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Removable, Sequestration Coatings for Mitigating Hazardous Contaminants Related to Deactivation and Decommissioning Activities

Baohua Gu
David Watson
Kenneth Lowe
Xiangping Yin

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OAK RIDGE National Laboratory

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REMOVABLE, SEQUESTRATION COATINGS FOR MITIGATING HAZARDOUS CONTAMINANTS RELATED TO DEACTIVATION AND DECOMMISSIONING ACTIVITIES

Baohua Gu, David Watson, Kenneth Lowe, and Xiangping Yin

January 2022

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OAK RIDGE NATIONAL LABORATORY
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ABSTRACT

This project is a collaborative effort between InnoSense LLC (ISL) and Oak Ridge National Laboratory (ORNL) aimed at mitigating the mercury (Hg) contamination problem at the Y-12 National Security Complex (NSC). The report summarizes the research and technology development work of the Phase IIB Project, led by Dr. Uma Sampathkumaran at ISL for the period of July 31, 2017–October 31, 2021. The Y-12 site cleanup includes technologies for safe and cost-effective facility demolition, soil remediation, and on-site treatment and disposal of mercury bearing wastes. One of the technology demonstration goals was a reactive strippable coating for targeted pre-demolition and de-contamination to limit Hg mobilization and the volume of debris requiring treatment for disposal. Toward this, ISL has developed a water-based reactive strippable coating (Trap & See-Hg™) to sequester elemental Hg(0) and mercuric Hg(II) species in the coating. The coating provides multiple benefits including (1) Hg vapor suppression, (2) sealing in the hazardous contaminants, (3) visually locating the contaminant through color changes, and (4) remediating the contaminated surfaces upon coating removal.

Through collaboration between ORNL and ISL in Phase II work, the team successfully demonstrated the Trap & See-Hg technology to (1) visually locate the presence of Hg on concrete, soil and drywall surfaces, (2) effectively sequester all mercury species (elemental, soluble and insoluble Hg) from lab-generated and ex-situ soil and brick/rubble samples collected from the Y-12 site with 80 to 95% efficiency to capture/reduce Hg vapor concentration, (3) be applied by manual and robotic spray applicators and cure at room ambient, (4) remain a cohesive and peelable coating upon drying, and (5) sequester Hg^0 from walls in the alpha-2 building basement at Y-12 during a small scale *in-situ* field trial.

In Phase IIB, ISL's effort was focused on refining the coating formulations with additives containing passive and reactive sorbents, colorimetric indicator, and thixotropic agents to achieve the desired performance. The paints were then evaluated at ORNL for applicability to metal and concrete surfaces of different sizes and shapes, and soil rubble, Hg-sorption capacity and achieving sag-free coating on surfaces when applied by a manual spray applicator. Ambient cure of the coating resulted in a dry thickness of 16–20 mil. The coatings were removed from smooth or rough surfaces with low to moderate force. Both lab-generated Hg-contaminated samples and soil and brick/rubble samples containing a range of Hg concentrations (collected from the Y-12 site by UCOR) were used in this project. Optimized formulations were applied to these samples and evaluated for ability to suppress Hg vapors, sequester Hg^0 and Hg(II) species and color mapping of mercury species on the test surface.

The ORNL team initially identified three to four potential sites at Y-12 and ORNL for potential demonstration. However, the pandemic closed any possible avenues for on-site demonstration of the robotic spray applicator. The metal and concrete samples with low to high Hg content provided by UCOR from an ongoing demolition were subsequently used for *ex-situ* sample characterization and analysis at ORNL over a period of 25 days. The coatings suppressed Hg vapors with an efficiency of 95–99% from heavily contaminated metal and 60% from concrete substrate. Analysis of total Hg in the peeled paints indicates sorption capacities ranging from 12 µg Hg/g of paint from concrete to 1.2 mg Hg/g of paint from the metal substrates while peeled paints from soil samples could capture a total Hg from 2.3 mg Hg/g to 66 mg/g paint.

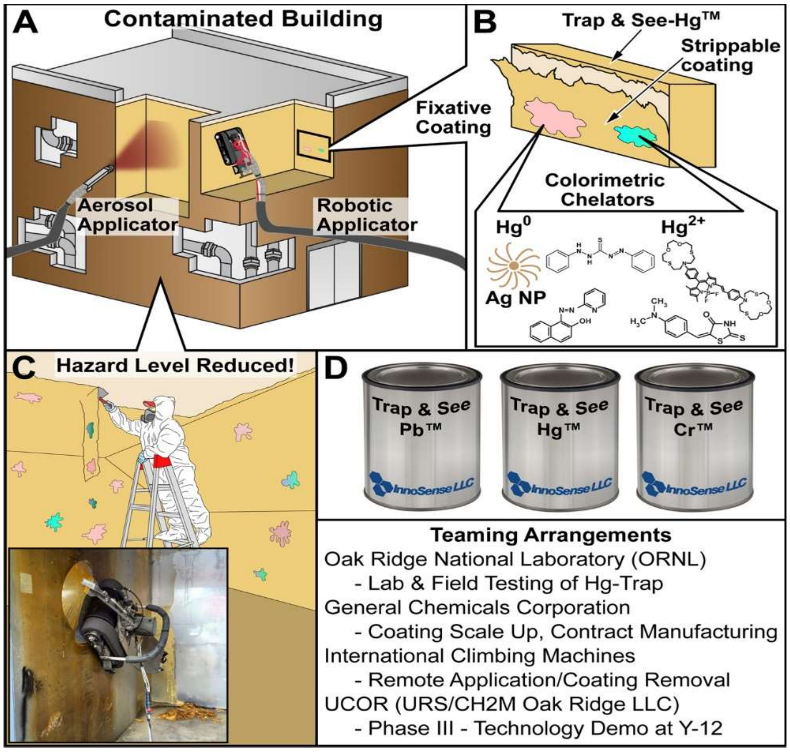
Toxicity characteristic leaching procedure (TCLP) tests recovered total mercury leached at 0.15±0.0008 µg/L for concrete to 126.81±0.67 µg/L and 377.25±5.67 µg/L for the two metal substrates, to allow for informed decisions on downstream disposal considerations. Together, this Phase IIB demonstrated the potential value of the Trap & See-Hg for Hg uptake, vapor suppression, and color mapping, and may thus be used to enhance worker safety during site cleanup, facility demolition, and downstream waste disposal at Y-12 NSC.

1.0 INTRODUCTION

The Oak Ridge Integrated Facility Disposition Project (IFDP) involves demolishing and upgrading infrastructure at the Y-12 Complex, along with the remediation of soils, groundwater, and surface water. Mercury has been identified as a high priority contaminant and new ways to remediate and monitor this contaminant are required. The Department of Energy (DOE) has estimated that $1B will be needed just for mercury remediation at Y-12.[1] During the 1950s and early 1960s, Y-12 buildings were used to process enriched lithium for hydrogen bombs.[2] Twenty four million pounds (lbs) of mercury were used in those operations, of which 2 million lbs of Hg were either spilled, lost or unaccounted for. An estimated 700,000 lbs entered the environment: air ~51,000 lbs; water ~240,000 lbs; soils/sediments unknown; building spills ~325,000 lbs and process systems ~60,000 lbs.[3]

Various forms of Hg (elemental, inorganic and organic) in the environment are toxic and hazardous materials known to damage the lungs, kidneys, and the central and peripheral nervous system.[4,5] Organic Hg species, such as methylmercury, cause nervous system damage leading to problems with coordination, numbness, and visual impairment.

Oak Ridge Environmental Management (OREM) has performed several projects to reduce offsite mercury migration since 1985 and is actively investigating technologies that can effectively remove it from the environment. Examples include elimination of some of the mercury sources in and around mercury contaminated tanks and facilities, cleaning of storm drain systems, excavation of contaminated soil, dredging of sediments, re-routing and removing old process piping, extraction of nearly five tons of mercury from old equipment, and the construction of an onsite mercury treatment system. To date, OREM has removed all of the old, deteriorated, mercury-contaminated Column Exchange (COLEX) equipment on the west side of Alpha-4. Crews also drained and cleaned out the equipment on the east end of Alpha-4 to continue preparing for eventual demolition. Decontamination methods were also tested to clean old mercury process piping, and a field test commenced for a newly developed fogging fixative and application process aimed at controlling mercury vapors during future deactivation and demolition projects at Y-12's hazardous, mercury-contaminated buildings.[2] Among the key objectives of the IFDP project at Y-12 complex relevant to Trap & See-Hg are: (1) developing a remediation and monitoring strategy for controlling contaminant release from source zones to ground and surface water, and (2) developing innovative cleanup technologies applicable to DOE sites. The Trap & See-Hg coating is aimed to fix, seal and aid in visualizing mercury during the remediation operations while reducing workplace hazards and minimizing hazardous waste volumes. An artist's rendition of the envisioned uses for Trap & See-Hg is shown in Figure 1.



Teaming Arrangements

Oak Ridge National Laboratory (ORNL)
  - Lab & Field Testing of Hg-Trap

General Chemicals Corporation
  - Coating Scale Up, Contract Manufacturing

International Climbing Machines
  - Remote Application/Coating Removal

UCOR (URS/CH2M Oak Ridge LLC)
  - Phase III - Technology Demo at Y-12

Figure 1. *Illustration of the strippable coating with a wall-climbing robot and spray applicator with potential for use in highly hazardous buildings/sites **(A)**. Strippable Trap & See-Hg coating with embedded chemochromic agents and sorbents to bind and visually indicate the presence of the hazardous contaminants **(B)**. Pre-demolition activity to minimize the: (1) safety risks for workers involved in deactivation and demolition activities, (2) waste volumes and (3) segregate the waste for disposal **(C)**. Hazard level is reduced allowing manual or robotic operations as needed **(D)**. Expanding Trap & See™ commercial product line.*

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Several commercially available products have been studied by DOE for their potential use as fixatives.[6] A robotic spray applicator from ICM was used to investigate the efficacy of three different coatings. The coatings, however, were not evaluated for removability in this study. Independently, the Environmental Protection Agency's National Homeland Security Research Center (NHSRC) evaluated two strippable coatings for radiological decontamination efficacy, to protect and safeguard an urban population in the event of a dirty bomb. [7,8,9,10] Most of the strippable materials are passive entrainment coatings or targeted to bind radionuclides which do not visually signal the presence of the contaminant and do not decontaminate in a single application.[11] Additional drawbacks include the use of carcinogenic solvents and chelators, and materials that are unpleasant to hazardous (e.g., ammonia).[12] Responsive, strippable coatings have been developed by researchers for capture of radionuclides. [10,11,12] A visible color change and high decontamination efficiencies are reported for uranium and plutonium.[11] Smart removable coatings with enzymatic degradation capabilities for chemical warfare agents are also reported.[13]

This project focuses on water-based, color-generating (chemochromic), strippable coatings for the high efficiency capture and visualization of mercury. During the initial Phase II, ISL developed water-based, strippable, chemochromic coatings (Trap & See™) to capture hazardous contaminants with visual indication capabilities. The paints for capturing elemental (Hg^0) and ionic mercury (Hg^{2+}) are branded as Trap & See-Hg™. Chemochromic indicators and reactive/passive sorbents in the coating selectively complex with Hg^{2+} and Hg^0 and indicate their localized presence through color formation. The visible limit of detection (LOD) is well below the land disposal requirements limit of 0.2 mg/L. The Hg-uptake capacity of the coatings ranged from a few tens of µg/g of total Hg to hundreds of µg/g of total Hg in brick and soil samples from the Y-12 site.

Trap & See-Hg was tested initially in a small scale *in-situ* trial at Y-12 by the Oak Ridge National Laboratory (ORNL). Small areas of the alpha-2 basement wall, beam and floor were manually sprayed with the paint after establishing Hg-vapors were present. The dried paint from two different locations was peeled and analyzed indicating that 4 µg/g of Hg and 12 µg/g of Hg, respectively, were sequestered in the paint.

2.0 STATEMENT OF OBJECTIVES

The main objectives of the Phase IIB project were focused on:

1. Refining Trap & See-Hg to characterize the Hg content from *ex-situ* samples and scale up for small and large scale site trials (by ISL).
2. Validating Hg-sequestration by headspace vapor reduction measurements and total Hg captured in the paint through testing at ORNL and a third party analytical test facility.
3. Demonstrating a safer method to remediate remote and highly contaminated areas using Trap & See-Hg and a robotic applicator.
4. Establishing the efficacy of Trap& See-Hg for *in-situ* characterization, sealing hazards and classifying waste for on-site Land Disposal Requirements (LDRs) or off-site processing.

Through collaboration between ISL and ORNL, all Phase IIB project objectives have been met with the exception of on-site demonstration and *in-situ* characterization due to the challenges of accessing the site for security and pandemic related remote working modality. The accomplished work offers a potential pathway for safe and cost-effective targeted pre-demolition decontamination for mercury contaminated buildings by: (1) characterizing the nature and extent of contamination, (2) limiting the volume of debris requiring treatment or disposal, and (3) minimizing environmental mobilization of mercury.

3.0 BENEFITS TO THE DOE MISSION

Development of the new and unique coatings and application systems is consistent with DOE's mission of Environmental Management and developing cost effective technologies for the remediation and demolition of legacy facilities. The work is also aligned well with ORNL mission and capabilities including: 1) Environmental Science and Engineering, 2) Applied Materials Science, 3) Chemical Engineering, 4) Mechanical Design and Engineering, and 5) Decision Science and Analysis (waste segregation and disposal pathway analysis). The need for the development of the coating technology has specifically been called out and discussed by DOE in the *Technology Plan to Address the EM Mercury Challenge* (DOE/ORO-2489, 2016). The findings of this work could potentially offer a pathway for a safe and cost-effective targeted pre-demolition decontamination for mercury process buildings by (1) limiting the volume of debris requiring treatment or disposal and (2) minimize environmental mobilization of mercury.

4.0 TECHNICAL DISCUSSION OF WORK PERFORMED

4.1 Materials and Methods

4.1.1 *Coating formulation*

The coatings are formulated by ISL with water-borne polymeric emulsions containing surfactants to aid wetting and dispersion, ligand-based chromogenic (or chemochromic) indicators, active and passive sorbents with specificity for mercury with non-hazardous solvents and buffers. Hg^{2+} is a soft metal cation, characterized by "soft sphere" of highly polarizable electrons in its outer shell. This leads to a preference for ligands of sulfur, the less electronegative halides and nitrogen over ligands containing oxygen.[14] Upon Hg^{2+} binding, they offer visual indication of mercury-entrained regions through color development. Several colorimetric and fluorescent indicators for heavy metal ion sensing are reported to be highly sensitive.[15,16] However, they often require custom synthesis or an external fluorescence excitation source to visualize color changes. They are expensive and, in some cases, have limited solubility in water. Hence our choice was narrowed to two commercially available complexing agents that are water/surfactant soluble with moderate to high specificity to Hg^{2+} and further down selected to one. Formulations were initially made in the laboratory and eventually scaled to 1-, 5- and 10-gallon quantities. In the initial Phase II, the paint's sag resistance was not optimized. Paints require viscosity be low at high shear rates for good workability (particularly spraying), and high viscosity at low shear rate to avoid excess spreading and sagging. Thixotropic formulations are useful to balance these two properties for the paints. The challenge is to obtain the level of thixotropy that leads to good deposition performance. In Phase IIB work, ISL investigated thixotropic agents and conducted a statistically designed experiment to identify the formulation ingredients and process parameters that impact the sorption capacity for Hg and color formation, while preventing spreading or sagging.

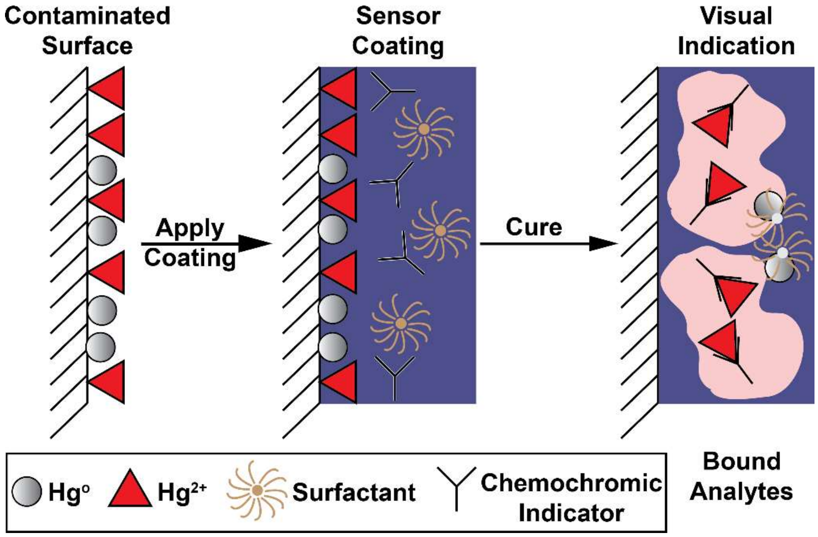

Figure 2. *Working principle of the Hg-sequestration and visualization coating. Binding events for ionic and elemental mercury are depicted through symbols and color changes in coatings visible to the eye.*

4.1.2 *Coating methods and substrates*

Doctor blade and spray coating methods were used to apply Trap & See-Hg on various substrates, namely drywall, concrete, plastic, glass, metal as well as soil and rubble from the Y-12 site. The wet coatings were cured at room temperature for at least 24 hours. The coatings were peelable after drying by applying low to moderate force for smooth and rough substrates. Viscosity was adjusted for both spraying and doctor blading to achieve dried coating thickness of approximately 0.25–0.3 mm (10–12 mil). The doctor blade method is good for applying coatings to flat substrates, such as glass and drywall. A Wagner Power Stainer Model 0525155 spray applicator was used to spray coat all test surfaces 15 cm x 15 cm and 60 cm x 60 cm or 60 cm x120 cm.

4.1.3 *Hg-contaminated samples*

Two types of samples were investigated. The first were elemental mercury and mercuric chloride-contaminated samples generated in the lab while the second was Hg-contaminated samples from the Y-12 site. For elemental Hg contaminated samples, concrete pavers (~9 cm x 9 cm) were exposed to mercury vapors generated with a custom flow through system shown in Figure 3. Slow moving UHP Nitrogen (N_2) carrier gas flows in from the top of the glass impinger with a small droplet of liquid Hg at the bottom. The Hg contaminated N_2 gas exits from the side port of the impinger and flows into a glass container with a sealed polymer lid. The polymer lid has an outlet and an inlet. The Hg carrying N_2 flows into the inlet and after infusing the interior atmosphere with a steady Hg concentration, exits through the outlet. The exiting Hg carrying N_2 flows through another glass bubbler filled with flowers of sulfur. The Hg reacts with the fine-grained sulfur to form a passive non-aqueous mercuric sulfide (HgS), insoluble in aqueous non-acidic environments.

For color formation and visible limit of detection tests, mercuric chloride was used as the contaminant. Stock solution of 10 mM mercuric chloride ($HgCl_2$) was prepared in distilled water and serially diluted to 1 mM, 100 μM, 10 μm and 1 μm. A fixed volume (20 μL) of these solutions were deposited to arrive at a final concentration of 40, 4, 0.4, 0.04 and 0.004 μg of Hg on the test substrates, typically 15 cm x 15 cm drywall or concrete pavers.

ORNL also collected contaminated concrete pads, ceramics, and other building materials samples from the Y-12 NSC site. These samples were from the surface and shallow subsurface surrounding the former Y-12 mercury retort building 81-10. These materials were evaluated for Hg contamination (by measuring

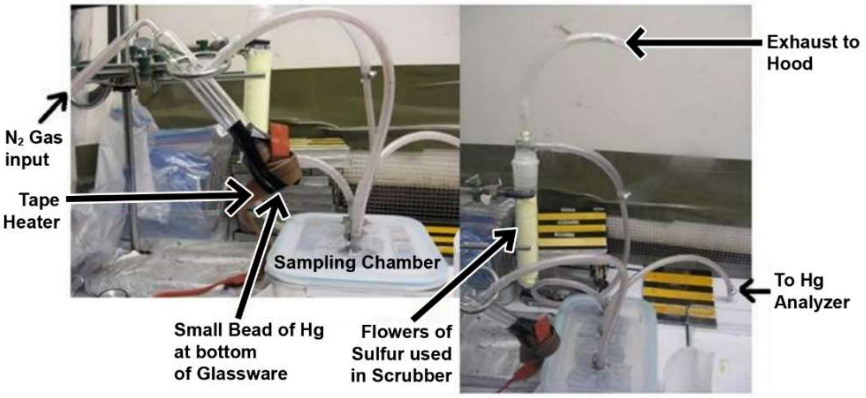

Figure 3. *Setup of the Hg contamination chamber for test sample preparation.*

elemental Hg^0 vapor in sealed bags) and radioactivity. Speciation of the Hg in these samples was evaluated using Hg^0 analyzer and chemical sequential extraction following techniques developed at ORNL.[17] Following complete evaluation of these samples, the ORNL team sent three non-radiological samples (concrete, rubble and brick) to ISL for additional evaluation. Leftover contaminated rubble and the highly Hg-contaminated soil sample (~815 mg/m^3 measured at ORNL) from the initial Phase II were subsequently used during the Phase IIB period. URS | CH2M Oak Ridge (UCOR) personnel provided additional Hg-contaminated debris (10 cm by 30 cm long concrete and cut metal plates from equipment) with varying Hg levels from an ongoing demolition at the Y-12 9201-4 building. These samples were used to test the efficacy of the paint to capture and suppress the vapors and were subsequently retained for *ex-situ* testing at ORNL.

Additional tests were performed with Drywall (15 cm x 15 cm) or concrete (15 cm x 15 cm) substrates contaminated with Hg(II) solutions in the concentrations from 40 µg to 0.004 µg in lanes separated by tape. The contaminated surfaces were allowed to dry overnight for a nominal of ~16 hours before applying the paint by doctor blading technique. The paint was allowed to cure at room temperature and evolution of color was observed by visual inspection. Influence of formulation additive on color formation and limit of visible detection were investigated.

4.1.4 *Measuring headspace reduction of mercury vapors*

Using the *ex-situ* EMA-2 soil rubble sample, the headspace reduction and mercury sorption capacity of the paints was evaluated by ISL using a Jerome J505 Mercury Vapor Analyzer. First, the vapor phase Hg concentration was measured (in triplicate) in the headspace of the container of Hg ORNL EMA-2 soil, by opening the bag and inserting a sampling tube from Hg analyzer. The Hg concentration was so high that aliquots of the soil sample (wt./wt. basis) were mechanically blended with different concentrations of clay. To be realistic in the testing, commercially available Tennessee Ball Clay SGP#1 was used with the ORNL soil and then placed in a rotating mill in a small bottle for a nominal 16 hours. This allowed for a layer of ORNL soil and native clay to be deposited as an aqueous slurry onto the substrates. The native Tennessee clay SG-1 acts as a binder to hold the ORNL soil in a coherent mass, to avoid the soil falling off. The intent was to provide an ORNL EMA-2 soil rich surface for the paint to adhere to and then remove the dried paint from the clay + ORNL EMA-2 soil. The samples were coated with pourable paints that dried in a nominal 24 hours to an adherent tape. Headspace measurements of the test samples were made before and after paint application and of the test surface after peeling the paint. Color changes due to the colorimetric indicator were seen and photographed in the paint applied to the soil samples.

4.1.5 *Peeled paint analysis*

All peeled paints from concrete and soil samples were shipped for total mercury analysis at ORNL. The peeled paints from the soil samples and drywall contaminated were also analyzed by two methods at ORNL. Since the paints have been formulated with the goal to sequester all soluble and insoluble mercury(II) compounds, the first step involved dissolving the dried paint in acetone to extract soluble Hg. A small aliquot of the extract was then used for determining the total Hg via bromine monochloride (BrCl) oxidation, followed by stannous chloride ($SnCl_2$) reduction. The total Hg concentration was determined by a previously developed method at ORNL.[17,18] Therefore, the Hg concentrations can be considered as the minimum estimates.

4.1.6 *Ex-situ testing of low and high-Hg contaminated UCOR samples*

ISL provided the ORNL team with one gallon of paint, a Safety Data Sheet, a protocol for manual (brush/pour) and/or spray application of the paint on the samples. Testing was conducted at ORNL who took custody of the Hg-contaminated samples from UCOR. All measurements and handling of Hg-contaminated samples were conducted in the presence of a safety specialist who recorded the Hg headspace readings around the work area. Briefly, Hg contaminated concrete and metal samples were placed inside buckets and capped. Hg headspace readings were obtained by sampling 100 or 200 µL of the headspace volume using a sampling syringe and analysis using a Lumex analyzer at 1 hour and 144 hours. The samples were painted, and headspace readings were obtained at 24 hours and 144 hours. Part of the paints were removed 2 weeks after the paint was applied and Hg readings were taken of the partially bared samples at 24 hours and 72 hours. The samples were then stripped of all of the paint, 10 days after the partial removal, and headspace Hg values were measured at 144 hours. The peeled paints were then tested again at ORNL for Hg content to determine the efficiency of the paint to sequester elemental Hg.

4.1.7 *Toxicity characteristic leaching procedure (TCLP)*

The Toxicity Characteristic Leaching Procedure (TCLP) analysis simulates landfill conditions. Over time, water and other liquids percolate through landfills, often reacting with the solid waste in the landfill, and may pose public and environmental health risks because of the contaminants it absorbs. The TCLP analysis determines which of the contaminants identified by the United States Environmental Protection Agency (EPA) are present in the leachate and their concentrations. TCLP is designed to determine the mobility of both organic and inorganic analytes present in liquid, solid, and multiphasic wastes. This is usually used to determine if a waste meets the definition of EP Toxicity, that is, carrying a hazardous waste code under RCRA (40 CFR Part 261) of D009 for Hg with a maximum limit of 0.2 mg/L.[19,20]

Guided by this information, TCLP analysis of the stripped coatings was carried out at ORNL on three samples, representative of low and high-Hg contents.

4.2 Results and Discussion

4.2.1 *Evaluating ISL refined paint formulations*

We first investigated the paint for color evolution on drywall samples. All sixteen paints showed color formation when the drywall surface was contaminated with 10 µg and 1 µg Hg from $HgCl_2$. We examined the following variations for the paints to see color evolution on drywall contaminated with $HgCl_2$.

1. No passive sorbent.
2. Solvent and passive sorbent stock solutions include surfactant.
3. Indicator added directly to Base Paint, eliminating solvent.
4. Alternate vendor source for solvent.
5. Control paint with no additives.
6. Replaced solvent used to dissolve the sorbent with water.
7. Used freshly prepared indicator solution (not stored).

8. Indicator added directly to Base Paint, no change to overall composition (made 3 days prior to use compared to other paints in this list).
9. CID# 2888: Scaled up paint from initial Phase II (2 year aged paint).

4.2.2 *Effect on color evolution*

The color evolution and results from the paint analysis are shown in Figure 4. It can be seen that the limit of detection in almost all the cases in the presence of Dithizone was 0.25 µg or 250 ng with some color fading in Panels 8 and 9. The observations for each variation identified above are summarized below.

Var 1 – Background paint color lighter affording slightly improved contrast and visible LOD of 250 ng (Panel 1).

Var 2 – Similar visible LOD as Panel 1 (Panel 2), lower contrast.

Var 3 – Paints that appeared to thin in the middle and thicker at the edges with more intense color (Panels 3 and 6).

Var 4 – 250 ng visible LOD (Panel 4), better contrast.

Var 5 – Control sample Base Paint – No color as it lacks indicator (Panel 5).

Var 6 –Similar thinning effect in paint (Panel 3), color visible with LOD at 250 ng to (Panel 6).

Var 7 – Color bright with visible LOD at 250 ng (Panel 7).

Var 8 – Color washed out, possibly poor indicator dissolution in paint (Panel 8).

Var 9 – Aged (2-year) paint from Phase II – visible LOD ~250 ng, color faint (Panel 9).

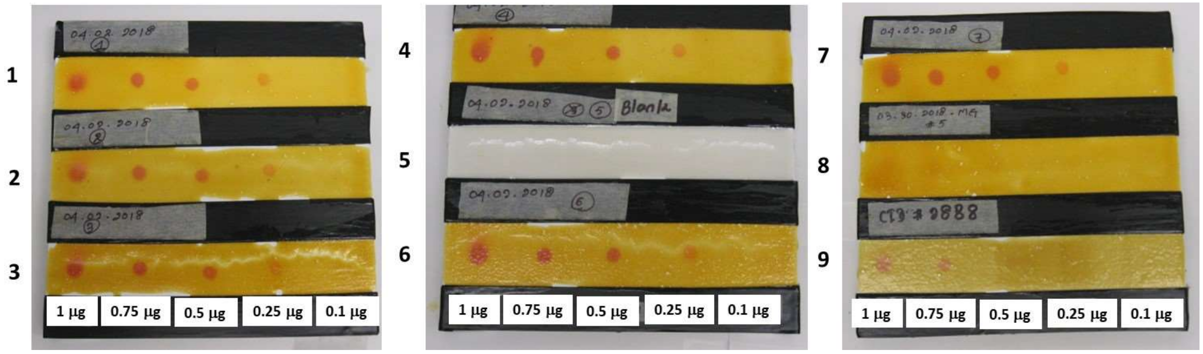

Figure 4. *Color evolution in various paints for $HgCl_2$ contaminated drywall surfaces. Limit of detection down to 250 ng.*

A similar study was conducted by contaminating concrete surfaces with 40 µg and 4 µg $HgCl_2$ shown in Figure 5 (Left). After paint application and drying, color can be noted in the contaminated regions on the concrete surface (Figure 5, Middle). After peeling the paint, the color appears more intense on the inner side of the paint that was in contact with the concrete surface (Figure 5, Right). These results suggest that the visible LOD on the concrete surface maybe hampered by the dark background color, however, the paint does sequester the contaminant from the surface.

4.2.3 *Testing elemental Hg in concrete samples*

Two groups of six concrete samples each were used to evaluate the effectiveness of the paint formulations. One group consisted of samples deliberately exposed to Hg vapors for an extended period of time at ISL. The other consisted of Hg-contaminated debris samples (ORNL) from Y-12 NSC. Three paints were used in each case, with two samples each for duplication.

Figure 5. *(Left) $HgCl_2$ contaminated concrete, (Middle) After paint application and drying - Limit of detection tested was 4 μg; (Right) The peeled paints showing color in the locations where Hg (II) is sequestered.*

To generate samples contaminated with elemental Hg, concrete pavers acquired by ISL were exposed to elemental mercury using the test setup shown in Figure 3. After the samples were exposed to Hg – N_2 vapors for a prescribed time, the headspace Hg was measured with the Jerome 505 Hg Analyzer. The ISL-generated Hg-contaminated concrete samples and ORNL samples are identified in Table 1. Three paints were used: 050818-MM-3111 (new paint composition reduced solvent and reactive sorbent), CID# 2888 (old paint composition), and CID #2923 (Control, no additives).

The ORNL debris samples did not have any measurable headspace Hg values. This matched the ORNL input that these samples had been lying around in ambient, so any elemental Hg was lost to the environment over time, but ionic Hg species could be present. All headspace measurements were made with a Jerome J505 Hg vapor analyzer.

Table 1. ISL and ORNL Sample IDs and Paint IDs

ISL Elemental Hg-contaminated Samples		**ORNL Concrete Samples (Hg^0 and Hg(II)?)**	
Concrete Sample ID (Hg^0)	**Paint ID**	**Concrete Sample ID (Hg^0 and Hg(II)?)**	**Paint ID**
050818-MM-3111-1	**050818-MM-3111**	**051118-RW-3111-1**	**050818-MM-3111**
050818-MM-3111-2	**050818-MM-3111**	**051118-RW-3111-2**	**050818-MM-3111**
050818-MM-3111-3	**CID# 2888 (2-yr old)**	**051118-RW-3111-3**	**CID #2888**
050818-MM-3111-4	**CID# 2888(2-yr old)**	**051118-RW-3111-4**	**CID #2888**
050818-MM-3111-5	**CID #2923 (Control)**	**051118-RW-3111-5**	**CID # 2923 (Control)**
050818-MM-3111-6	**CID# 2923 (Control)**	**051118-RW-3111-6**	**CID # 2923 (Control)**

4.2.4 *Paint application and testing*

Samples from both groups and the corresponding paints that were applied are identified in Table 1. A small volume pipette (5–10 mL) was used to dispense paint onto one of the sides of the test substrate. The paint was spread on the surface using a paddle to cover the entire surface and then worked into any crevices in the concrete. More paint was pipetted as needed. The paint was dried overnight followed by application of the paint on the exposed surface, followed by drying. The procedure is repeated until the entire substrate is covered with the paint and dried to fully encase it.

The dried paints were inspected for color, measured for elemental Hg vapor concentration, and then peeled for analysis. The mass of the twelve dried paints were measured. A portion from each of the twelve paint samples was shipped to ORNL for total Hg analysis by Cold Vapor atomic absorption analysis.

Headspace measurements were made using the Jerome J505 Mercury Vapor Analyzer as previously described. All measurements were made of the samples placed in sealed triple Ziploc bags after 30 minutes of equilibration. Headspace measurements were made four times:

1. Before paint application,
2. After paint application and drying,
3. Of the peeled paint, and
4. Of the residual Hg concentration on bare concrete.

Table 2. Summary of Headspace Measurements

	Total Headspace Hg ($\mu g/m^3$)			
Paint ID	**Before Paint Application**	**After Paint Application**	**Peeled Paint**	**Concrete After Paint Removal**
050818-MM-3111-1	21.65	0.04	0.12	0.79
050818-MM-3111-2	24.00	0.09	0.09	0.72
050818-MM-3111-3	30.62	0.18	2.92	2.12
050818-MM-3111-4	22.95	0.54	0.21	1.90
050818-MM-3111-5	60.20	0.68	0.98	3.14
050818-MM-3111-6	49.72	1.06	0.80	7.04

From the summarized data in Table 2, the initial [Hg] in concrete samples appear high, before coating with paint. For dried painted samples, the [Hg] levels are at their lowest before peeling the paint. After the dried paint is peeled, the paint [Hg] levels appear generally slightly higher than the samples before paint peeling possibly due to exposed surfaces. After the paint is peeled, the residual [Hg] on the bare concrete is higher than the paint, but much lower than the initial base concrete, indicating sequestration in the paint and remediation of the substrate surface. Overall, 27x, 34x, 14.4x,12x, 19x and 7x reduction in [Hg] was noted indicating the paint had vapor suppression efficiencies ranging from 97% to 86% for the duplicate samples tested with three paint variants.

It should be noted that the headspace measurements are not a quantitative measure of the Hg concentration in the samples – initial bare concrete, painted concrete, paint after peeling, or bare concrete after the paint was peeled. During the drying process, Hg can easily penetrate further into the concrete, making for a lower headspace value, for a sample potentially with a higher Hg content. On the other hand, some Hg may have diffused through the paint into the atmosphere during the drying process. It is also feasible that the [Hg] values in the paint could be low because the paint sequestrants are doing their part to prevent the Hg vapors from escaping to the atmosphere. By averaging the headspace values for samples (from Table 2) with same paint ID (paired formulations in Table 8), we get an average reduction in headspace of 97% for the new paint formulation, 92% for the aged paint and 89% for the control paint for the limited sample size of two.

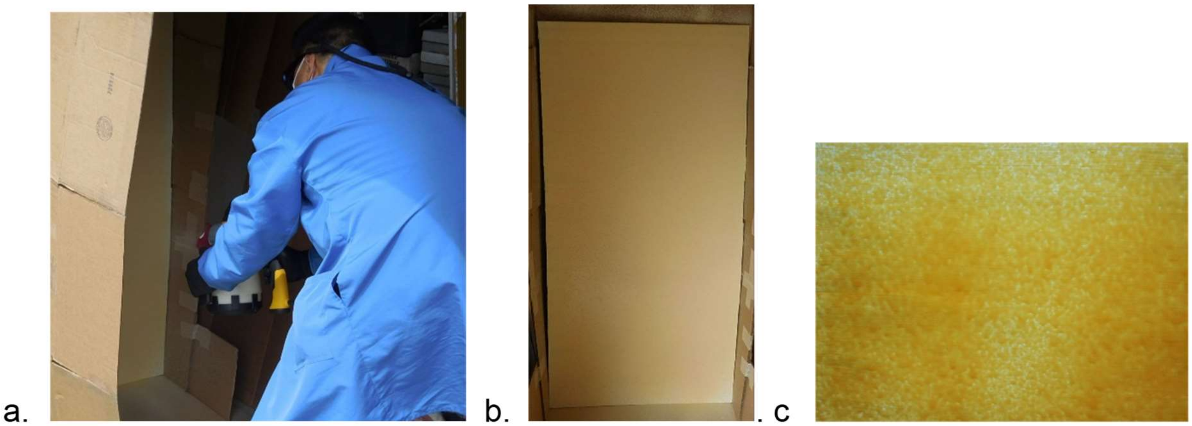

Figure 6. (a) *Spray painting on 4' x 2' dry wall,* ***(b)*** *after spray coating, and* ***(c)*** *24-mil wet thickness sprayed coating.*

4.2.5 *Spray painting on dry wall*

ISL paint 050818-MM-3111 was tested for spray application on drywall, concrete, and metal surfaces. The viscosity of this paint was ~7000 cP at 5 rpm, similar to materials used in the initial Phase II. This paint was scaled up and shipped to ORNL for their testing.

A Wagner Flexio 570 paint sprayer was used for manual spray application on a 4 ft long 2 ft wide dry wall panel. A spray booth was assembled from cardboard boxes to contain the spray. Spraying was done from a distance of 1 ft from the surface to have a good spread of the paint and achieve full coverage. Figure 6a shows an ISL engineer spraying the paint on the 2 ft x 4 ft drywall surface. The middle panel (b) shows the fully coated panel while the right panel (c) shows a close up of the painted wall, showing some orange peel effect. The measured sag-free wet thickness of the sprayed paint (KTA wet film thickness gauge) was 24-mil, the target thickness was 30-mils.

To improve the sag resistance and spread of the paint for vertical spray application, ISL tested two additional commercially available thixotropic agents. To achieve a sag-free 30 mil wet coating, formulations with viscosities higher than the 050811MM-3111 batch were prepared by incorporating thixotropic additive 1. Additive 1 was readily dispersible in the paint formulations. The sag resistance and viscosities for various paint formulations are summarized in Table 3. At viscosities of ~15 kcP (5 rpm with spindle #2), sagging was eliminated to yield a uniform dried coating.

Table 3. Summary of Wet Thickness and Sag Versus Paint Viscosity

Paint Batch ID	Viscosity (kcP)	Wet Thickness (mil)	Sag Free	Notes
100218RG3111-1	11.42	29	No	Large Sag Ridge
100218RG3111-2	12.33	30	No	Medium Sag Ridge
101218RG3111-3	14.63	30	Yes	Wave-like features
101218RG3111-4	14.72	30	Yes	No features

4.2.6 *Manual application of paints on different substrates*

As part of the paint (050811-MM-3111) evaluation and testing at ORNL, ISL developed protocols to manually apply Trap & See™-Hg paint on a variety of substrate materials and geometries (similar to the debris samples from Y-12 NSC) and peel them when dry. Test substrates included planar sheet metal, J-connectors and steel pipes in addition to the soil, concrete, drywall and rubble typical of Hg-contaminated waste. If the test surface was contaminated with mercury, the paint application process was carried out in ventilated fume hood. Personal protective equipment such as lab coat or overalls, safety glasses and gloves are also recommended. For manual application 1" and 2" foam brushes, a transfer container with a mouth/opening wider than the width of the foam brush are used. Before coating, the paint is shaken 5 minutes manually or 1 minute in a paint shaker.

The paint was applied by using foam brushed onto the following substrates using a one stroke/dip technique. For curved surfaces such as the J-connector and steel rod, the first coat is applied by painting the surface, and subsequent layers by a tapping technique using the foam brush. A 15 to 30 minute wait time is recommended for the paint to dry before applying additional coats to increase the thickness.

- 5" x 7" sheet metal steel
- J-connector
- Steel pipe
- Cement brick

The coated parts were allowed to dry for 3-4 days under room ambient in order to peel from the surface. The coating thickness of film peel from the steel substrate was ~6-mil for one coat, ~17-mil for the 3-layer coating on the J-connector and ~16.5-mil for the coating on the steel rod surface. Figure 7 shows steel sheet before coating, after coating, peeling the paint, and peeled coating and bare substrate. Figure 8 shows the application of coating to a J-connector at various stages and the peeled coating while Figure 9 shows similar images for coatings on a steel pipe.

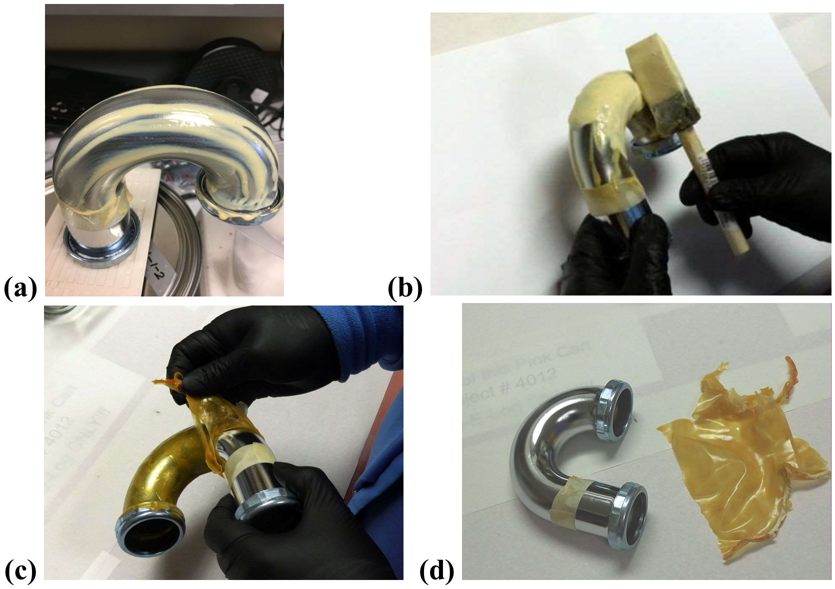

***Figure 8. (a)** J-connector after 1st coating, **(b)** paint tapping process, **(c)** peeling off coating, and **(d)** peeled off paint and steel sheet.*

***Figure 9**. **(a)** Steel pipe after 1st coating, **(b)** after coating has dried, **(c)** peeling coating, and **(d)** peeled off paint and steel pipe.*

Figure 10 shows the application of the paint to a cement brick. The paint is applied to the top and four sides initially and dried for an hour prior to application of a second coat. For a peelable coating of ~2-mil dry thickness from this rough substrate, six coats were required. The coatings were tack free after 2 days and fully cured and peelable after 4 days of ambient drying. The paint tested here was a low viscosity (~7000 cP), hence the need for additional coats. It was also the paint that was shipped and used in testing at ORNL. Later paint formulations for spray application had a higher viscosity and could achieve the target thickness in one spray application.

***Figure 10**. **(a)** Cement brick after 1st coating, **(b)** dried 4-layer coating, and **(c)** peeling a thin coating (13-mil) that breaks, recommend thicker coatings by increasing number of layers to 6, targeting ~20-mils.*

4.2.7 *Ex-situ analysis of low to high hg-containing samples from the Y-12 site*

Due to the challenges of accessing the Y-12 NSC site, the site contractor provided the debris materials (4-inch by 1-foot long concrete and cut metal plates from equipment) from a demolition site to the ORNL team. A Research Safety Summary document was generated and approved at ORNL prior to commencing the work. A Senior Field Technician in the EM division took custody of the samples, characterized and tested them. All measurements and handling of Hg-contaminated samples were conducted in the presence of a safety specialist who recorded the Hg headspace readings around the work area. ISL provided the ORNL team with one gallon of paint, safety data sheet (SDS) and a protocol for manual and/or spray application of the paint.

ISL shipped a scaled-up paint batch based on 050818MM-3111 for testing at ORNL with Y-12 Samples. Briefly, Hg contaminated concrete and metal samples were placed inside buckets and capped. Hg headspace readings were obtained by sampling 100 or 200 µL of the headspace volume using a sampling syringe and analysis from a Lumex analyzer at 1 hour and 144 hours. The samples were coated with the paint and headspace readings were obtained at 24 hours and 144 hours. Part of the paints were removed 2 weeks after first applying the paint and Hg readings were taken of the partially bare samples at 24 hours and 72 hours. The samples were then stripped of all of the paint 10 days after the partial removal and headspace was taken at 144 hours. Table 4 summarizes the headspace Hg values for the concrete and two metal samples at the various time points before and after paint application and after partial as well as complete removal of the paint.

Table 4. Comparison of Hg Headspace Measurements from Y-12 Samples

		Concrete (Bucket #1)	Metal (Bucket #2)	Metal (Bucket #3)
		#1 total Hg	**#2 total Hg**	**#3 total Hg**
	Sample	in Headspace	in Headspace	in Headspace
Before paint	**time (h)**	ug/m^3	ug/m^3	ug/m^3
2:30-3:00pm	1	24.07	224400.00	25978.00
7/12/2018 3:00pm	144	107.00	12190.00	37850.00
After paint, 7/18/18				
7/19/2018 3:15pm	24	102.00	5260.00	2650.00
7/25/2018 3:00pm	144	43.30	619.50	401.50
Percent Change in Hg from 7/12/2018 Reading		-60%	-95%	-99%
After partial paint removal (2x2", 3 spots for the Concrete, and 2 spots for Metal #1 and #2), 7/30/18				
7/31/2018 3:00pm	24	18.79	2312.60	484.20
8/02/2018 3:30pm	72	18.55	1963.70	457.10
Percent Change in Hg from 7/12/2018 Reading		-83%	-84%	-99%
After ALL paint removal, 8/10/18				
8/16/2018 3:00pm	144	170.00	12840.00	5290.00
Percent Change in Hg from 7/12/2018 Reading		59%	5%	-86%

The headspace readings taken over Y-12 samples after the paints had dried indicate a significant reduction in Hg vapor formation. Partial paint stripping indicates a continued suppression in Hg vapor release although there is an increase in Hg due to the exposed surface. After complete removal of the paint and with an equilibration time of 6 days, Hg in headspace for the concrete sample was slightly higher than the initial reading. For one of the metal samples, the total Hg in the headspace was increased by ~10x from the painted readings but was still an 85% reduction in Hg compared to the untreated metal scrap. For the concrete sample, an increase in total Hg present in the headspace was observed. This may be due to a shifted equilibrium resulting from stripping off the coating from the surface. The newly generated low Hg concentration surface may enable release of mercury vapor trapped in interior pores of the concrete structure. Likewise, porous surfaces caused by rusting on metal surfaces can exhibit similar behavior, although to a lower extent since the porosity is limited to the surface. Residual Hg trapped below the rusted layers on the metal pieces could be contributing to the increased headspace measurements noted from one of the two metal samples.

The temporal data from Table 4 indicates that longer incubation periods of the surfaces with the paint formulation leads to increased sequestration from the headspace.

The peeled paints were also analyzed at ORNL for Hg content to determine the efficiency of the paint to sequester elemental Hg. The method of paint analysis uses standard protocols at ORNL. The results of the analyzed paints are summarized in Table 5.

Table 5. Comparison of Hg Captured in Peeled Paints from Y-12 Samples

Sample ID	Total Paint wt (g)	Hg removed by paint (ug/g)	Hg in residual (ug/g)	Final total Hg in peeled paint THg (ug/g)	Average THg (ug/g)	Total THg (mg)
Concrete area1-1A	79.28	9.276	3.174	12.45		
Concrete area2-2A		4.189	4.312	8.50		
Concrete area3-3A		4.946	11.501	16.45	**12.47**	**0.99**
Metal #1-1A	70.89	783.802	584.871	1368.67		
Metal #1-2A		612.506	492.662	1105.17	**1236.92**	**87.69**
Metal #2-1A	48.3	317.136	499.221	816.36		
Metal #2-2A		176.197	221.223	397.42	**606.89**	**29.31**

From the data in Table 5, we can conclude that the paints can sequester Hg from the Y-12 samples at capacities ranging from 12 µg/g from concrete up to 1.2 mg/g from a metal surface. Note the paint recovered 66 mg Hg/g of paint from a soil sample. The results to date suggest that Trap & See-Hg has demonstrated the following features:

- Ability to sequester very low levels (0.03 ug/g to 10 ug/g of Hg in the paint) of mercuric species (Hg(II) and Hg0) from brick rubble provided from the Y-12 site.
- Ability to trap and suppress Hg headspace from higher levels of Hg-contaminated waste (107 ug/m^3 to 37,850 ug/m^3) by about 60% on concrete and 95–99% on metal surfaces when paint is applied to the surface.
- Ability to sequester very large amounts of mercury in the paint ranging from 12 ug/g to 1.2 mg/g for a total Hg removal ranging from 0.99 mg to 87.69 mg. This would aid in classifying the waste for on-site or off-site disposal.

4.2.8 *Technology demonstration*

The team discussed the logistics of the technology demonstration of Trap & See paint with ICM's wall climbing robot. Potential sites for the technology demonstration were identified and shown in the photos in Figure 11, these include basements of buildings identified for demolition or repurposing. Figure 12 shows the exterior of other buildings that could also serve as potential sites for the technology demonstration. The site(s) could not be finalized due to the ongoing pandemic. However, the project team discussed the requirements for water, power and other logistics and provided a fact sheet to the Y-12 management including a safety manual for the operation of the robot.

Figure 11. *Photo images of the potential demonstration locations.* ***(A)*** *Building 7601 Basement, and* ***(B)*** *Building 7603 Basement1.*

Figure 12. *Photo images of the potential demonstration locations.*
(A) *Exterior of Building 7610a, and* ***(B)*** *an adjacent building.*

4.2.9 *Paint analysis for LDR considerations*

A modified TCLP test[21] was conducted at ORNL on the peeled paint samples from the three Y-12 samples tested at ORNL to assess the downstream hazardous waste disposal requirements. The total Hg in the paint samples were representative of low and high-Hg content. About 5 g/L of solids from each of the paint samples was used in the tests. Table 6 summarizes the three types of Y-12 samples tested, the % headspace reduction after paint application and % headspace Hg on test surface after removal of paint, the total Hg captured by the paint and the Total Hg leached from 5 g/L of solids from the TCLP test. Only the leachate from the peeled paint from the Metal#1 sample exceeded the EPA's Resource Conservation and Recovery Act (RCRA) toxicity characteristic limit of 0.2 mg/L of solids due to its high Hg content.

Table 6. Summary of Headspace Hg, Total Recovered Hg, and TCLP Tests for Y-12 Samples

Substrate Type	Headspace Hg Reduction after Paint Application	Headspace Hg After Paint Removal	Total Hg Captured in Paint	TCLP Tests Total Hg leached (5 g/L of solids)
Metal #1	-95%	+5%	1236 µg/g	377.25 (±5.67) µg/L
Metal #2	-99%	-86%	606.9 µg/g	126.81 (±0.67) µg/L
Concrete	-63%	+46%	12.47 µg/g	0.15 (±0.0008) µg/L

5.0 SUBJECT INVENTIONS

InnoSense LLC is the lead organization of this Phase IIB SBIR project and has been granted a U.S. Patent 11,021,615 entitled "Removable Smart Sequestration Coatings for Hazardous Metals" on June 1, 2021. No invention disclosures or patents were produced from this work at ORNL.

6.0 COMMERCIALIZATION POSSIBILITIES

InnoSense LLC, the lead organization of this project, is actively engaging stakeholders for commercialization possibilities. Since Hg-contamination is the major concern at the Y-12 NSC site, we focused outreach efforts on discussions and meetings with the OREM and site contractor, UCOR. ISL also attended the Waste Management Symposia in 2019 and 2021 (WM2021). At WM2021 ISL hosted a booth and also presented the work on the mercury remediation coatings titled "Reactive Strippable Coatings for Visualization and Remediation of Mercury Contaminated Surfaces – 21292" on March 12, 2021. This attracted additional visibility from large primes identified in Table 7 who expressed interest in Hg and other hazardous contaminants.

Table 7. Customers and Key Stakeholders Contacted by ISL

Company Name	Point of Contact
Amentum	Bill Badger, Business Development & Strategy Director bill.badger@amentum.com
Atkins Global	Brad Bowan, SVP, Engineering and Technology brad.bowan@atkinsglobalns.com
Huntington Ingalls Industries	Dave Carlson, Oak Ridge Capture Manager dave.carlson@hii-nss.com
Perma-Fix Environmental Services	Sally McQuaid, Assistant Manager, Technical Services, smcquaid@perma-fix.com
Oak Ridge National Laboratory	Mark Ford, Technical Support, Integrated Operations, Non Reactor Nuclear Facilities Division, fordmk@ornl.gov
Savannah River National Laboratory	Dr. Jen Wohlwend, Savannah River Site Technology Manager Jennifer.Wohlwend@srnl.doe.gov Dr. J. Connor Nicholson, Test Bed Director, National Advanced Proving Ground, Connor.Nicholson@srnl.doe.gov

7.0 CONCLUSIONS

Through collaboration between ISL and ORNL, we have successfully developed and demonstrated Trap & See-Hg as a reactive strippable coating for Hg decontamination & decommissioning (D&D) applications. It could potentially provide a sprayable solution designed to actively seal-in, suppress hazardous Hg vapors for worker safety, aid in visualizing hazardous materials and remediate the contaminated surface. The specific achievements are recapped here:

- Trap & See formulations were refined, down selected, and scaled up to 1 gallon and 5-gallon quantities.
- The reactive strippable coating was demonstrated with *ex-situ* samples for both visualization and fixing of elemental and mercuric Hg compounds reported to be present in the buildings and environments at the Y-12 NSC site.
- The paint was applied to metal, drywall, concrete, soil, plastic or other surfaces that are either smooth or rough. Paint removal is relatively easy from a smooth surface and requires moderate force to remove from a rough or highly jagged surface.
- The visible LOD ranges from 4 µg/cm^2 to 400 ng/cm^2 and sometimes as low as 250 ng/cm^2 depending on the substrate surface color and texture.
- The paint specifically developed to capture and seal in elemental mercury and Hg(II) compounds has demonstrated Hg vapor suppression efficiencies of 95–99% on metals and ~60% on concrete debris samples from the Y-12 site.
- The paints sequestered a significant amount Hg when peeled from various sample types (12 µg Hg/g of paint from concrete, 1.236 mg Hg/g of paint from metal and 66 mg Hg/kg of paint from soil) which was characterized by Total Hg analysis at ORNL.
- TCLP tests showed promise for on-site disposal with leachate values ranging from 0.15 (±0.0008) µg/L to 377.25 (±5.67) µg/L for a total of 5 g/L of solids tested. One leachate from the peeled

paint (Metal sample) exceeded the EPA's RCRA toxicity characteristic limit of 0.2 mg/L of solids due to its high Hg content.

- Commercialization efforts were carried out at ISL, paving the path towards scaling up, and beta test and validation with potential strategic partners.

These results demonstrate the potential for Trap & See coatings to be used in various D&D applications with mercury contamination and may provide environmental and health benefits to DOE, prime contractor, and the society in general.

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