DECONTAMINATING RADIOACTIVE LEAD SOLIDS

James N. Taylor
Jeffrey C. Bell
CWM Remedial Services, Inc.
ENRAC - Nuclear Waste Division
Columbia, South Carolina

William L. Matthews
Mason & Hanger-Silas Mason Co., Inc.
Pantex Plant
Amarillo, Texas

ABSTRACT

Lead has been and continues to be used extensively at nuclear facilities and DOE\DOD sites to shield workers from ionizing radiation. Because it is often used in highly contaminated areas, the lead itself often becomes radioactively contaminated, thus creating a Mixed Waste. If the lead is contaminated above specified limits, it must be decontaminated prior to release for unrestricted use. In most cases, where the lead cannot be decontaminated using conventional methods, the lead is stored until a viable decontamination method and/or disposal alternative is determined. At many facilities, large quantities of stored lead are creating a significant problem.

The U.S. EPA treatment standard for radioactive lead is, "Macroencapsulation with surface coating materials such as polymeric organics (e.g. resins and plastics) or with a jacket of inert inorganic materials to substantially reduce surface exposure to potential leaching media." Since the Toxicity Characteristic Leaching Procedure (TCLP) requires size reduction to less than 9.5 mm particles for solid waste, macroencapsulation is not a practical or economically feasible option for processing lead.

The U.S. EPA originally proposed "Surface Deactivation" as the treatment standard. Because there exists no demonstrated available technology, this method was dropped from the final treatment standard.

INTRODUCTION

Mason & Hanger-Silas Mason Co., Inc. contracted with Chemical Waste Management (CWM) Remedial Services, Inc. for surface decontamination and declassification of approximately 22,700 kg (50,000 pounds) of D008 radioactive lead solids in the form of weapons components at the Pantex Facility. The weapons components were surface-contaminated with depleted uranium (uranium-238) as a result of weapons disassembly operations at Pantex. The services requested by Mason & Hanger included decontamination of the lead to meet DOE Order 5400.5 release limits, stabilization of the secondary waste generated such that it passed the TCLP test for lead, and shearing of the decontaminated lead for declassification of shape.

The decontamination and stabilization processes were based upon a successful bench-scale demonstration completed at Pantex prior to full-scale operations. The decontamination process involves the use of a widely accepted acid etching technique for the dissolution of lead surface layers. The stabilization process involves solidification of the secondary mixed waste consisting of the decontamination acid, dissolved lead, and dissolved uranium-238 contamination. Solidification formulas based upon modified pozzuolanic chemistry were used to stabilize the secondary waste such that the final solidified product passed the TCLP test for leachable lead.

Following decontamination operations, the lead objects were surveyed to meet DOE criteria and then declassified utilizing an hydraulic shearing unit to cut the lead into unclassified shapes. The resulting lead pieces were packaged for transport to a lead recycling facility. The stabilized secondary waste was prepared for shipment to the approved federal radioactive waste disposal facility.

The successful completion of this project demonstrated that surface decontamination of lead can be cost-effectively performed such that:

- the decontaminated or "clean" lead can be reclaimed through recycling at a lead smelter for beneficial reuse.
- the secondary waste generated can be successfully stabilized to meet disposal requirements for burial as Low Level Radioactive Waste (LLRW) at a federal disposal site.

This report summarizes the activities involved in the successful completion of the decontamination and solidification work and presents the data generated during the project.

LEAD DECONTAMINATION

Scope Of Work

The original project scope of work consisted of decontamination of approximately 22,700 kg of lead in the following forms:

- 200 large classified cylindrical shapes of dimensions 60 cm X 60 cm X 90 cm
- 400 small classified hemispherical shapes of dimensions 60 cm X 60 cm X 30 cm
- 2 fiberboard drums of declassified shapes of dimensions 5 cm X 5 cm X 0.5 cm

Process Description

Decontamination was performed using a chemical stripping process to remove thin layers of the lead object's surface,

and the contamination along with the layer. The process uses an acid and hydrogen peroxide (H_2O_2) solution to dissolve the contaminated lead surface layers. This stripping process is commonly used in the metal finishing industry to dissolve lead coatings from copper alloys and steel.

Preparation

Once the lead objects were staged in the processing area, initial contamination and radiation surveys were conducted to obtain representative levels for fixed and removable alpha contamination.

In many cases some preliminary disfiguring had been performed on the lead objects in an attempt to partially declassify them. Many objects were twisted, smashed, crimped, etc. such that surveys of the contaminated interior surfaces were impossible. In order to adequately survey these objects and to ensure that all surfaces of the object would come into contact with the decontamination acid, the objects had to be pried open, cut, and flattened.

The reshaped lead was transferred from the staging area into the processing system or to the survey area. A forklift with lifting slings was used to transfer heavier objects into the decontamination system tanks.

Decontamination Operations

Following equipment setup and pre-operational testing to ensure proper radiological and industrial safety, the decontamination acid solution was prepared in one of the decontamination tanks. Lead components were then loaded into the empty decontamination tank. Decontamination acid solution was pumped into the empty decontamination tank until the lead objects were fully submersed in acid. After a predetermined decontamination period, the lead components were removed from the acid solution and placed in a rinse tank. The length of the decontamination period depended on several factors including the height of objects in the tank, pumping capacity, the concentration of lead in solution, and expected contamination levels. Figure 1 shows a basic flow diagram for the decontamination process.

Upon completion of the specified decontamination period the lead objects were rinsed with water to remove residual acid and lead from the surfaces. The rinse water was collected and regenerated to minimize secondary waste production. Lead objects, free of residual acid, were transferred to a clean area and allowed to dry. Once the components were dry, they were surveyed for contamination levels and either released for shearing or transferred back to the acid tanks for additional decontamination.

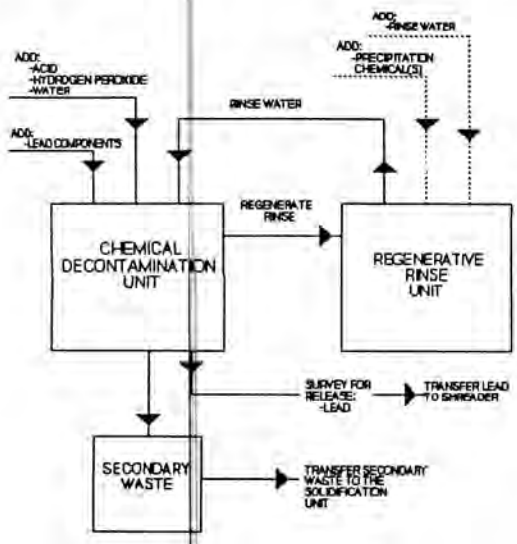

Fig. 1. Decontamination flow diagram.

All lead objects were surveyed as required by DOE Order 5400.5 to ensure that they met the specified release criteria for fixed and removable contamination. In cases where components did not meet the release survey criteria, the objects were returned to the decontamination tanks for re-processing. In a few cases (less than 1%), the contaminated lead surfaces were wire-brushed to remove surface contamination embedded below the depth removed by the chemical process.

Upon completion of decontamination and release surveys, lead components were transferred to the shearing area for declassification. Declassification of lead shapes was accomplished using a hydraulic shearing assembly to cut the objects into squares approximately 5 cm by 5 cm in size. Once the lead objects were sheared they were packaged in 208 L (55 gallon) drums. The packaged, declassified lead was stored for eventual shipment to a lead recycling facility.

Periodically during the decontamination process, as the concentration of lead in solution increased, samples of acid were drawn, solidified, and sent to an approved lab for TCLP analysis. These samples were generated to verify that the full-scale solidification product would pass the TCLP analysis for lead (Pb) and therefore meet the requirements for burial as Low Level Radioactive Waste (LLRW) at the federal disposal site.

Table I summarizes the average parameters measured for the decontamination process at Pantex.

RELEASE SURVEYS

Release Criteria

The release limits were based on DOE Order 5400.5 guidelines for alpha contamination due to the presence of depleted uranium (uranium-238) contamination. To ensure release limits were met and for ALARA considerations, release limits of 20% of the DOE Order 5400.5 for alpha contamination were utilized. These limits were as follows:

- Maximum fixed alpha contamination: $<1{,}000$ dpm/100 cm^2
- Maximum removable alpha contamination: <200 dpm/100 cm^2

In addition, Beta/Gamma contamination limits were established below the 5400.5 guidelines for conservatism.

TABLE I

Summary of Decontamination Data

Decon Period	17 min.
Weight	140 kg
Surface Area	54,515 cm^2
Removal Rate	0.84 μm/min
Depth Removed	14 μm

Surveys were performed for both Alpha and Beta/Gamma contamination on 100% of the surfaces, documented, reviewed and approved prior to release of the material for unrestricted use.

Representative Decontamination Factors

Table II is a summary of representative initial and final alpha contamination levels and the associated decontamination factors (DF's) which were obtained.

TABLE II

Contamination Levels and DF's

Fixed Contamination (dpm/100cm^2)	
Initial Levels	250 - 23,400
Final Levels	ND* - 500
Average DF	18
Removable Contamination (dpm/100cm^2)	
Initial Levels	4 - 5,000
Final Levels	ND* - 81
Average DF	54
* Non-Detectable	

SECONDARY WASTE STABILIZATION

Scope Of Work

The secondary mixed waste generated by the lead decontamination process consists of the acid solution, dissolved lead, and dissolved uranium-238 contamination. This secondary waste was solidified using a proprietary solidification formula which is based on modified pozzuolanic chemistry. (This formula is not cement-based.) The solidification formula was successful in stabilizing the secondary waste for lead such that a sample of the solidified waste form passed the toxicity characteristic leaching procedure (TCLP) for lead. Figure 2 provides a simplified flow diagram of the solidification process.

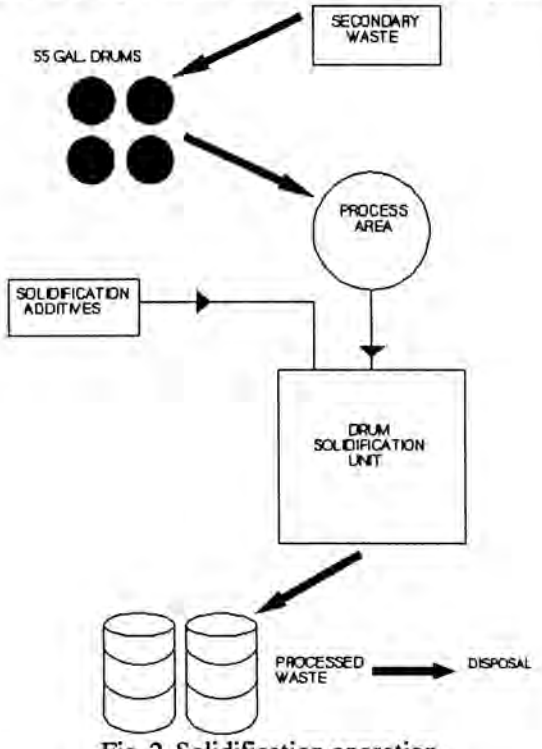

Fig. 2. Solidification operation.

Solidification Operations

As stated previously, samples of the acid were periodically drawn, stabilized, and analyzed for leachable lead to verify that the full-scale solidification product would meet the disposal requirements. These solidified samples of the secondary waste were prepared in accordance with NRC approved Process Control Procedures (PCP) and inspected for proper solidification characteristics (free-standing liquid, hardness, etc.). The concentration of lead in the acid solution was monitored closely throughout the project. The lead concentration was not allowed to exceed the maximum lead concentration in samples that had successfully passed TCLP for lead during treatability testing.

When the lead concentration reached the maximum allowable level, a sample of the acid was drawn, solidified in accordance with the project work plan and forwarded to the approved lab for TCLP analysis. The TCLP results for this process control procedure (PCP) sample were used as representative of the entire secondary waste batch. The secondary acid waste solution was pumped into 208 L (55 gallon) DOT 17H epoxy-coated steel drums lined with 60 mil rigid polyethylene liners in preparation for solidification. A new decon acid solution was mixed in the decontamination tanks so that decontamination of lead objects could continue in parallel with the solidification operation.

A specially designed and fabricated drum solidification unit was used to solidify the secondary waste in the 208 L drums. This unit consists of an electric lift, a fillhead port for addition of solidification agents, and a mixing unit. Each drum of secondary waste was solidified, labelled with a unique drum identification number, and temporarily stored.

Due to the use of the regenerative rinse unit, the volume of secondary waste was significantly reduced. Only three batches of secondary waste were produced during the project. The first two batches consisted only of the decon acid solution with dissolved lead and uranium-238 contamination. For the third and final secondary waste batch the rinse water and sludge from the Rinse and Precipitation system were mixed with the decon acid solution to re-dissolve lead that precipitated out of the rinse water. A PCP sample for each secondary waste batch (batches 1-3) was subjected to TCLP analysis and verified to pass prior to solidification of the entire waste batch. The average concentration of lead in solution prior to solidification was 30 g/l.

Volume of Secondary Waste

The original 22,700 kg (50,000 lbs) of lead objects were transported to the processing area on three flatbed trailers approximately forty feet long. The three trailers contained fifty (50) wire baskets (120 cm X 120 cm X 90 cm) containing the lead objects of various sizes. This equates to approximately 68 m^3 (2,400 ft^3) of initial waste volume. The decontamination process generated a total of twenty-one (21) 210 L drums of secondary waste. This equates to approximately 4.5 m^3 (158 ft^3) of final waste volume. Therefore, a volume reduction of approximately 93% was achieved through the decontamination process.

As demonstrated, approximately 0.9 m^3 (31.5 ft^3) of stabilized secondary waste was generated for each 4,540 kg (10,000 lb) of lead decontaminated.

The object's surface area directly corresponds to the quantity of secondary waste generated. The surface area per kilogram of lead decontaminated on this project was approximately 394 cm^2/kg.

TABLE III

TCLP Results

Acid Batch Number	TCLP Lead (mg/l)
1	0.09
2	0.17
3	0.40

SUMMARY OF TCLP ANALYSIS

Table III lists the results of the TCLP analyses performed on the solidification samples. As shown, the leachable lead level is well below the limit of 5.0 mg/l.

CONCLUSION

As demonstrated through the decontamination of 22,700 kg of lead at Pantex, surface deactivation of D008 radioactively contaminated lead solids can be successfully accomplished using existing, cost-effective technology. Additionally, the resultant secondary mixed waste can be stabilized to meet land disposal restrictions and qualify the waste for burial as LLRW. Finally, the decontaminated or "clean" lead can be released for unrestricted use or recycled.