

Fixation of Radiological Contamination; International Collaborative Development

Rick L. Demmer

March 2013



The INL is a U.S. Department of Energy National Laboratory
operated by Battelle Energy Alliance

Fixation of Radiological Contamination; International Collaborative Development

Rick L. Demmer

March 2013

**Idaho National Laboratory
Idaho Falls, Idaho 83415**

<http://www.inl.gov>

**Prepared for the
U.S. Department of Energy
Assistant Secretary for Environmental Management
Under DOE Idaho Operations Office
Contract DE-AC07-05ID14517**

March 25, 2013

CCN 229877

Andrew P. Szilagyi
EM-13/Cloverleaf Building
U.S. Department of Energy
1000 Independence Ave., S.W.
Washington, DC 20585-2040

SUBJECT: Fixation of Radiological Contamination; International Collaborative Development

Executive Summary

A cooperative international project was conducted by the Idaho National Laboratory (INL) and the United Kingdom's National Nuclear Laboratory (NNL) to integrate a capture coating with a high performance atomizing process. The initial results were promising, and lead to further trials. The somewhat longer testing and optimization process has resulted in a product that could be demonstrated in the field to reduce airborne radiological dust and contamination.

Introduction

Contamination control is crucial during D&D activities, as well as during modifications to existing systems. In some cases, substantial contamination is present throughout the exhaust ductwork of older facilities due to the years of processing in hoods, glove boxes, etc. A critical concern during the decommissioning of radioactive facilities is to prevent the spread of radiological contamination. This is more serious for areas where large accumulations of radioactive dust and lint are present and have the potential to "go airborne", such as buried drum exhumation, laundry facilities, exhaust ventilation ducting and exhaust stacks.

Around the DOE complex, these airborne contamination issues pose hazards to the workers and result in significant productivity loss. At the Idaho National Laboratory, Radioactive Waste Management Complex, excavation in a highly contaminated pit required that a backhoe operator be placed in a contamination proof, positive pressure cab to prevent inhalation of airborne contamination. This required change of operators every two hours due to time limitations of using "respiratory protective equipment". Together with the accompanying change of anti-contamination clothing, survey requirements and multiple restarts during the day, this amounted to a loss of thousands of dollars per day in lost productivity. In a recent demolition of large ventilation ducting (about 10-ft cross section and 100-ft long) at Brookhaven National Laboratory, a spray coating was applied to the duct from inside using a painter dressed in anti-contamination clothing and an airline respirator system [3]; effectively controlling the hazard, but requiring a manned entry into a dangerous environment. In still other facilities highly contaminated airborne activity areas are closed to entry and no maintenance is performed. These

“infinity rooms” (with airborne contamination levels beyond the scope of current instrumentation to quantify) exist in several locations throughout the DOE Complex; such as pump pits at Savannah River and filter cubicles at Hanford. Yet another case at the Idaho Site was an area of significant accumulations of lint at a radioactive laundry. That duct, shown in Figure 1, is contaminated with lint and radioactive materials attached to the walls that require control during decommissioning.

There have been previous attempts of applying a coating to capture the high levels of airborne contamination with varying degrees of success. Typically these attempts have not been successful in penetrating and causing adherence of contaminated lint and dust to the ductwork. As a result, that contamination was not fixed and ultimately became airborne when it was disturbed. Also, traditional methods of capturing this contamination have been very labor intensive, i.e. placing a worker in the area to spray a coating, and exposing personnel to the hazard. Likewise, spraying a coating in an area with penetrations, auxiliary piping and other equipment usually does not fix contamination that is “shadowed” by such items. Some coatings, such as glycerin, leave a residue is slippery and could cause a slipping hazard. Using copious amounts of water to control dust causes a radiological waste issue. Thus, an improved capture coating and delivery mechanism was identified as a need to resolve these contamination issues.

Figure 1, Lint inside ducting at the INL.



Development of an Advanced Contamination Capture System

During 2011 it was recognized that two separate technologies, the United Kingdom's (UK) National Nuclear Laboratory's (NNL) PDX large volume atomization technology and the Idaho National Laboratory's FX1 fogging solution, could be integrated into a powerful contamination capture system. The NNL's PDX was developed through the National Nuclear Laboratories work within the Chemical, Biological, Radiological and Nuclear (CBRN) response arena, where the laboratory has been using its expertise in waste characterization, waste management, and decontamination to allow post terrorist-incident recovery. The INL's FX1 chemical solution was developed in a joint Small Business Innovative Research (SBIR) grant with Vista Engineering (Hanford, WA) in 2006 to solve the problem of loose, airborne contamination during the D&D of facilities.

The PDX atomization technology has its roots in the development of marine drive systems, and was subsequently developed by *PDX Limited* for applications including sterilization, decontamination, and health care. The NNL recognized the potential for the technology to have numerous hazard mitigation applications within the nuclear industry, notably the knock-down and tie-down of airborne and other particulates. The technology utilizes the convergence of compressed air, and the solution (e.g. a decontamination reagent) through the atomizer head, within which rapid acceleration and impact of the two streams occurs, leading to atomization of the solution into an ultra-fine mist. The geometry of the unit is configured to provide a planar 360 degree exit, through which the mist exits at speed (Figure 2). In operation, the mist is initially projected laterally, in a cascade.



Figure 2, 360° atomizer

The high air flow generates a turbulence that maintains the movement and buoyancy of the mist. It is this turbulence that ensures the droplets disperse throughout a space and interact with both the line of sight and non-line of sight surfaces. Non-line of sight surfaces are not wetted in a paint spray technique, e.g. the underside of horizontal surfaces, the reverse sides of objects from the spraying location. It is this ability to effect such non-line-of-sight coverage/delivery that makes this technology such a step-change in capability. A single atomizer can be placed within a cell and within minutes deliver an ultra fine coating to all surfaces. An activity which would otherwise require a manned-entry, the erection of scaffolding to reach ceiling and upper walls, the movement of objects to permit access to the reverse and undersides of objects.

The initial objective of the FX1 solution development was to achieve a fogged capture coating that would allow for easy application, unmanned operation and flow through facilities and equipment. The solution had to penetrate through the surface layer of dust and “freeze” the dusty contaminants, fixing them to the substrate. Laboratory tests were performed to develop new formulations using different surfactants, binders and other components. These were applied in simple scoping studies to samples of lint and dust to simulate contamination. The simplest tests consisted of spraying these solutions onto pads of lint mixed with talcum powder. These tests indicated whether solutions were able to capture the dusty lint type of contaminants prepared in the beakers. Twenty different simple tests were completed using latex paint, glue, surfactants, glycerin, waxes and commercial fixatives common to the dust and asbestos industries. These solutions were evaluated on the ability of the solution to penetrate the top layer, contain dust and bind the material. Based on the results of these simple spray tests, solutions containing latex paint and surfactants were taken to fogging tests.

Two types of fogging equipment were employed during fogging tests. The first used a very small ultrasonic fogger (a home humidifier type). This was found to be unsatisfactory because the power was low (about 30 watts on the transducer) and could not atomize the higher viscosity and solids contained in these solutions. We found that even the typical industrial fogging equipment, that uses kilowatt sized transducers simply aren't capable of driving viscous solutions to a fog over long time periods. A second type of fogger, a low pressure medical nebulizer device, was ultimately employed to test the fogging ability of candidate solutions. Figure 3 shows the fogging unit as deployed in the laboratory using a water aspirator (left side) to manage the off-gas. Also visible are the red hot water bottle feed system going through a drip flow and the white air pump on the right side of the picture.



Figure 3, Tabletop nebulizer fogging unit used to test FX1.

This new coating demonstrated better penetration of the lint and dust, and an ability to stabilize respirable particles. All of the additives are common materials, found in paint and shampoo for example, and none are considered harmful (beyond being irritants in higher concentrations). The solutions are aqueous and generally clean up with soap and water. They show superior penetration into felt coupons exposed in the fogging system and contain talc dust added to the coupons (prior to fogging) much better than the baseline solution. In fact, no dust from the talc can be seen on the two candidate solutions.

Based on the laboratory tests that compared wetness, stickiness, dustiness and penetration, several coatings showed improved performance over the conventional glycerin fogging technique. A field demonstration of one of these fogging solutions was performed late in February 2007 at the INL in a 6 ft X 6 ft X 4 ft waste container. A 6 hr test was planned, but was cut short to 1.5 hrs (because of the failure of the large ultrasonic equipment). The solutions were not optimized for use in commercial fogging equipment, but were able to produce a fog and penetrate simulated lint coupons in this pilot scale test. No further testing was completed due to lack of funding.

Integrated Testing with NNL and INL

Based on the positive indications of previous testing, The Department of Energy, Environmental Management Organization (DOE-EM) provided funding to integrate the FX1 and PDX technologies. The purpose of this effort was to optimize the FX1 and PDX technologies so that a field demonstration of the integrated platform could be performed at a DOE site. While the collaborative effort proved that these two pieces could be integrated, the effort to optimize them into one technology took longer than originally estimated and didn't arrive at a satisfactory product until virtually the end of the project; thus precluding a field demonstration.

The integration began with a testing program to establish the FX1 compatibility with the PDXTM atomization technology. These tests were based upon the assumption that the viscosity and rheology of FX1 is similar to that of water under ambient conditions. Using a single PDX 360° nozzle, three trials were carried out to assess its ability to effectively fill a 45m³ the test chamber shown in Figure 4. Within the enclosure was a Malvern Spraytech particle size distribution apparatus that gave a readout of the physical characteristics of the fog.



Figure 4, FX1 Test enclosure showing particle size analyzer on floor.

For each test, the PDX 360° nozzle was placed in the center of the chamber, approximately 0.3 m below the ceiling to maximize the benefit of the Coanda effect (ability of the mist to adhere to the ceiling before falling to the floor) between the gas and atomized water exiting the nozzle and the chamber ceiling. During atomization of the water trials, the ventilation was set at the maximum to determine whether the mist created by the atomizer would remain stable.

During the first FX1 tests (FX1-1) the solution was fogged into the room at a liquid to air ratio of 2:1 (typical ratio for decontamination chemicals). The test included evaluation of coating various targets, both line of sight and non-line of sight, general ability to fog, fixation of particulate and particle size measurement. The targets including metal, plastic, wood and brick were arranged to allow in-line and non-line of sight surfaces to be tested (Figure 5). To assess tie-down of particulate, a tray containing fine particulate (powder <100 µm) was placed on the floor.



Figure 5, Objects for line of sight / non-line of sight testing

The 360 PDX nozzle was effective in atomising FX1 using the high liquid flowrate of 2:1 (2 parts liquid to 1 part air). Figure 6 (before and after) shows that the nozzle during FX1-1 was able to fill the 45m³ enclosure with a dense mist of FX1 within 45 seconds. A thick mist filled the room remained for 3 to 5 minutes after the spraying was stopped. However, this had cleared significantly within after 10 minutes. Figure 7 shows the monitoring of the key parameters, the

air flowrate to the nozzle, the pressure (in the liquid line), the ventilation flowrate and the ambient temperature during the FX1-1 trial.



Figure 6, Before and After 45 seconds of fogging FX1.

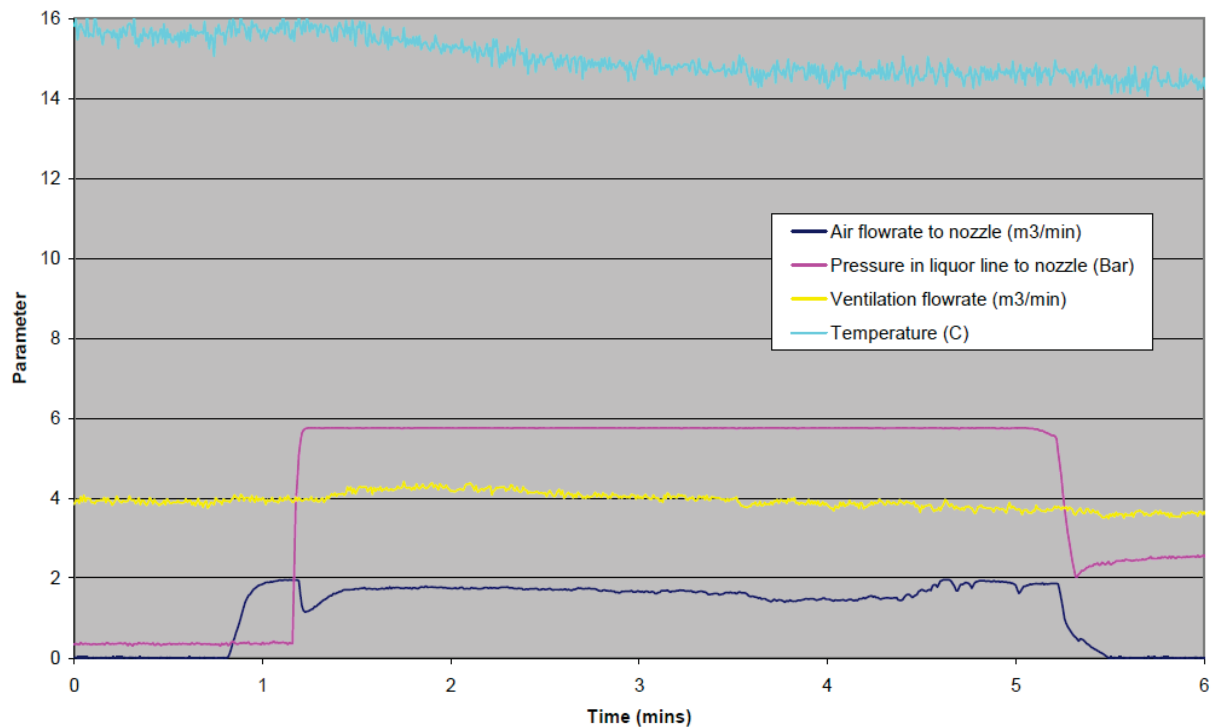


Figure 7, Operational Parameters for FX1-1 fogging test.

During these tests the particle size was much larger than anticipated. The expected particle size was in the neighborhood of 10 μm , with a good airborne particulate fog being less than about 20 μm . In these tests a small portion (10%) of the fog was in this size range, while most of it was in the 100-250 μm size range. This data is shown on Table 1. This larger size range of the fog particles leads to rapid agglomeration and settling. This could be advantageous in some

applications, particularly if just interested in just “clearing the air”. But our D&D application concept requires that the fog stay suspended for a longer duration in the air. Also note in Table 1 the “growth, or agglomeration, of particles after the fogger discontinued use. While this was more rapid than expected during this test, it is desirable for reducing airborne contamination; these size particles cannot remain airborne for long and will precipitate from the air.

Table 1, Particle size distribution for FX1-1 Test.

Solution	liquid to air ratio	Air Pressure (Bar g)	Dv 10 um	Dv 50 um	Dv 90 um
FX1 1 minute after start	2 to 1	6	8.7	116.2	335.2
FX1 5 minute after start	2 to 1	6	8.2	234.2	288.2
FX1 7 minute after end	2 to 1	6	14.3	238.5	278.6
FX1 37 minute after end	2 to 1	6	43.5	242.4	222.4

During this test we noted that the drying and coalescence of the latex film did not occur within a reasonable time. This was probably due to scaling factors that have to be overcome from the smaller, bench top fog (operating at milliliters per minute) unit, moving directly to a 4 liter per minute rate; the characteristics observed in the small scale weren't the same when orders of magnitude larger flow are involved. But, aside from the change in scale, the temperature and humidity during the FX1-1 test were different than when the small scale was performed. The small scale tests (2006) were performed at about 21°C, with a relative humidity of about 15%. The FX1-1 was at 16°C with about 90% humidity (raining during these tests). That is not to say that the FX1-1 conditions were inappropriate, it is important to know the limitations of the process. The conditions observed during the small scale tests were more ideal for proper coalescence.

While these FX1-1 tests were underway in the UK, supporting tests were being performed at the INL. Eighty liters of solutions were prepared and shipped to the UK for testing. Viscosity testing was performed on several different solutions in order to support the pump design evaluations. The viscosity data is shown in Table 2 indicates the viscosity data collected on these various solutions. This data shows the curious characteristic that the base formula displays a higher viscosity than one containing twice (and four times) the level of latex (shown as LTX), while reducing the quantity of surfactant (shown as SLS) has an inversely proportional result on the viscosity. The later effect is probably an anomaly, because that data was collected in a different series of experiments. However, none of these results had a significant effect on the pumping requirements because the cutoff for the pump is greater than 100 cP. An additional piece of viscosity data was collected with the addition of polyvinyl acetate (PVA). The use of PVA could

allow the fixative to potentially be used as a strippable coating. The effect of PVA on the viscosity was dramatic and likely would not allow a strippable formulation to be processed in this kind of misting head.

Table 2, Viscosity data on selected FX1 solution variants

Solution	Viscosity (cP)	Temp (C)
FX1	48.2	20.1
FX1 25%SLS	2.3	20.3
FX1 50%SLS	3.7	20.6
FX1 2XLTX	13.3	20.9
FX1 4XLTX	18	20.8
FX1 10%PVA	191.7	22.4

After these viscosity tests were completed, an additional round of FX1 (FX1-2) were performed in the UK. These tests took place in a smaller “cargo” container, so the volume of the test was reduced to about 33 m³ (and with less direct ventilation). The FX1-2 solutions contained twice the amount of latex (FX1 2XLTX) and twice the amount of latex and one-half the quantity of surfactant (FX1 2XLTX 50%SLS). Table 3 shows the particle size results of this second round of tests. The particle sizes were much lower, which is greatly improved over the FX1-1 tests. The best results from the FX1-1 test were a Dv50 of 116 um, while the FX1-2 tests ranges from 59 um to 11 um, up to a 90% improvement. The goal for these tests was to get to a Dv50 of 50 um, which was achieved in three of the five tests. Figure 7 show the inside of the cargo container with the test equipment.

Table 3, FX1-2 Test Results.

Solution	liquid to air ratio	Air Pressure (Bar g)	Dv 10 um	Dv 50 um	Dv 90 um
FX1 2XLTX	2 to 1	7	25.1	49.9	92.5
FX1 2XLTX, 50%SLS	2 to 1	8	27.2	58.7	116.5
FX1 2XLTX, 50%SLS	1 to 1	8	29.4	57.1	113.9
FX1 2XLTX, 50%SLS	1 to 2	8	4.3	13.1	36.1
FX1 2XLTX, 50%SLS	1 to 2	8	3.9	10.8	27.4

The FX1-2 series showed marked improvement in particle size distribution and seemed to have general improvement in vertical stability of the residue. Figure 8 shows that a good coating of the fog had settled on the particle size instrument and appears much more stable (less liquid) than



Figure 8, Cargo container with PDX and instrumentation.

in the FX1-1 test. Wetting was observed on both line of sight and non-line of sight surfaces (as in the FX1-1 test also), which is a positive outcome, and wetting of surfaces up above the height of the nozzle was demonstrated. The fog was able to “knockdown” any airborne contamination and coalesce on the floor. Since this was a smaller 33 m³ area, it was no surprise that the floor was completely covered with latex.



Figure 8, Solution retained on surfaces after fogging solution in the cargo container.

In real terms there is very little difference in the behavior of the two formulations tested in FX1-2. The droplet sizes are very similar (measured using a Malvern Spraytech placed just over 2 m from the nozzle). By reducing the liquid to air ratio to 1:1 more material was within aerosol than with the higher liquor to air ratio (ie 2:1). Decreasing the liquid to air ratio 1:2 showed smaller droplet sizes, but likely caused the nozzle to have a partial blockage. Therefore, a 1:1 ratio seemed to be the best compromise. Additional testing could result in finer droplet sizes if the solution viscosity was reduced.

Conclusions

The integration tests of the fogging solution and the misting technology have shown that they are compatible and have significant potential for use as a contamination control method. The two technologies were developed without consideration of using them together, thus it is little surprise that some development was required to achieve a proper balance of characteristics. The FX1 fogging solution had been developed to a “bench-top” level of maturity, using entirely different equipment and conditions than the pneumatic PDX atomizer. Given that, the first formulation of FX1 and PDX showed many good qualities:

- A dense fog was produced that lasted over 10 minutes.
- Coalescence that binds dry contamination to the substrate.
- Line of sight and non-line of sight coverage.

These are highly desirable traits and balanced the lack of thickness and dryness of the product that were observed. However, the results of those first tests were inadequate to recommend immediate transfer into a full demonstration.

These are highly desirable traits and balanced the lack of thickness and dryness of the product that were observed. However, the results of those first tests were inadequate to recommend immediate transfer into a full demonstration.

Instead, a second round of testing was initiated with the purpose of further developing the two technologies into a more optimized system. In this second round the FX1 fogging solution was reformulated to reduce the concentration of surfactant and to increase the latex base formula. The results were that the droplet size distribution was dramatically reduced (with twice the latex and one-half the surfactant). This was combined with a hardware change of reducing the liquid to air ratio (1:1) which improved results. However, a 1:2 ratio, which gave even more ideal droplet size, caused plugging of the PDX nozzle. As the new formulation settled it seemed to give a more uniform coating. While drying may still be an issue, with proper ventilation, that is likely to be a lesser concern. Unfortunately, due to recent business issues apart from this effort, it appears PDX will be unable to supply a nozzle for a demonstration. However, alternatives are being examined to satisfy this gap and you (Mr. Szilagyi) will be involved in the selection.

One of the primary advantages within this collaboration is the pathway for cooperative development of other projects. Our international collaboration on this project allowed both organizations to progress with the technology development and build on their individual strengths, with a strong focus on both US and UK decommissioning challenges. An additional outcome is the concepts for follow-on work that have been developed; among these are using the coating to disperse sub-micron mercury absorbent and to develop it as a sealant that could be used against indoor pollutants like allergens.

Sincerely,

A handwritten signature in blue ink, appearing to read "Rick L. Demmer". The signature is fluid and cursive, with a large initial "R" and "D".

Rick L. Demmer
R&D Scientist
Aqueous Separations & Radiochemistry

cc: J. J. Grossenbacher, INL, MS 3695

bcc: INL Correspondence Control, MS 3105, email: BEACC@inl.gov
R. L. Demmer Letter File: RLD-13-002

Uniform File Code: 8403

Disposition Authority: RD1-A-1

Retention Schedule: Permanent. Cut off after project/program completion, cancellation, or termination.

NOTE: Original disposition authority, retention schedule, and Uniform Filing Code applied by the sender may not be appropriate for all recipients. Make adjustments as needed.