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Bench Scale Study of Coating Formulations for Abatement of Mercury Vapor and Recovery of Mercury Spills

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SUMMARY

A sealant formulation has been developed that effectively mitigates human exposure to elemental mercury vapor evolving from spilled mercury. The sealant may be dispensed using a fogging methodology (for industrial settings) or a common spray bottle (for home or school settings). In parallel, the potential for Hg recovery using strippable coatings was evaluated.

Four liquid coatings were evaluated: one fogging fixative (FX2) developed at INL for the containment of surface and airborne contamination and three commercially available strippable decontamination gels (Carboset 441, Encor 449, and Stripcoat TLC Free).

Bench scale mercury vapor abatement tests were conducted in an enclosure using FX2 with the addition of a mercury absorbent and a mercury reactant. Testing with FX2 showed the reactant contributed significantly to reducing the rate of mercury evaporation. The reactant had a minimal impact on the mercury evaporation rate for the strippable coatings. The potential of strippable coatings to remove/recover mercury was inferred from the adhesive and cohesive performance of sprayable dilutions of the coatings applied to porous and non-porous surfaces.

Testing provided the following results:

- Mercury vapor concentration was reduced by a factor of 40 using the reactant enhanced FX2 solution, as well as with dilutions of Carboset and Encor. Dilute Stripcoat was less effective at reducing Hg evaporation.
- A 100× reduction in Hg evaporation rate was achieved using a coating of FX + the Hg reactant (a.k.a. FX Hg), compared to uncoated Hg.
- A 2× reduction in the Hg vapor accumulation rate was achieved using FX Hg, compared to the strippable coatings.
- Strippability and adhesion tests show that dilutions of Carboset and Stripcoat can be stripped from a variety of indoor surfaces. Dilute Encor was not strippable.

Results suggest FX Hg and dilute Carboset will likely reduce Hg vapor concentrations, in an indoor setting, to a level at or below the Occupational Safety and Health Administration 8-hour human exposure limit (0.1 mg/m^3). FX Hg dispensed via fogging would increase worker safety during remediation and demolition of industrial facilities with high levels of elemental mercury contamination. When applied via spray bottle, FX Hg may provide a safe and convenient method for the mitigation of Hg vapor in residential or commercial settings (e.g., offices, homes, schools). This approach would be most beneficial where recovery of the spilled Hg is impractical.

A sprayed then stripped application of dilute Carboset may provide a safe and convenient method for recovery of small Hg spills in residential and commercial settings. Further testing is required to determine if liquid Hg sufficiently adheres to the strippable coating to be recovered in this way. If it can be dispensed as a fog, dilute Carboset could be employed to recover Hg contamination in industrial settings. Testing of the foggability of dilute Carboset is required to assess this potential.

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1. INTRODUCTION

In the broadest sense, liquid mercury (Hg) contaminated sites present a hazard to both humans and the environment. There are 714 liquid Hg contaminated sites in the United States – 49% of the 1,467 hazardous waste sites on the U.S. Environmental Protection Agency's (EPA) National Priorities List.[1] The distribution of identified Hg contaminated sites is shown in Figure 1. Clearly, the extent of Hg contamination in the United States is large, and the need for remediation is considered a priority by the U.S. EPA.

In industrial, residential, and commercial settings, accidental Hg spills can produce concentrations of Hg vapor in indoor air that are poisonous to adults and can be lethal to children. The most recent estimate indicates that about 152,000 people are potentially exposed to Hg vapor in workplace environments in the United States.[1] Industrial exposure to Hg vapor occurs from Hg incorporated into manufacturing processes (such as the production of chlorine gas, Portland cement, caustic soda, sulfuric acid, automobile sensors and switches, liquid crystal displays, dry cell batteries, measurement and control devices, fluorescent and Hg vapor lamps, and disposal/recycling of Hg and Hg contaminated items).[2] In commercial and residential settings, adults and children are exposed to Hg vapor when Hg containing items (such as fluorescent lamps, blood pressure cuffs, barometers, and fever thermometers) are broken.

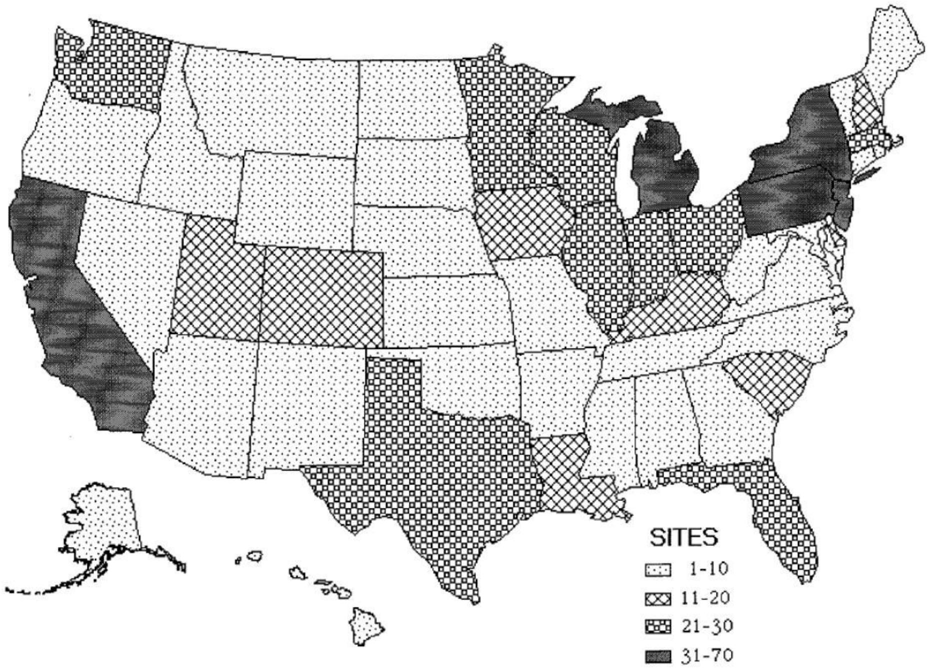

Figure 1. Frequency of elemental (liquid) Hg contamination at EPA National Priorities listed sites.

Kits are available for decontamination of small Hg spills in accessible locations (e.g., breakage of a compact fluorescent light bulb on nonporous flooring), and larger spills may be recovered by specially equipped workers using a sophisticated vacuum system. However, the use of vacuum recovery may increase the spread of Hg vapor, and not all spills are easily recoverable because of the materials and locations involved. For these reasons, the need exists for mitigation of human exposure to Hg vapor resulting from:

- Prolonged Hg recovery efforts,
- Containment of non-recoverable Hg spills,
- Contaminated facility demolition efforts.

In this study, the potential uses of sprayable/foggable coatings for Hg vapor abatement and/or Hg recovery were investigated as a cost-effective alternative to current decontamination methods.

Mercury in any of its three forms (i.e., elemental, inorganic, organic) is highly toxic to humans, affecting the neurologic, gastrointestinal, and renal organ systems.[1] The pathway of human exposure to Hg depends on which form of Hg is considered. This study considers the elemental (i.e., liquid) form of Hg and inhalation of Hg vapor, by liquid Hg evaporation, as the exposure pathway. The main objective of this study is to provide a basic formulation for sealants (i.e., coatings) that:

1. Effectively mitigates human exposure to Hg vapor, and
2. May be employed in industrial and commercial/residential settings, using a fogging methodology or a common spray bottle, respectively.

Because of Hg's relatively high vapor pressure (0.002 mm Hg at 25°C),[3] toxic levels of Hg vapor can accumulate rapidly in indoor air following a liquid Hg spill. Prompt action is necessary to prevent elevated vapor concentrations and prolonged human exposure following even a small Hg spill. Complete recovery of spilled Hg is desirable to avoid prolonged exposure to Hg vapor; however, Hg is heavy and small beads readily sink into floor cracks and disperse into inaccessible places. Therefore, recovery can be a challenge for even small Hg spills (e.g., the amount in one thermometer). When recovery efforts require prolonged worker exposure to Hg vapor, development of a coating and application methodology that minimizes Hg evaporation would be beneficial. If recovery is impractical due to location and/or the surface involved, use of coatings for in-place containment could provide a cost-effective alternative to recovery.

2. TESTING APPROACH

This study evaluates the effectiveness of three strippable coatings and one fixative coating for reduction of Hg evaporation under indoor conditions. The four coatings under evaluation include three commercial, strippable decontamination solutions (Carboset 441, Encor 449, and Stripcoat TLC Free) and INL's proprietary fogging fixative (FX2). The commercial coatings were formulated for recovery of loose contaminants from minimally porous surfaces by peeling the dried (i.e., cured) sealant as a cohesive film from surfaces, with contaminants adhering to the film. FX2 was developed for in-place containment of loose contaminants on surfaces, porous and nonporous.

The overall goal is to provide a formulation for coatings that can be applied in a fogging application at the industrial scale and in a hand-sprayer application in the home or school setting. The consistency of each strippable coating was adjusted (by water dilution) to be approximately compatible with the fogging process developed for FX2.[4] A second objective of this study was to evaluate the potential for liquid Hg recovery using strippable coatings, based on their adhesive and cohesive nature. Coating formulations that performed well relative to Hg vapor abatement were selected for additional formula optimization and fogging application studies.

The ability of dilute coating formulations to reduce Hg evaporation was evaluated by applying the coating to an aliquot of Hg in an enclosure. The subsequent equilibrium concentration of Hg vapor in the enclosure was used to indicate reduction of Hg evaporation relative to the vapor concentration achieved without a coating (i.e., the base case). Tests were conducted to establish performance of each coating without additives. Additional tests incorporating a Hg absorbent and/or Hg reactant were conducted. Equilibrium Hg vapor concentrations recorded for all coating formulations were compared to the Occupational Safety and Health Administration (OSHA) permissible exposure limit for Hg vapor in air (0.1 mg/m^3).[5]

Liquid Hg recovery was not quantified in this study. The potential for Hg recovery was based on the adhesive and cohesive nature of the three strippable coatings evaluated. Recovery potential is inferred from evaluations of the adhesion and strippability (i.e., the ease with which cured sealants are peeled by hand), relative to porous and nonporous construction materials. Recovery tests using small Hg beads consistent with a spill would be appropriate for the most promising coatings.

2.1 Experimental Setup

2.1.1 Mercury Vapor Capture

Figure 2 shows the experimental setup used in this study. The experimental enclosure consisted of a pyramid type glove bag (Captair, model 2200ANM) that has butyl rubber gloves, a port for gas sampling, and an approximate internal volume of 0.07 m3. The enclosure was placed in a fume hood for evacuation of Hg vapors between tests. A vinyl tube (0.3 cm ID) was routed through the gas sampling port with the open end suspended 20 cm above the Hg aliquot and the opposing end connected to an Hg vapor analyzer (described in Section 2.3). A consistent 0.2 mL aliquot (2.7 g, assuming Hg density of 13.53 g/ml) of liquid Hg (Sigma-Aldrich) was used for all tests. Aliquots were measured and dispensed using a 1 mL hypodermic syringe. Aliquots were placed in a plastic cup lined with Glad Cling Wrap (see the insets in Figure 2). A glass Petri dish was used for secondary containment. 0.5 mL of each coating was applied to Hg aliquots via a 1 mL syringe. 0.5 mL was sufficient to fully cover an aliquot.

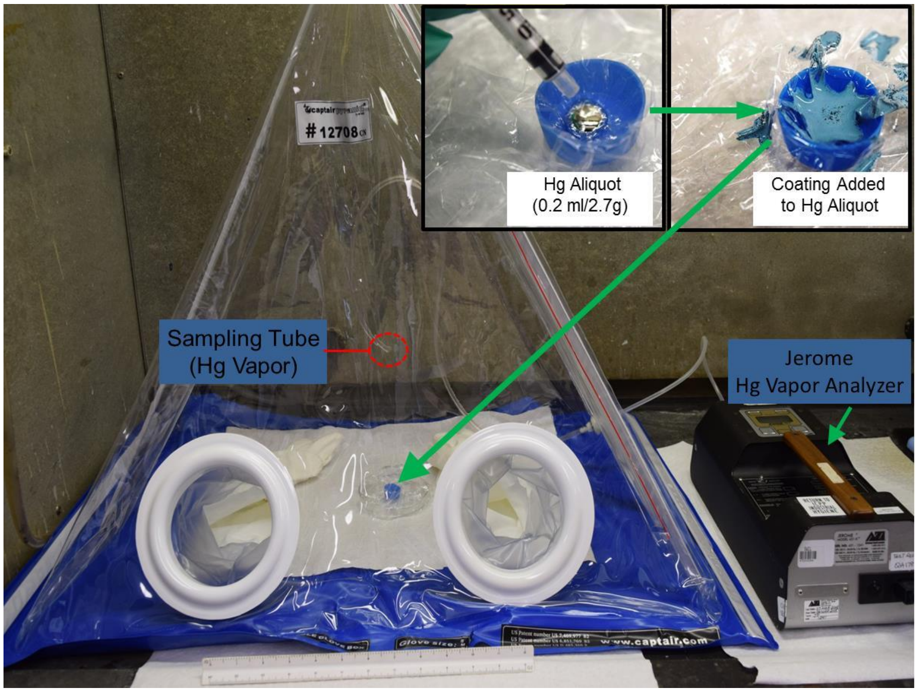

Figure 2. Experimental setup for Hg vapor abatement tests.

Vapor abatement tests were conducted at ambient laboratory temperature (16 to 26°C). Laboratory temperatures were documented using an ERTCO (Germany) model 1328 thermometer. Relative humidity in the laboratory was not documented, but a range of 16 to 80% (48% average) was recorded during the test period by the National Weather Service station at the Idaho Falls Regional Airport, approximately 52 km east of the laboratory.

2.1.2 Strippability Test Surfaces

The strippable coatings under evaluation were originally formulated for recovery of loose surface contaminants. The dried coating adheres to particles and peels from the surface as a cohesive film. It is acknowledged that adjusting the consistency of coatings (by water dilution) for use as a fogging agent may alter the adhesive and cohesive (i.e., strippable) characteristics of the coatings. As such, determination of adhesion and strippability, relative to porous and nonporous surfaces, was the first step taken in evaluation of their potential as a fogged coating for Hg removal.

Materials used for strippability evaluations include:

- Plastic weigh pans (4 × 4 in.), representing a smooth, nonporous surface
- Concrete coupons (6 × 6 in.), representing a flat, porous surface
- Granite coupons (6 × 6 in.), representing an uneven, porous surface

2.2 Evaluated Coatings and Additives

The strippable coatings were too viscous to be sprayed using a common hand operated spray bottle. Dilutions were made using 18 MΩ water. Sprayability was determined to be adequate when a mist visually comparable to that of FX2, with minimal splatter, was obtained using the spray bottle. Future viscosity testing and formula optimization are suggested for development of solutions as fogging agents.

The coatings used in this study are as follows:

1. FX2 – A solution of latex paint, glycerol, and sodium lauryl sulfate in water that was developed at INL as a non-strippable fogging agent for containment of surface and airborne contaminants. In these tests, FX2 was made with blue tinted paint to improve visibility. The FX2 consistency is optimized for fogging application; no additional dilution was required for these tests.
2. Stripcoat TLC Free (TLC Sanchem, Inc., Chicago, Illinois) – Stripcoat TLC Free is a yellow tinted strippable coating that is formulated for removal of radiological and chemical particles from indoor contaminated surfaces. A 33% water dilution (i.e., two parts Stripcoat to one part water) was used to make Stripcoat sprayable.
3. Carboset 441 (Lubrizol Corp., Sheffield Village, Ohio) – Carboset is a strippable coating that was formulated for protection of metal (i.e., nonporous) surfaces. A 20% water dilution (i.e., four parts Carboset to one part water) was used to make Carboset sprayable.
4. Encor 449 (Arkema, Inc., King of Prussia, Pennsylvania) – Encor is a strippable coating that was formulated for protection of metal (nonporous) surfaces. A 40% water dilution (i.e., three parts Encor to two parts water) was used to make Encor sprayable.

Chemicals were added to the coatings to evaluate their ability to absorb and/or react with the mercury. Mercury absorbers were added to the coating solutions to reduce Hg diffusion to the coating’s free surface (i.e., the coating-atmosphere interface). Mercury reactants were added to the coating solutions to react with Hg to form a stable compound that does not evolve mercury vapor, thereby reducing the coating’s permeability and Hg vapor diffusion to the coating’s free surface. The solid additives were slowly stirred into solution, then vigorously stirred for 1 minute.

Absorbent and reactive additives tested:

- Additive 1 – Mercury reacting chemical, ≤ 150 µm particle size, ASTM #100 sieve.
- Additive 2 – Mercury absorbing chemical, sieved to a ≤ 25 µm particle size using an ASTM #500 sieve.
- Additive 3 – A combination of additives 1 and 2.
- Additive 4 – Mercury absorbing and reacting compound.

- Additive 5 – Commercial mercury absorbing and reacting compound, sieved to a ≤ 25 µm particle size using an ASTM #500 sieve.

Additive concentration was held constant at 0.035 g/mL (3.5 g per 100 mL solution). This concentration was selected based on observation of a concentration in solution that was sprayable as a relatively fine mist without plugging the spray nozzle. Additive 3 is the exception. Being a combination of additives 1 and 2, the concentration of each chemical was maintained at 0.035 mg/L, thus a total of 7.0 g of solids were added per 100 mL solution (7.0 wt. % solid addition). These subjective amounts of solid additions form the basis for formula optimization tests. Based on results from initial evaluations with FX2, additive 1 was selected for evaluation with the strippable coatings. Table 1 is a matrix of coating solutions evaluated in this study.

Table 1. Matrix of evaluated coatings and additives.

	Additives					
Coating	**No Additive**	**1**	**2**	**3**	**4**	**5**
FX2	X	X	X	X	X	X
Carboset	X	X	—	—	—	—
Encor	X	X	—	—	—	—
TLC	X	X	—	—	—	—

2.3 Measurement of Vapor Concentration

Evaluations of Hg vapor abatement were conducted in an enclosure. Evaporation in such an enclosure is expected to be an equilibrium process that can be defined simply by the ideal gas law for a unit volume:

$$N = \frac{P}{RT}$$

where:

P = Hg vapor pressure, 0.002 mm Hg at 25°C (2.632E−06 atm)

R = Universal gas constant, 0.08206 L·atm/K·mol

T = Temperature in Kelvin, 298.15°C (25°C)

N = moles/L

This relation yields a calculated equilibrium Hg vapor concentration of 21.6 mg/m^3 (2.6 ppm), using the molar mass of Hg (200.59 g/mol) for conversion of N from moles/L to mg/m^3.

Measurements of vapor concentration were performed using a Jerome 431-X portable Hg vapor analyzer (visible on the right side of Figure 2). The Jerome analyzer utilizes adsorption of Hg onto an internal gold foil, which alters the foil's electrical resistance relative to a reference gold foil. Prior to each day's measurements, adsorbed Hg was removed from the gold foil using an automatic regeneration function (i.e., electric heating of the foil) and the instrument zero was checked (ambient laboratory air was used as a zero reference). Additionally, prior to measurements, the enclosure's gloves were waved around for 30 seconds to eliminate potential sampling error caused by stratification of Hg vapor. The instrument has an accuracy of 65% at 100 mg/m^3 Hg and precision of 5%.[6] When vapor concentrations exceeded the instrument saturation limit, a 1 L gas syringe (Trajan Scientific Americas, Inc., Austin, Texas) was used to draw a gas sample from the enclosure via the gas port. Sample dilution was performed by drawing laboratory air into the syringe until a total volume of 1 L was contained. The dilute gas sample was injected into an evacuated 3 L Tedlar bag (CEL Scientific Corp., Santa Fe Springs, California), which was subsequently connected to the vapor analyzer for measurement of the dilute vapor concentration. Reported concentrations are adjusted for the dilution.

Calculated and measured equilibrium Hg vapor concentrations were in disagreement for uncoated Hg aliquots. Measured equilibrium concentrations were approximately 10× lower than the calculated equilibrium concentration of 21.6 mg/m^3. The source of this disagreement was not determined, but is believed to be related to a scaling coefficient programmed into the analyzer's software. Because relative, not absolute, comparisons are needed to evaluate coating performance, the accuracy of concentration measurements is not critical to the purpose of this study. For the purpose of presenting vapor concentration data on the most likely (and most conservative, relative to meeting regulatory limits) concentration scale, all measured vapor concentrations reported were multiplied by 10.

NOTE: *Measurement precision was confirmed by two base case replicate tests that are not presented here.*

3. RESULTS AND DISCUSSION

3.1 Mercury Evaporation Reduction

3.1.1 FX2 Evaluation

This study first evaluated FX2 with and without additives. Figure 3 shows Hg vapor concentrations in the enclosure for the base case (i.e., no coating) and the various formulations of FX2. The coatings demonstrated three advantages:

1. A reduction in the rate at which vapor concentration increases prior to achieving equilibrium in the enclosure (i.e., a more gentle slope on the initial portion of the vapor concentration curve);
2. A decline in the equilibrium concentration over time (i.e., the uncoated sample curve is flat once equilibrium is achieved, whereas the coated samples' curves show a gradual decline in equilibrium vapor concentration);
3. A reduction in the total mass of Hg evaporated, which is indicated by lower maximum vapor concentrations measured for the coated samples.

The inset in Figure 3 magnifies the lower portion of the graph, showing Hg vapor concentrations without the base case, in order to accentuate differences between the various FX2 formulations (see the Appendix for the complete datasets used to generate Figure 3).

Maximum vapor concentrations occurred within 24 hours for the base case and within approximately 24 to 48 hours for the various FX2 formulations. Relative to the base case, use of coatings resulted in a decrease in the rate of Hg vapor concentration increase. The evaporation rate (i.e., the rate of vapor diffusion across a coating's free surface) was calculated by fitting a linear expression to the first 24 hours of concentration measurements. Table 2 shows the calculated vapor diffusion rates (in mg/m^3/hr) for each test series. The diffusion rate for FX2 + Additive 1 (0.009 mg/m^3/hr) was 100× lower than that of the base case (0.915 mg/m^3/hr) and significantly lower than that of FX2 with other additives. The calculated diffusion rate for the base case is conservative. The R^2 value of 0.748 indicates a relatively poor fit. Based on the base case curve shown in Figure 3, fitting a linear expression to the first 12 hours of data would achieve a better fit and a considerably higher value for the diffusion rate.

Coating application resulted in maximum vapor concentrations that decline with time, unlike the base case concentration, which was relatively constant with time. For the base case, unabated Hg evaporation resulted in a rate of vapor replenishment that was greater than enclosure leakage. The coatings diminished Hg evaporation to a rate less than the rate of enclosure leakage.

The inset in Figure 3 shows FX2 only and FX2 + Additive 2 performed similarly in reducing Hg evaporation; however, greater reductions were achieved when additive 1 was added to the recipe (i.e., additive 3 is additive 1 + additive 2). This outcome reveals the minimal contribution provided by additive 2 for the reduction of vapor diffusion through the coating. The figure also shows a reduction in maximum vapor concentration > 40× for the other four additives (< 0.5 mg/m^3), compared to the base case (> 21 mg/m^3). Based on the irrelevance of additive 2, a Hg absorber, and the importance of additive 1, a Hg reactant, to lower vapor concentration, additive 1 was selected as the best additive and the one to be included in the evaluation of strippable coatings. Additives 3, 4, and 5 were different combinations of Hg absorber and Hg reactant.

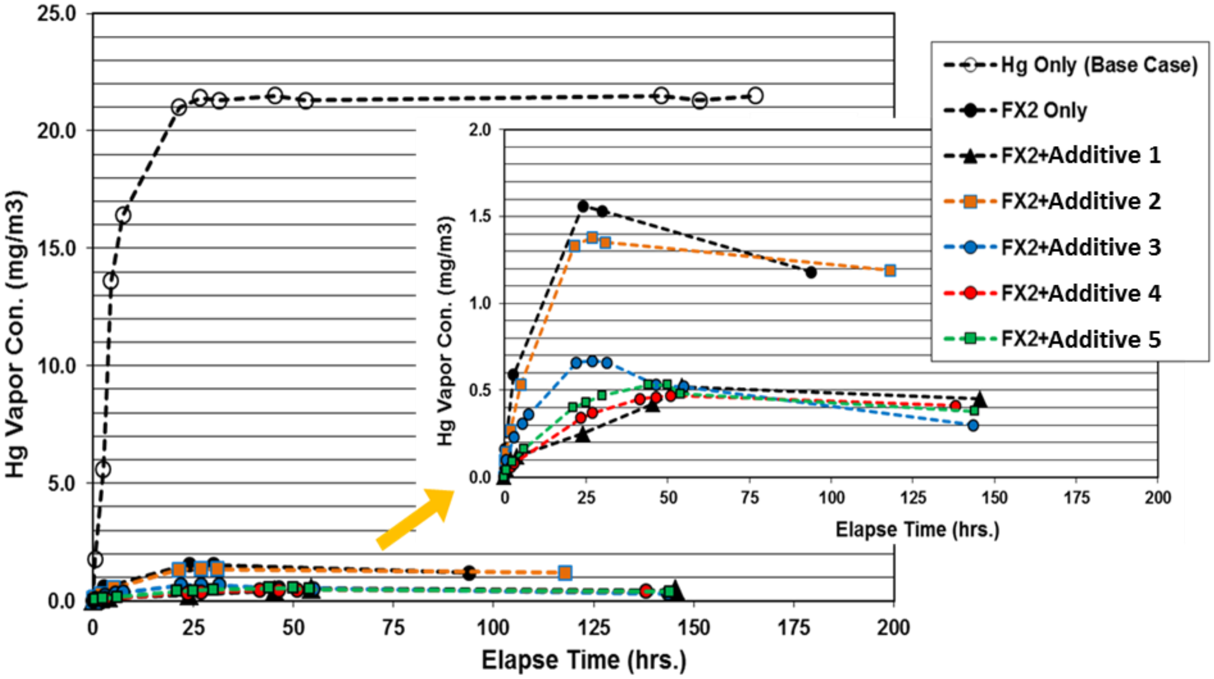

Figure 3. Evaluation of Hg vapor reduction for various FX2 formulations, including the uncoated, Hg-only base case. The inset shows a magnification of the lower portion of the main graph, omitting the uncoated base case in order to accentuate differences between the various FX2 formulations.

Table 2. Hg vapor diffusion rates for FX2 additives over the first 24 hours after coating.

	FX2 Additive						
	Hg Only (Base Case)	**FX2 Only (No Additive)**	**Additive 1**	**Additive 2**	**Additive 3**	**Additive 4**	**Additive 5**
Rate (mg/m^3/hr)	0.915	0.061	0.009	0.058	0.027	0.014	0.017
R$^{2(a)}$	0.748	0.935	0.862	0.974	0.922	0.987	0.985

a. R^2 indicates the quality of the linear fit. The closer the value is to 1, the better the curve fits the data.

Figure 4 shows Hg aliquots coated with FX2 (at 94 hours) and FX2 + Additive 1 (at 150 hours). (Note: Cracks visible in the FX2 coating [left pane of Figure 4] occurred during handling and were not present during testing.) During drying, the FX2 + Additive 1 coating became black in color (right pane of Figure 4), as the additive reacted with Hg. Formation of a solid, non-reactive Hg phase in the coating further inhibited Hg vapor diffusion through the coating, as indicated by the relatively low vapor diffusion rate of FX2 + Additive 1 (see Table 2).

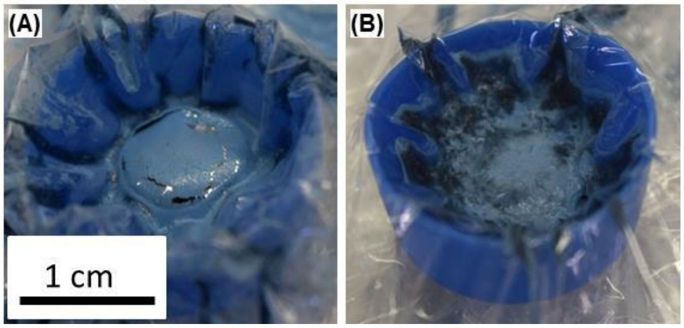

Figure 4. Final images of (A) FX2 only coating and (B) FX2 + Additive 1 (a.k.a. FX Hg) coating.

3.1.2 Strippable Coating Evaluations

Following tests with FX2, three strippable coatings (Carboset 441, Encor 449, and Stripcoat TLC Free) were evaluated. Figure 5 shows the evolution of Hg vapor concentrations in the enclosure for strippable coatings with and without the mercury reactant. Results for FX2 and FX2 + Additive 1 (a.k.a. FX Hg) are shown for comparison. (For the complete datasets used to generate Figure 5, see the Appendix.) Two primary results are evident from Figure 5 and Table 3:

1. Hg evaporation rates for Carboset and Encor are similar and clearly outperform Stripcoat TLC;
2. Hg vapor diffusion rates across the three strippable coatings were comparable and roughly 2× that of FX Hg (see Table 3).

Carboset and Encor formulations achieved maximum Hg vapor concentration (~0.5 mg/m^3) within approximately 24 hours. The maximum vapor concentration measured for Stripcoat was ~1.2 mg/m^3 at ~76 hours. Application of Stripcoat affected a 20× reduction of maximum vapor concentration.

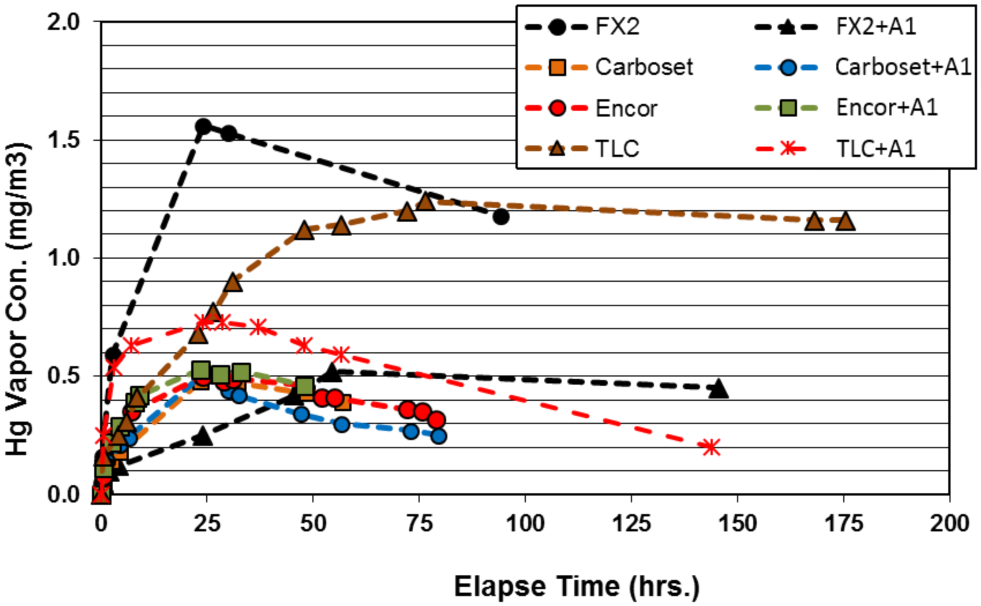

Figure 5. Hg vapor reduction for evaluated commercial coatings.

Table 3 shows vapor diffusion rates calculated by fitting a linear expression to the first 24 hours of vapor concentration measurements. Note the unreliable fit for the TLC + Additive 1 data due to the sharp rise and then round off of that curve in the first ~12 hours. Rates calculated for FX2 and FX Hg are repeated from Table 2 for comparison.

Table 3. Strippable coatings' Hg vapor diffusion rates over the first 24 hours after coating.

	Coating							
	FX2	**FX Hg**	**Carboset**	**Carboset+A1**	**Encor**	**Encor+A1**	**TLC**	**TLC+A1**
Rate (mg/m^3/hr)	0.061	0.009	0.019	0.019	0.018	0.020	0.026	0.022
R^2	0.935	0.862	0.953	0.916	0.781	0.756	0.914	0.549

Unlike FX2, the addition of the Hg reactant to Carboset and Encor contributed negligibly to the reduction of Hg evaporation, as indicated by the consistent ~0.5 mg/m^3 maximum measured vapor concentrations with and without the additive. The additive improves Stripcoat performance markedly, but the maximum vapor concentration remains larger than for Carboset and Encor. Figure 6 shows Hg aliquots coated with Carboset (after 57 hours), Carboset + Additive 1 (after 80 hours), Encor (after 79 hours), and Encor + Additive 1 (after 57 hours). Consistent with what was observed with FX Hg, strippable coatings containing the reactant became black in color (Figures 6b and 6d) as the Hg stabilization reaction occurred. However, the rate of diffusion reduction achieved in FX Hg was not achieved in Carboset and Encor. This outcome is likely due to the longer reaction time provided by FX Hg's ~48 hour drying time, compared to Carboset's and Encor's ~24 hour drying time.

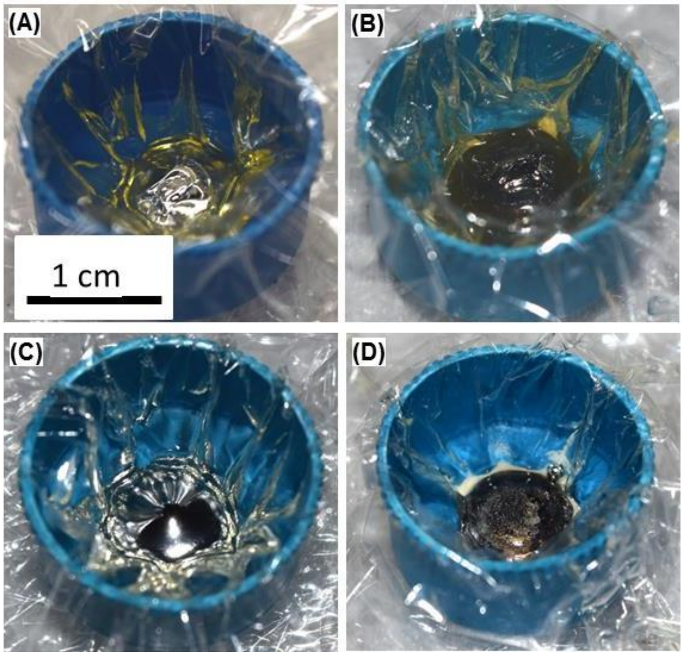

Figure 6. Hg aliquots coated with (A) Carboset, (B) Carboset + Additive 1, (C) Encor, and (D) Encor + Additive 1, after 57, 80, 79, and 57 hours drying time, respectively.

Although the maximum vapor concentrations measured for strippable coatings and FX Hg are consistent (~0.5 mg/m^3), the vapor diffusion rate of strippable coatings (0.02 mg/m^3/hr on average) is 2× greater than that of FX Hg (0.009 mg/m^3/hr). Following the initial use of an Hg vapor abatement coating in an indoor setting, knowledge of the vapor diffusion rate, as it relates to vapor accumulation, can provide guidance for follow-on actions. These actions may include planning of remediation efforts (e.g., ventilation rates, filter capacities, and personal protective equipment requirements) or determining the amount of time required for attenuation to reduce human exposure to a level permissible under regulatory laws. Currently, the OSHA limit for human exposure to airborne Hg is 0.1 mg/m^3 over a weighted 8 hour period. Given a measurement period longer than the 3 to 4 days considered in this study, the 0.5 mg/m^3 maximum vapor concentrations measured for the best performers in this study (FX Hg, Carboset 441, and Encor 449) would have most likely declined, due to enclosure leakage, to a level consistent with the OSHA limit (0.1 mg/m^3). Further, vapor concentrations may have continued to decline to a level consistent with the more stringent 0.025 mg/m^3 8-hour exposure limit of the American Conference of Governmental Industrial Hygienists.

Based on the comparable 40× reduction in Hg evaporation of FX Hg, Carboset, and Encor, and on the 2× lower vapor diffusion rate of FX Hg, this study found the basic formulation of FX Hg to be the best coating for Hg vapor abatement. Because of their comparable Hg evaporation reductions, optimization of dilute Carboset and Encor for fogging application and strippability is suggested. Evaluation of strippability is presented in the following subsection.

3.2 Coating Strippability and Adhesion

Student interns Jesse Viera and Janesler Gonzalez from FIU’s DOE Fellows Program conducted tests on strippable coatings under consideration for Hg recovery. Water dilutions of Carboset, Encor, and Stripcoat were prepared (see Section 2.2) with no additional additives and applied, using a common spray bottle, to porous and nonporous coupons (see Section 2.1.2). Except on the nonporous surface, dilute coatings were applied in multiple layers per manufacturer recommendations. One thick application was applied to the nonporous surface (i.e., a plastic weighing pan). Because of surface porosity and subsequent fluid absorption, 15 layers of dilute Carboset and Encor were applied to the porous surface samples (i.e., granite and concrete) in order to obtain a thick and potentially strippable coating. Dilute Stripcoat frequently plugged the spray nozzle; therefore, a spatula was used to spread the bulk of the dilute Stripcoat onto the porous samples. It was noted that dilute Stripcoat is much more adhesive (i.e., sticky) than dilute Carboset and Encor. Fogged application of Stripcoat may not be possible, even with additional dilution. Coatings applied to plastic pans were allowed to dry overnight. Coatings applied to granite and concrete were dry to the touch within 4 hours, aided by fluid absorption into the porous surfaces.

Carboset, Encor, and Stripcoat were easily removed (i.e., stripped) from the nonporous surface as single cohesive pieces. However, after drying for approximately 48 hours, the Encor piece that was stripped from the plastic pan became very brittle. A similar result was observed for Encor on porous surfaces. Figure 7a provides an example of good nonporous surface strippability recorded for dilute Carboset. Figure 7b shows the loss of cohesion (i.e., friability) seen for Encor on a concrete coupon. Dilute Carboset (Figure 8) and Stripcoat (Figure 9) maintained strippability and adhesive properties throughout porous and nonporous surface evaluations. Retained cohesion indicates that Carboset and Stripcoat will be strippable from a variety of indoor surfaces. Their adhesive properties suggest they have potential to retain embedded Hg when stripped from surfaces.

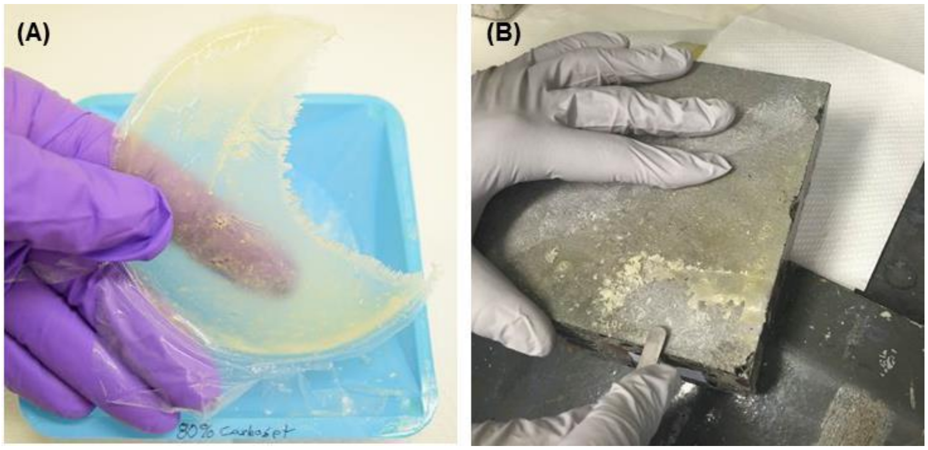

Figure 7. Examples of good and poor strippability performance: (A) good strippability of dilute Carboset from a nonporous, plastic surface, and (B) the loss of strippability of dilute Encor from a porous concrete surface. Note how the coating pictured in (B) crumbles instead of peeling as it is pried from the surface.

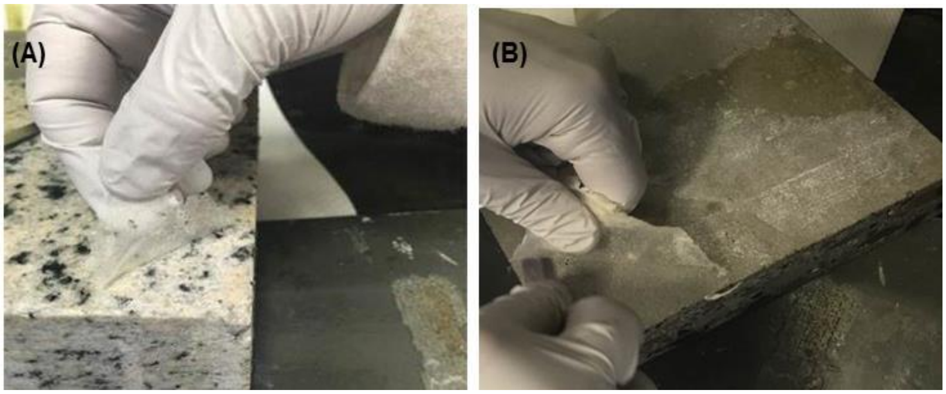

Figure 8. Examples of Carboset strippability from porous surfaces: (A) on an undulating granite surface and (B) on a smooth concrete surface.

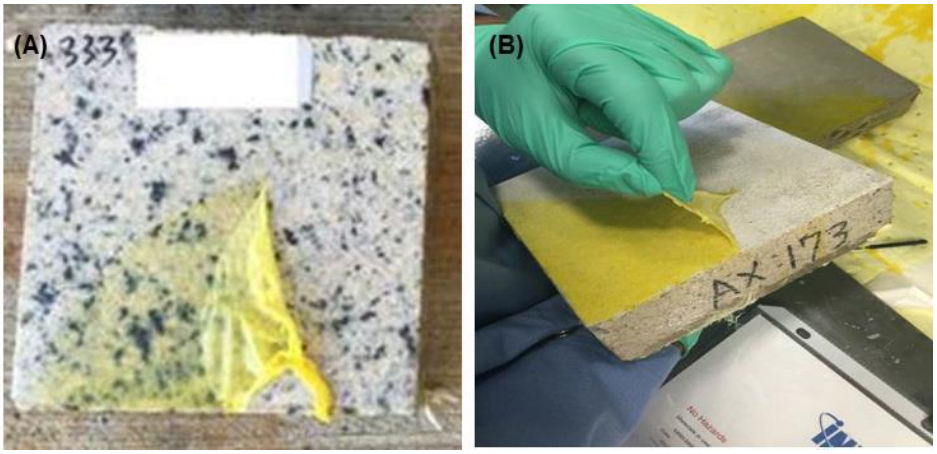

Figure 9. Examples of Stripcoat TLC Free strippability from porous surfaces: (A) on an undulating granite surface and (B) on a smooth concrete surface.

4. CONCLUSION

Coatings of Carboset, Encor, and FX Hg reduced Hg evaporation equally well. The application of dilute Carboset, dilute Encor, or FX Hg effected a > 40× reduction of maximum vapor concentration in the enclosure (from > 21 mg/m^3 without coating to < 0.5 mg/m^3 with coating).

The time required for Carboset, Encor, and FX Hg coatings to cure was estimated to be the elapsed time between coating application and measurement of maximum vapor concentration. Cure times of ~24 hours for Carboset and Encor and ~48 hours for FX Hg were reported. Rudimentary calculations of Hg vapor diffusion rates through the coatings suggest that the additional cure time required for FX Hg prolonged the intended Hg stabilization reaction, resulting in an FX Hg coating that was less permeable to Hg vapor than the commercial coatings. The FX Hg mercury vapor diffusion rate (0.009 $mg/m^3/hr$) was 2× lower than the rates of Carboset and Encor (~0.019 $mg/m^3/hr$) and was 100× less than the rate for uncoated Hg (0.915 $mg/m^3/hr$). FX2 has been extensively tested as a foggable agent for contamination control. As such FX Hg is the most promising option for use a Hg fixative.

Regarding Hg recovery, dilute Carboset and Stripcoat maintained their cohesive and adhesive properties on porous and nonporous surfaces. Retained adhesion suggests Carboset and Stripcoat stripped from surfaces may retain embedded Hg. Dilute Stripcoat proved difficult to dispense from a spray bottle, suggesting it may be difficult to apply via fogging or direct spraying. Further, Stripcoat did not perform as well as Carboset and Encor in reducing Hg evaporation. Dilute Carboset, on the other hand, is readily sprayable and significantly reduces Hg vapor concentration. Dilute Carboset is a good candidate for development as a foggable solution for Hg recovery.

5. REFERENCES

[1] Toxicological Profile For Mercury, U.S. Department of Health and Human Services, Public Health Service, Agency for Toxic Substances and Disease Registry, March 1999.

[2] Sources of Mercury in Industrial Facilities, http://www.glrppr.org/docs/mercury_in_industry.htm, accessed July 2015.

[3] Health Effects Notebook for Hazardous Air Pollutants – Mercury Compounds, https://www.epa.gov/sites/production/files/2016-09/documents/mercury-compounds.pdf, accessed February 2018.

[4] R.L. Demmer, D.T. Fox, and K.E. Archibald, “Pilot Scale Advanced Fogging Demonstration,” INL/EXT-14-34022, January 2015.

[5] 29 CFR [Code of Federal Regulations] 1910.1000, revised 1 July 1999.

[6] Jerome 431-X Mercury Vapor Analyzer Operation Manual, Arizona Instrument LLC, Tempe, AZ, 2000. The Certificate of Instrument Calibration (ID 720924, 6/2/15) indicates a precision of +/- 5%.

APPENDIX

Test Data

Hg Only (base case)

Date	Time	Elapse Time (hrs)	Measured (mg Hg/m^3)	Dilution	Hg (mg/m^3)	Temp (°C)
1/27/2015	9:30	0	0.000	0	0.000	20.5
	10:00	0.5	1.76	0	1.760	20.5
	12:00	2.5	1.4	4	5.600	20.0
	14:00	4.5	1.36	10	13.600	20.0
	17:00	7.5	1.64	10	16.400	20.0
1/28/2015	7:00	21.5	2.10	10	21.000	20.0
	12:15	26.75	2.14	10	21.400	20.0
	17:00	31.5	2.13	10	21.300	20.0
1/29/2015	7:00	45.5	2.15	10	21.500	20.0
	14:30	53.0	2.13	10	21.300	19.5
2/2/2015	7:30	142.0	2.15	10	21.500	20.0
	17:00	151.5	2.13	10	21.300	20.0
2/3/2015	7:00	165.5	2.15	10	21.500	20.0

FX2 Only (no additives)

Date	Time	Elapse Time (hrs)	Measured (mg Hg/m^3)	Dilution	Hg (mg/m^3)	Temp (°C)
10/8/2014	10:45	0.0	0.000	0	0.000	18.0
10/8/2014	10:48	0.1	0.05	0	0.050	18.0
10/8/2014	11:00	0.3	0.16	0	0.160	18.0
10/8/2014	13:30	2.8	0.59	0	0.590	19.0
10/9/2014	10:45	24.0	1.56	0	1.560	19.5
10/9/2014	4:45	30.0	1.53	0	1.530	19.0
10/13/2014	8:45	94.0	1.18	0	1.180	18.5

FX2 + Additive 1

Date	Time	Elapse Time (hrs)	Measured (mg Hg/m^3)	Dilution	Hg (mg/m^3)	Temp (°C)
11/4/2014	10:30	0.0	0.000	0	0.000	19.5
	11:00	0.5	0.050	0	0.050	19.5
	12:30	2.0	0.100	0	0.100	19.0
	14:30	4.0	0.12	0	0.120	19.0
11/5/2014	10:30	24.0	0.250	0	0.250	20
11/6/2014	8:00	45.5	0.420	0	0.420	21.5
	17:00	54.5	0.520	0	0.520	21.0
11/10/2014	12:00	145.5	0.450	0	0.450	21

FX2 + Additive 2

Date	Time	Elapse Time (hrs)	Measured (mg Hg/m^3)	Dilution	Hg (mg/m^3)	Temp (°C)
10/15/2014	10:00	0.0	0.000	0	0.000	23.0
	10:05	0.1	0.100	0	0.100	23.0
	10:30	0.5	0.14	0	0.140	23.0
	12:00	2.0	0.27	0	0.270	24.5
	15:00	5.0	0.53	0	0.530	24.5
10/16/2014	7:30	21.5	1.33	0	1.330	23.5
	13:00	27.0	1.38	0	1.380	22.5
	17:00	31.0	1.35	0	1.350	24.5
10/20/2014	8:00	118.0	1.19	0	1.190	23.5

FX2 + Additive 3

Date	Time	Elapse Time (hrs)	Measured (mg Hg/m³)	Dilution	Hg (mg/m³)	Temp (°C)
11/11/2014	9:30	0.0	0.000	0	0.000	21.5
	10:00	0.5	0.100	0	0.100	23.0
	12:30	3.0	0.230	0	0.230	22.0
	15:00	5.5	0.310	0	0.310	21.0
	17:00	7.5	0.360	0	0.360	22.0
11/12/2014	7:30	22.0	0.660	0	0.660	24.5
	12:30	27.0	0.670	0	0.670	19.0
	17:00	31.5	0.660	0	0.660	18.5
11/13/2014	8:00	46.5	0.530	0	0.530	17.5
	4:30	55.0	0.520	0	0.520	16.5
11/17/2014	9:00	143.5	0.300	0	0.300	14.0

FX2 + Additive 4

Date	Time	Elapse Time (hrs)	Measured (mg Hg/m³)	Dilution	Hg (mg/m³)	Temp (°C)
11/18/2014	14:00	0.0	0.000	0	0.000	14.5
	16:00	2.0	0.060	0	0.060	15.5
	17:00	3.0	0.080	0	0.080	16.0
11/19/2014	13:30	23.5	0.340	0	0.340	17.0
	17:00	27.0	0.370	0	0.370	17.5
11/20/2014	7:30	41.5	0.450	0	0.450	18.0
	12:30	46.5	0.460	0	0.460	18.5
	17:00	51.0	0.470	0	0.470	19.5
11/24/2014	8:00	138.0	0.410	0	0.410	18.5

FX2 + Additive 5

Date	Time	Elapse Time (hrs)	Measured (mg Hg/m^3)	Dilution	Hg (mg/m^3)	Temp (°C)
12/2/2014	11:00	0.0	0.000	0	0.000	19.5
	11:30	0.5	0.040	0	0.040	19.5
	13:30	2.5	0.090	0	0.090	19.5
	17:00	6.0	0.160	0	0.160	20.3
12/3/2014	8:00	21.0	0.400	0	0.400	19.5
	12:00	25.0	0.430	0	0.430	20.0
	17:00	30.0	0.470	0	0.470	21.0
12/4/2014	7:00	44.0	0.530	0	0.530	21.0
	13:00	50.0	0.530	0	0.530	18.5
	17:00	54.0	0.480	0	0.480	18.5
12/8/2014	11:00	144.0	0.380	0	0.380	20.0

Carboset (no additives)

Date	Time	Elapse Time (hrs)	Measured (mg Hg/m^3)	Dilution	Hg (mg/m^3)	Temp (°C)
9/8/2015	8:00	0.0	0.000	0	0.000	16.0
	8:30	0.5	0.050	0	0.050	16.0
	10:30	2.5	0.140	0	0.140	17.5
	12:30	4.5	0.180	0	0.180	18.0
9/9/2015	7:30	23.5	0.480	0	0.480	18.5
	16:00	32.0	0.470	0	0.470	21.0
9/10/2015	8:30	48.5	0.430	0	0.430	19.5
	17:00	57.0	0.390	0	0.390	22.5

Carboset + Additive 1

Date	Time	Elapse Time (hrs)	Measured (mg Hg/m^3)	Dilution	Hg (mg/m^3)	Temp (°C)
9/14/2014	8:30	0.0	0.000	0	0.000	21.0
	9:00	0.5	0.040	0	0.040	21.0
	10:00	1.5	0.14	0	0.140	21.5
	13:00	4.5	0.21	0	0.210	25.5
	15:00	6.5	0.24	0	0.240	26.0
9/15/2015	8:00	23.5	0.5	0	0.500	23.0
	14:30	30.0	0.44	0	0.440	22.0
	17:00	32.5	0.42	0	0.420	23.0
9/16/2015	7:30	47.0	0.34	0	0.340	20.5
	17:00	56.5	0.3	0	0.300	20.0
9/17/2015	9:30	73.0	0.27	0	0.270	18.0
	16:00	79.5	0.25	0	0.250	19.0

Encor Only (no additives)

Date	Time	Elapse Time (hrs)	Measured (mg Hg/m^3)	Dilution	Hg (mg/m^3)	Temp (°C)
10/5/2015	10:00	0.0	0.000	0	0.000	15.5
	10:30	0.5	0.080	0	0.080	15.5
	12:30	2.5	0.240	0	0.240	16.0
	17:00	7.0	0.350	0	0.350	17.5
10/6/2015	10:00	24.0	0.500	0	0.500	15.5
	15:00	29.0	0.480	0	0.480	18.0
	17:00	31.0	0.490	0	0.490	18.5
10/7/2015	10:00	48.0	0.460	0	0.460	16.0
	14:00	52.0	0.410	0	0.410	17.5
	17:00	55.0	0.410	0	0.410	18.5
10/8/2015	10:00	72.0	0.360	0	0.360	16.5
	13:30	75.5	0.350	0	0.350	19.5
	17:00	79.0	0.320	0	0.320	20.5

Encor + Additive 1

Date	Time	Elapse Time (hrs)	Measured (mg Hg/m^3)	Dilution	Hg (mg/m^3)	Temp (°C)
9/28/2015	8:00	0.0	0.000	0	0.000	21.0
	8:30	0.5	0.110	0	0.110	21.0
	10:30	2.5	0.220	0	0.220	21.5
	12:30	4.5	0.290	0	0.290	22.0
	16:00	8.0	0.390	0	0.390	22.0
	17:00	9.0	0.420	0	0.420	22
9/29/2015	7:30	23.5	0.530	0	0.530	21.5
	12:00	28.0	0.510	0	0.510	21
	17:00	33.0	0.520	0	0.520	21.5
9/30/2015	8:00	48.0	0.460	0	0.460	21.5
	17:00	57.0	2.640	0	2.640	22.0

Stripcoat TLC Free Only (no additives)

Date	Time	Elapse Time (hrs)	Measured (mg Hg/m^3)	Dilution	Hg (mg/m^3)	Temp (°C)
10/12/2015	8:30	0.0	0.000	0	0.000	16.0
	9:00	0.5	0.160	0	0.160	16.0
	12:30	4.0	0.250	0	0.250	16.0
	14:30	6.0	0.310	0	0.310	21.5
	17:00	8.5	0.410	0	0.410	21.5
10/13/2015	7:30	23.0	0.680	0	0.680	21.5
	11:00	26.5	0.770	0	0.770	22.5
	15:30	31.0	0.900	0	0.900	18.0
10/14/2015	8:30	48.0	1.120	0	1.120	23.5
	17:00	56.5	1.140	0	1.140	19.0
10/15/2015	8:30	72.0	1.200	0	1.200	25.5
	13:00	76.5	1.240	0	1.240	20.5
10/19/2015	8:30	168.0	1.160	0	1.160	19.0
	16:00	175.5	1.160	0	1.160	20.0

Stripcoat TLC Free + Additive 1

Date	Time	Elapse Time (hrs)	Measured (mg Hg/m^3)	Dilution	Hg (mg/m^3)	Temp (°C)
10/20/2015	8:30	0.0	0.000	0	0.000	23.5
	9:00	0.5	0.250	0	0.250	22.0
	11:30	3.0	0.540	0	0.540	20.5
	15:30	7.0	0.630	0	0.630	19.5
10/21/2015	8:30	24.0	0.730	0	0.730	24.0
	13:00	28.5	0.730	0	0.730	24.5
	17:00	37.0	0.710	0	0.710	19.5
10/22/2015	8:30	48.0	0.630	0	0.630	25.5
	17:00	56.5	0.590	0	0.590	24.0
10/26/2015	8:30	144	0.200	0	0.200	23.0