sprayer application. The roller method of application averaged less than 14 mils of thickness per application on a vertical surface and approximately 12 total applications are needed to reach the requisite 1/8" coating thickness. With a 24-48 hour curing period between applications, this option does not appear like a viable for field deployment. On the other hand, the results with the hand-held sprayer were much more promising in terms of labor, time, and number of applications. FIU used a cordless GRAYCO UltraMax Handheld Airless Paint Sprayer at a setting 10 to apply the IC to a 36" x 40" vertical (wall) surface of the hot cell test bed to a 1/8" coating thickness. Approximately 2 quarts of product were consumed. Including time needed to refill the sprayer, the total application time with the sprayer was 5 minutes. (Figure 6)

Epoxy Foams

To ensure conservation of time and additives, an initial down-selection of the foams was performed based on the expansion profiles of the foam during curing in a closed environment. Initial expansion characterization of each foam was taken upon receipt to ensure that a given liquid volume (~60 mL for this test) foamed to the expected final volume. Figure 7 shows the two epoxy based foams following curing within an enclosed environment. Overall, these materials behaved well with no material failure noted for sufficient initial liquid volume necessary to fill the void volume. Density of these foams was found to be controllable based on initial liquid volume. Characterization of density alteration will be performed in future testing using water displacement methods.

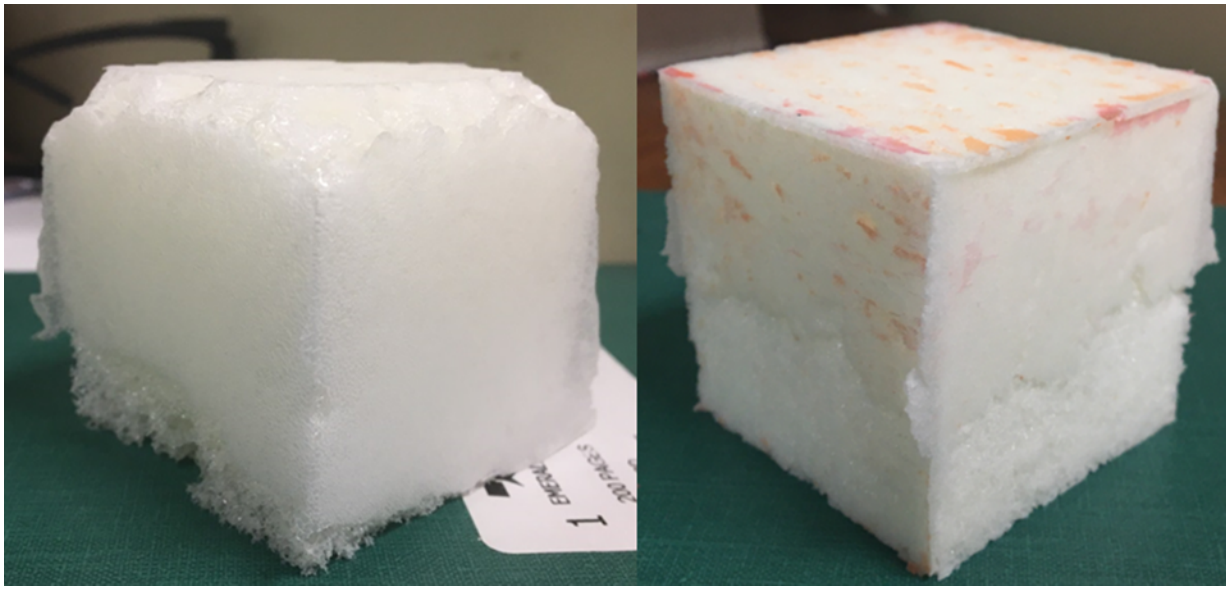

Figure 7: Flex Foam-iT 7 FR (left) and 23 FR (right) [2-part epoxy].

Figure 8 shows the two canisterized foams performance following curing within an enclosed environment. The Abesco sample develops large void spaces that collapse upon curing and disassembly of the mold. Collapsed areas for these materials do not reach full curing, leaving semi-liquid areas within the volume. The Great Stuff material fully cures, however it leaves large void volumes and generates excessive pressure within the confined mold during curing.

Figure 8: Abesco FireRated FP200 (left) and Great Stuff Fireblock (right) [Canisterized].

Radiation shielding capabilities were tested for both F8 and F14 loaded foams using three gamma emitting sources of differing energies and dose rates after aging 10 months. Each proprietary compound was loaded into foams F8 and F14 at a 0.1, 0.3, 0.5, 0.7, and 1.0 ratio of additive to foam weight. Radiation dose rate measurements were conducted in SRNL Building 735-2B using gamma sources of varying energies. Table 1 provides the initial non-shielding dose readings at a 10-cm standoff distance from the initial and ten-month testing, along with the associated energies of each source. A 10 cm standoff distance (distance from the source to the FLIR identiFINDER 2) was selected to ensure that the differing foam thicknesses did not affect the sources distance from the detector.

Table 1: Sources used for radiation shielding testing

Source	Source ID	Energy (MeV)	Initial Dose at 10 cm (mrem/hr)	Dose at 10 cm for ten-month evaluation (mrem/hr)
Am-241	003/11	0.0595	4.94	5.57
Cs-137	F2-785	1.18	15.34	15.19
Co-60	K2-125	2.82	3.78	3.75

Following the radiation measurements, shielding efficiency percentages were calculated. An example of this for both Foam-IT 8 and 14 is shown below in Figures 9 and 10 for Am-241. Shielding percent difference (~2-3%) is attributed to error of the device as well as experimental error in measuring the distance of the initial testing which has been remedied through the new fixed experimental setup.

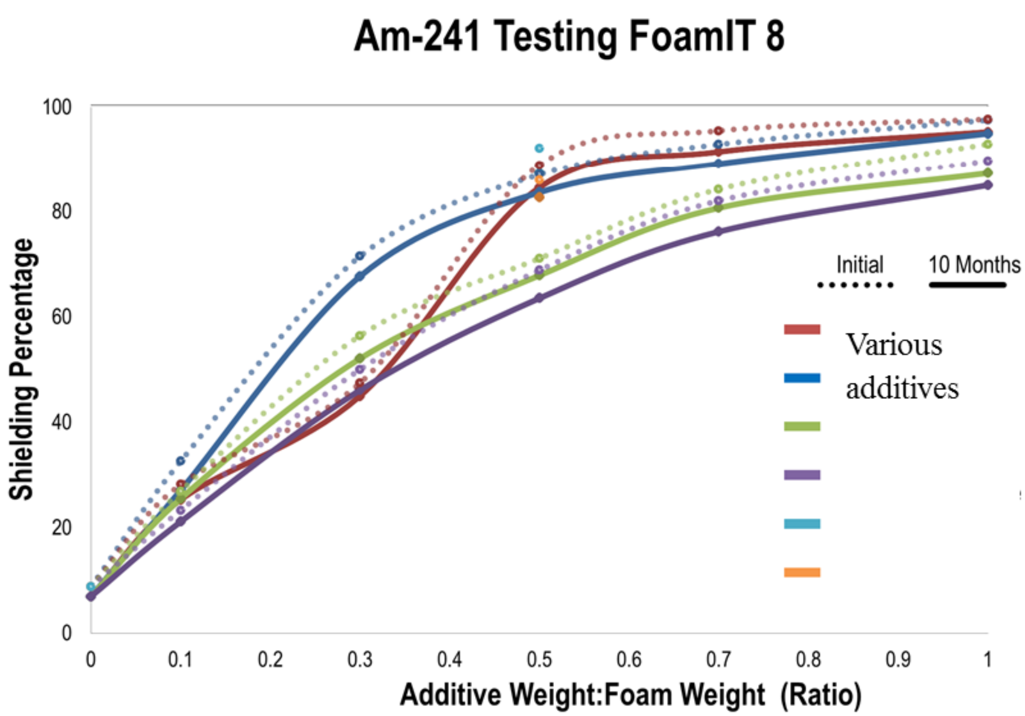

Figure 9: Am-241 Foam-IT 8 Test Data

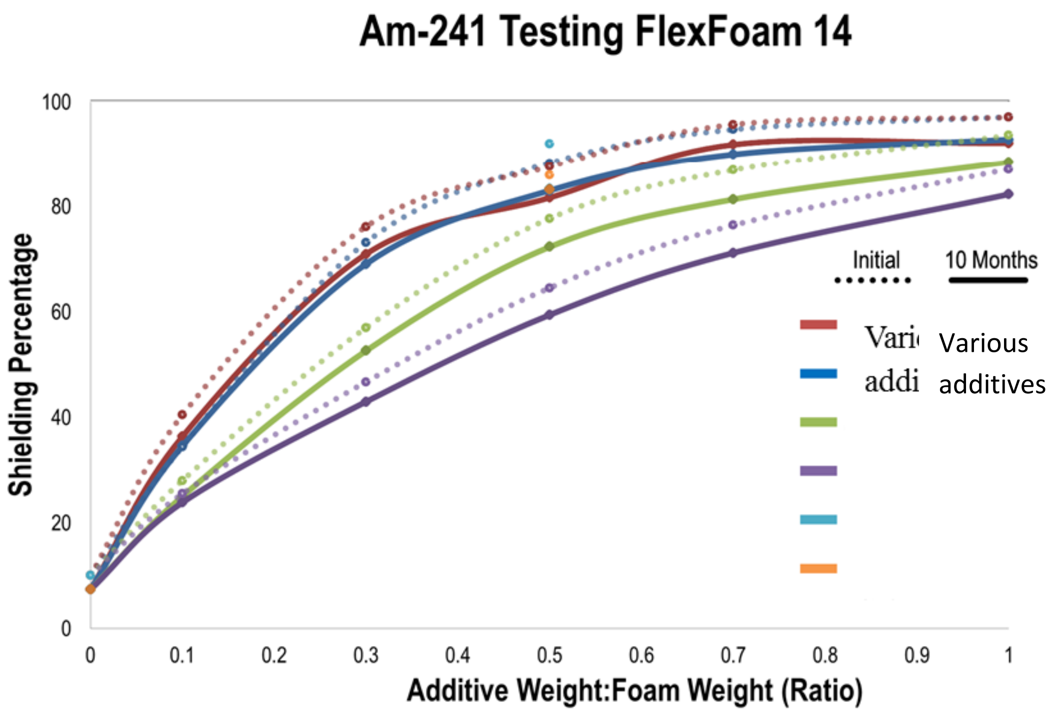

Figure 10: Am-241 Flex Foam 14 Test Data

CONCLUSIONS

Since the January 2016 brief to site personnel, ARC and SRNL have conducted extensive coordination with SRS 235-F personnel to facilitate planning to expedite the R&D related to this effort. An initial set of documents associated with the specific facility hot cells, including schematics, assessment of the manipulator arms, anticipated particulate sizes of the Pu-238 contamination, an approved tooling list, etc., were formally reviewed and approved for release by the site to ARC. Utilizing these as a foundation, pre-planning for further adaptation of the intumescent coatings and the application during a full-scale cold demo at a hot cell testbed at ARC has commenced in anticipation of funding. Based on current work, four objectives have been completed with respect to intumescent coatings: 1) confirmed the specific operational, safety, and regulatory requirements that the intumescent coating fixative will need to satisfy; 2) continued baseline testing of additional commercial-off-the-shelf intumescent coatings with the intent of identifying the one to two that best satisfy those specific requirements; 3) constructed a full-scale mock-up of the SRS 235-F shielding cell to the greatest extent possible at the ARC testbed; and 4) conducted a full-scale cold test demo of the intumescent coating and its application in the testbed at ARC with collaboration from SRNL. FIU could reach all locations within the hot cell configuration using the site approved tools. FIU did not include obstacles in the hot cell configuration that could complicate access and application. The primary challenge encountered during the application of the IC using the site approved tools is that multiple coats of the IC, with related curing time between coats, will be needed to reach the requisite 1/8” coating thickness. Further evaluation of the thickness of each coat (initial and each subsequent coat) and estimated total number of coats needed is currently being performed.

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