BNL Building 650 Lead Decontamination and Treatment Feasibility Study

Final Report

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EXECUTIVE SUMMARY

Lead has been used extensively at Brookhaven National Laboratory (BNL) for radiation shielding in numerous reactor, accelerator and other research programs. A large inventory of excess lead (estimated at 410,000 kg) in many shapes and sizes is currently being stored. Due to its toxicity, lead and soluble lead compounds are considered hazardous waste by the Environmental Protection Agency. Through use at BNL, some of the lead has become radioactive, either by contamination of the surface or through activation by neutrons or deuterons. Regardless of whether the lead is contaminated or activated, the presence of radioactivity converts the material from a hazardous waste to a mixed waste which is considerably more complicated and expensive to dispose of. This study was conducted at BNL's Environmental and Waste Technology Center for the BNL Safety and Environmental Protection Division to evaluate feasibility of various treatment options for excess lead currently being stored. The objectives of this effort included investigating potential treatment methods by conducting a review of the literature, developing a means of screening lead waste to determine the radioactive characteristics, examining the feasibility of chemical and physical decontamination technologies, and demonstrating BNL polyethylene macro-encapsulation as a means of treating hazardous or mixed waste lead for disposal.

A review and evaluation of the literature indicated that a number of physical and chemical methods are available for decontamination of lead. Many of these techniques have been applied for this purpose with varying degrees of success. Methods that apply mechanical techniques are more appropriate for lead bricks and sheet which contain large smooth surfaces amenable to physical abrasion. Lead wool, turnings, and small irregularly shaped pieces would be treated more effectively by chemical decontamination techniques. Either dry abrasion or wet chemical methods result in production of a secondary mixed waste stream that requires treatment prior to disposal.

Evaluation of potential activation products and a radiological survey of random lead samples stored in Building 650 revealed that, in some cases, a unique gamma spectroscopy "fingerprint" can be used to identify lead that has been activated. For example, lead samples that contained isotopes of Cs, Sr, and Co (easily identified by gamma spectroscopy) are the result of contamination and thus, are subject to potential decontamination processing. Samples containing isotopes of the activation product Bi-207, however are poor candidates for potential decontamination. Based on preliminary findings, a gamma-scanning survey of lead would be useful for screening and identifying materials that are either clean, contaminated, or activated.

Results of bench-scale investigation of decontamination methods resulted in identification of several wet chemical methods that were very effective in removing layers of lead oxide from the surface, and thus would be likely candidates for decontamination processing. Most effective was a solution of hydrogen peroxide + 10% acetic acid solution. For all solutions tested, considerable improvement in removal efficiency was achieved using ultrasonic agitation. Attempts at dry abrasion decontamination were not successful using low-density polyethylene beads, but silica sand blasting was effective in removing lead oxide from the surface of bricks.

Polyethylene macroencapsulation was successfully demonstrated for the treatment of contaminated lead as an alternative to decontamination, or for cases where decontamination is either not technically or economically feasible. This process is being commercialized by Envirocare and will be available for disposal of contaminated DOE lead. BNL is currently considering shipping of 10,000 lbs of contaminated lead to Envirocare under this DOE sponsored initiative.

It is readily apparent from this evaluation that various wet chemical and dry abrasion can be used to remove radioactive contamination from lead, thus enabling potential recycling of the material or disposal as hazardous rather than mixed waste. However, the ultimate decision on whether or how to decontaminate lead currently stored at BNL or treat it for disposal will require consideration of process economics which were beyond the scope of this study.

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1. INTRODUCTION

Lead is one of the most effective materials for radiation shielding and has been used extensively at Brookhaven National Laboratory (BNL) in numerous reactor, accelerator and other research programs. A large inventory of excess lead (estimated at 410,000 kg) no longer being used has accumulated over many years and is currently being stored in four modular storage facilities adjacent to Building 650. Due to the diversity of applications, the lead is present in many shapes and sizes including thin strands of "lead wool," turnings and machine shop scraps, sheet stock and bricks. Lead and soluble lead compounds are considered hazardous waste (Toxicity Characteristic metals) by the Environmental Protection Agency under 40 CFR 268 because of their highly toxic properties.[1] Through use at BNL, some of the lead stored at Building 650 has become radioactive. The radioactivity is either a result of accumulation of radioactive particles on the surface through contact with radioactive liquids and solids (contamination) and/or a result of interaction between neutrons and other particles with the bulk lead, creating activation products. Regardless of whether the lead is contaminated or activated, the net result is conversion of a hazardous waste into a mixed waste (hazardous and radioactive) with the concomitant complexity and expense associated with disposal. Because bulk lead storage practices predate enactment of hazardous waste regulations, clean, contaminated, and activated lead are stored in a co-mingled fashion at Building 650, with no means of identifying radioactive characteristics.

This study was conducted at BNL's Environmental and Waste Technology Center for the BNL Safety and Environmental Protection Division to evaluate feasibility of various treatment options for excess lead currently being stored. The ultimate disposition of the lead will depend on whether the material is non-radioactive, contaminated, or activated. Non-radioactive material can either be recycled for reuse as shielding or other industrial applications such as battery manufacturing, or may be macro-encapsulated for disposal as a hazardous waste. Lead that is surface contaminated may potentially be decontaminated by chemical or physical means and then placed with the non-radioactive inventory for disposition. Activated lead would be much more difficult or impossible to decontaminate because of the extent and depth of penetration of radioactive isotopes within the bulk material. Activated lead would best be treated by macro-encapsulation for disposal as a mixed waste. Thus, the objectives of this effort included:

- investigating potential treatment methods by conducting a review of the literature,
- developing a means of screening lead waste to determine the radioactive characteristics,
- examining the feasibility of chemical and physical decontamination technologies, and
- demonstrating BNL polyethylene macro-encapsulation as a means of treating hazardous or mixed waste lead for disposal.

2. REVIEW OF POTENTIAL TREATMENT TECHNOLOGIES

The Environmental Protection Agency has identified numerous technology-based treatment standards known as Best Demonstrated Technologies (BDAT) for both toxicity characteristic (TC) and listed hazardous wastes.[2] By employing a recommended BDAT for a particular waste, the generator is not required to demonstrate compliance with performance-based test criteria, thus considerably simplifying disposal requirements. The BDAT for lead prior to land disposal is macroencapsulation with a surface coating material (e.g., plastic polymer) (see Table 3 of 40 CFR 268.42, waste code D008). However, land disposal is not the only option available in the case of radioactively contaminated lead. If the material can be successfully decontaminated, either totally or to an acceptably low level, the lead can be recycled into use. Activities in this area are already in place at other national laboratories, including the Idaho National Engineering Laboratory (INEL)[3], the Lawrence Livermore National Laboratory (LLNL)[4] and the Savannah River Site (SRS)[5].

Decontamination of materials and components can be accomplished in many ways and brief descriptions of these processes are readily available.[6,7] Those procedures that have been used specifically to decontaminate lead can be categorized roughly as either mechanical or chemical in nature. The former are usually based on the mechanical removal of surface contaminants using the forces generated by the impact of matter with the surfaces of the component. The latter rely on chemical action to remove the contaminants and include such methods as acid dissolution and foaming cleansers. There are also a few processes, such as metal refining and light ablation, that resist classification in either of the two categories. Summaries of many of these different procedures and their attributes can be found in.[3,5]

The following sections review the various techniques on an individual basis, assessing their effectiveness in removing or reducing the contamination and the minimization of secondary waste streams. At the same time, it should be recognized that optimum decontamination might be achieved through the employment of two or more of these methods in some cases. For completeness, a short section is included on macroencapsulation.

2.1 Mechanical Methods

The methods that fall under the category of mechanical decontamination can be subdivided into dry and wet types. The simplest forms of mechanical decontamination involve manual deployment of such implements as swabs, brushes and gritpaper, either in air or in the presence of a fluid medium. However, because of the high potential for spread of contamination and exposure of workers to radiation, these methods are used for removing only the very slightest amounts of contamination. The following subsections describe the types of systems most commonly encountered in decontaminating lead and other materials.

2.1.1 Dry Abrasive Blasting

The term "dry abrasive blasting" is used here to encompass all those processes in which hard particles are blasted via a compressed air jet at the surface to be decontaminated. Thus it includes shot blasting (which incorporates steel shot in the air jet), blasting with glass, sand, alumina, etc., blasting with ice and blasting with dry ice (solid carbon dioxide). Each method can be used to remove surface contamination such as oxide films and loosely-embedded particles. The degrees of success in decontamination are variable, and depend significantly on the shape and form of the material or component in question.

Decontamination of lead using shot blasting has been investigated at INEL.[3][8] It can produce high decontamination factors but is basically limited in application to brick, sheet and other forms with relatively large surfaces. The method would thus not be appropriate for decontaminating lead wool or shot, or very intricate shapes. Shot blasting has the advantage of being a long established, readily available and inexpensive technology. A disadvantage is that it will result in the removal of some base material and the creation of a secondary waste stream. In addition, there will be a significant risk of spreading the contamination unless the activity is performed in an enclosed environment (e.g., glove box, special enclosure).

Blasting with glass, sand, alumina, plastic and similar materials will produce similar decontamination results as shot blasting, and enjoy the same advantages and disadvantages. High-pressure blasting with polymer beads is being used at Department of Defense (DOD) facilities for removal of paint from aircraft and other equipment, and may have applicability for lead decontamination.[9]

A recent innovation is the use of dry ice blasting to decontaminate reactor components and parts [10,11,12,13,14]. The method relies on the abrasive action of blasting with solid particles (in this case, dry ice pellets), the agitation associated with the shattering of the particles on impact, the thermal shocking associated with the presence of dry ice, and the action resulting from the sublimation of the particles and subsequent expansion of the CO₂ gas produced. Unlike the other dry abrasive methods, no significant secondary waste stream is created because the cleaning medium, carbon dioxide, sublimates and the resulting gas is exhausted through a series of HEPA filters into the atmosphere. As with other blasting methods, the dry ice jets are most effective in decontaminating parts and components with relatively simple configurations (e.g., lead bricks and sheets). Decontamination factors are apparently comparable to those obtained with other dry blasting techniques and it is planned to incorporate the method in the INEL program for decontaminating lead [1]. The equipment needed to conduct dry ice blasting is available commercially and consists of an integrated system which includes a decontamination room, decontamination cell, air filtration system, and CO₂ storage and supply facilities.

Ice blasting [15] is a relatively recent addition to the decontamination options available. It uses low pressure air to direct ice particles onto contaminated surfaces, where the particles fracture on impact. The physical agitation that accompanies the fracturing is apparently sufficient to remove most non-bonded foreign particles. Later, as the ice melts, the resulting

water flushes the loosened material from the surface. The water can then be filtered and recycled, minimizing the size of the secondary waste stream. In addition, because the process is considerably less abrasive in its action than methods such as shot blasting, very little, if any, base metal is removed. The method has been used to decontaminate lead bricks at the Oak Ridge National Laboratory (ORNL) and fixed contaminants were reportedly reduced to below free release levels without the creation of a secondary mixed waste stream. However, no quantitative details have been obtained concerning its effectiveness.

2.1.2 Wet Abrasive Blasting

Wet abrasive blasting encompasses techniques such as hydrolasing and slurry blasting. In these procedures, fluid media are used to provide the main mechanical force or to supplement the effects due to impact of solid media. In the latter instance, the fluid may act in several capacities - transfer medium, washing solution, dust and debris minimizer, etc. The fluid medium of choice is usually water but other, non-aqueous fluids have been used. Freon, in particular, showed considerable promise at one time but is rarely considered now because of environmental concerns.

Hydrolasing (water or steam jetting) has been employed over the years for a great variety of cleaning operations, from common domestic and industrial equipment to various items associated with the nuclear industry.[16] Although a plain water/steam system is often sufficient to produce the required degree of cleanliness, improvements can be gained by incorporating agents such as detergents and acids in the fluid [6]. Hydrolasing is reportedly used by SEG, Inc., for removing surface contamination from lead brick and sheet [3]. As with other blasting methods, though, it is not considered to be efficient in removing contamination from lead wool, shot, turnings and small scrap pieces. When a simple water/steam jet is used, there will be essentially no removal of base material. However, if an acid or other strong chemical is incorporated into the fluid stream, some lead dissolution can be anticipated. After loose material and debris have been filtered from the liquid, it can be reused in subsequent decontaminations, thus minimizing the generation of a significant secondary waste stream.

Slurry blasting involves many of the same abrasives that are used in dry abrasive blasting (e.g., alumina, glass, etc.) [3,4,5,16,17]. Thus, in effect, the process combines the abrasive actions of a dry medium with the washing action of the fluid medium. This type of process is well established and commercial equipment is readily available, though at a cost in excess of that of a dry system. As with shot blasting and similar dry systems, there is the ultimate problem of a secondary waste stream. Also, as with other decontamination processes based on blasting, slurry blasting is most effective when applied to relatively large flat surfaces, such as lead brick and sheet. Los Alamos National Laboratory (LANL) has reported success in decontaminating lead bricks and shielding using low pressure blasting with a mixture of alumina, water and air, a system which removes only a few thousandths of an inch of lead.[17] It is not recommended for decontamination of small or intricately shaped items (e.g., lead wool, shot or shavings).

Although not currently in favor because of environmental concerns, blasting with Freon showed considerable promise as a decontamination method during the early 1980's.[6,18,19] However, when Freon decontamination of lead was investigated by INEL, the results were reportedly very poor.[8,19] The Freon process is not included in INEL's overall lead decontamination program.

2.2 Chemical Methods

Chemical methods of decontamination can be divided into two categories: those that use a detergent or cleansing agent and those that rely on aggressive reagents (acids and bases). For convenience, electrochemical methods for decontamination are also included in this general category. In practically all cases, the chemical methods can be used to decontaminate lead in all forms and are not restricted to bricks, sheet and similar forms.

2.2.1 Detergent Solutions

The term "detergent" is used here as an umbrella to cover such agents as soaps, degreasing agents, foaming cleansers, gels and pastes; that is, agents that have low or non-existent corrosivity. The usefulness of detergent solutions is thus basically limited to the removal of loose surface contamination. In addition, although decontamination can be accomplished via simple immersion in the solution, the effectiveness can be significantly increased by the addition of some mechanical action such as swabbing, scrubbing or brushing [3,6]. Vibratory or ultrasonic equipment can also be used to supplement the detergent action when activity levels require remote operations. The supplemental equipment needed is readily available and the technology is common and familiar. Consequently, capital costs are low compared with other decontamination processes. On the negative side, the process has the potential for creating large volumes of liquid secondary waste and the associated costs of disposing of such a waste stream. A listing of some of the commercially available detergent-type decontamination agents is given in [20] but no data are contained in that reference on their effectiveness with contaminated lead.

INEL has indicated that it will employ these agents in the first stage of its multi-stage process for lead decontamination but does not appear to have carried out any actual tests to determine their effectiveness or the amount of associated secondary waste generated.[3] Rockwell International has reported success decontaminating non-porous surfaces using foam applications of Turco products, one of which was the mildly alkaline detergent cleaning powder, Turco 4324.[21] Similarly, SRS has reported encouraging results using Turco 4324 NP in its lead decontamination program.[5] However, SRS notes that the used solutions contained lead and created a disposal problem. The lead could be effectively removed using ion exchange media but the process might be prohibitive on a large scale because of the excessive volume of ion exchange media that would be needed.

2.2.2 Acidic and Alkaline Solutions

There are a variety of commercially-available decontamination agents that rely on the chemical actions of acidic and basic (alkaline) solutions, examples of which can be found in [18]. They differ from the detergent type of decontamination agent in that they typically dissolve some of the underlying base metal in addition to loosening and dissolving the surface contaminants. Thus, as with the mechanical (blasting) methods, they can be used to reduce subsurface contamination levels. However, in doing so, a secondary mixed waste stream, with its attendant problems, is generated. This issue was noted above with respect to the decontamination experiments done by SRS using Turco 4324 NP.[5] The SRS experiments also looked at using 1N nitric acid but concluded it was less effective than the proprietary agent. In addition, SRS reportedly identified a nitric-oxalic acid solution used to decontaminate lead but further details on this agent and its effectiveness have not been located.[21] In general, though experiencing some success with chemical decontamination processes, SRS has not favored their use because of the secondary mixed waste stream problem.[5,7]

Successful decontamination of more than 20 tons of lead using an acid-based process has been reported from the Pantex weapons facility.[22] The process employed a solution containing an (unidentified) acid and hydrogen peroxide, and is apparently in common use commercially for stripping lead coatings from copper alloys and steel. The lead components investigated were initially in different forms: classified cylindrical shapes, classified hemispherical shapes and assorted declassified shapes. Many had been subjected to some preliminary disfiguring (twisted, crimped, etc.) so, to ensure contact with the decontamination solution, these had to be pried open, cut and flattened. Decontamination efficiency varied with the type of contamination. This process was more effective in removing loosely bound than tightly fixed contamination. Reference 21 also reports on the amount and treatment of secondary waste resulting from the process. Typically, decontamination using this acid/peroxide mixture has a lead removal rate of about $0.84 \mu\text{m}/\text{minute}$ and a operation lasting 17 minutes would remove $14 \mu\text{m}$ of material.

INEL has indicated that it will employ some of these agents in its multi-stage process for lead decontamination but does not appear to have carried out any actual tests to determine their effectiveness or the amount of associated secondary waste generated. [3] Both acid and alkaline solutions are included but the actual chemicals are not identified. INEL notes that Scientific Ecology Group, Inc., (SEG) in Oak Ridge prefers washing with an alkaline rather than an acid solution when dealing with lead because of the very insoluble solids formed.

2.2.3 Electrochemical Methods

Electrochemical methods have been used to remove contamination from metal surface and have numerous advantages, including short application times and high decontamination factors. [6,16] However, though details are available on the application of such methods to the decontamination of stainless steel and other reactor alloys (e.g., [23]), no reference has been found to its use in decontaminating lead.

Rocky Flats Plant (RFP) demonstrated the feasibility of decontaminating stainless steel from plutonium-containing glove boxes using a basic electrolyte containing sodium nitrate, sodium tetraborate and sodium fluoride.[24] The report also includes data on the dissolution rates of lead and several other metals using this electrolyte and concludes that the method would also be applicable to decontamination of these other metals. However, no subsequent testing to demonstrate this was found in the literature.

INEL has reviewed the advantages and disadvantages of electrochemical methods of decontamination and recommended that none be included in their lead decontamination program.[3]

2.3 Melt Refining

Decontamination using melt refining is based on the principle that contaminants in a molten metal tend to partition between the melt and an overlying slag medium, with the contaminants preferentially concentrating in the slag.[25] The practice is common in the commercial metal refining industry and is now being adapted for decontaminating radioactively contaminated material, including lead.[3,4,6,8,26,27] The unique attribute of this process is that it has the ability to remove contaminants from within the bulk of the material and reduce the activity of the base material to an acceptable release level without significant dissolution.

— Much of the early development work in the DOE complex was done at ORNL and covered a range of materials, including lead.[25,27,28] The experiments investigated the effects of such variables as slag composition, melting technique and crucible material. The results of this early work on removal of uranium impurities indicated that the technique had great potential, with decontamination factors of greater than 1000 being noted for lead.

The INEL investigations into decontamination of lead through melt refining are described in [29,30] The results from the preliminary bench tests using isotopes of strontium, cesium and cobalt indicated an average decontamination factor of approximately 4700.[29] Later pilot scale tests (using strontium, cesium and cobalt isotopes) also recorded a high average decontamination factor, of around 5000.[30] Subsequently, melt refining was recommended as one of the steps to be included in the INEL lead decontamination program. As such, it would be a final stage if initial attempts at decontamination by the detergent washing, acidic and basic washing, and dry ice blasting did not produce the desired results.

Melt refining can be used on all forms of lead if there is no desire to retain the original shape. Thus it could be particularly appropriate for lead shot, wool and scrap. However, although equipment is available on the commercial market, initial costs could be high compared with other methods of decontamination. There will also be a secondary mixed waste stream (the dross from the melt) that would probably require macroencapsulation or some other treatment before disposal.

2.4 Laser Ablation

The use of laser ablation to decontaminate radioactive material is a very recent innovation.[13] High-power, short-pulsed laser beams are focused on the surface of a contaminated component, removing material from the surface. From there, the released material can be collected in a particulate air filter system to prevent redeposition. The technique is still in the development stage at the Ames Laboratory and is probably some years away from becoming a viable method in actual component decontamination operations.

2.5 Macroencapsulation

As discussed in Section 2, the technology-based standard treatment for radioactively-contaminated lead prior to land disposal is macroencapsulation with a surface coating material (e.g., plastic polymer). It is also conceivable that macroencapsulation could be used for sealing secondary waste products such as the slag formed during the melt-refining process. (Other secondary waste products such as wash solutions would be more appropriately treated by solidification or microencapsulation processing.) Provisions for macroencapsulation are contained in the INEL lead decontamination program [3], and are under consideration as viable mixed waste treatment options at SRS and RFP [31,32]. A synopsis of this process can be found in both [31] and [32]. Both SRS and RFP are looking into encapsulation using organic polymers, specifically, in the case of RFP, polyethylene. A review of RFP's macroencapsulation process was conducted following a recent site visit and is summarized in Reference 33. The general intent in all cases is that macroencapsulation will be used when recycling is not a viable or cost-effective option. This does not preclude the conducting of one or more decontamination steps prior to the encapsulation to remove loosely-bound surface contamination before coating. While this may improve adhesion between the coating and the base material, it must be weighed against the disadvantages inherent in creating a secondary waste stream.

2.6 Review of Potential Treatment Technologies Summary

It is readily apparent from this evaluation that none of the methods have universal applicability, i.e., no method can completely decontaminate lead in all forms regardless of the type of contamination.

Those processes that take advantage of mechanical action, whether under wet or dry conditions, are capable of effectively removing surface contamination and, in some instances, an amount of subsurface material. Application of such methods is basically limited to brick, sheet and similar substantial forms with relatively large contiguous surface areas. Equipment costs range from the mundane to the relatively expensive but all systems generate a secondary waste stream that will require treatment and disposal. In some cases, savings in initial equipment costs may be more than offset by increased costs in waste disposal. Of the

commercial systems available, the most attractive appears to be dry ice blasting. Its major attribute is generation of a minimal secondary waste stream. Fully integrated systems can be bought, rented or leased (in the U.S., the major supplier is NDC, Inc.).

A variety of decontamination processes are also available that rely principally on chemical action to remove the contaminants. These include washing with soap and detergent-type materials, immersion in acid and alkaline solutions, and electrochemical polishing and etching. There is virtually no experience with decontaminating lead using electrochemical methods, so this does not appear to be a viable option at this time. The other two types of chemical methods are well established and have their individual advantages and drawbacks. Both have the potential to decontaminate lead in any physical configuration. The detergent type of solution is generally non-aggressive, does not attack the lead substrate and is appropriate for the removal of loosely-attached contaminants. The more aggressive acid/alkaline solutions will dissolve the more adherent contamination that might be associated with the oxide layer and the metal substrate. Inevitably, both processes will generate a secondary waste stream, which will, in the case of the more aggressive media, contain lead and be a mixed waste. Of the two types, use of a system employing only soaps and detergent agents appears to be the best option if it is known that the contamination is mostly loosely bound to the substrate.

Melt refining appears capable of accommodating all decontamination needs but the process, though becoming established, nevertheless involves high capital equipment costs. A more rigorous engineering cost benefit analysis will be required to evaluate fully its usefulness. It would obviously represent overkill if all the contamination were known to be surface type and easily removable by other methods such as washing with detergents or dry ice blasting. However, it offers the only reasonable option if the contamination is tightly bound within the bulk of the material. The process also produces a secondary waste stream but one which can be relatively easily treated using macroencapsulation.

Decisions are required concerning the extent to which decontamination should be attempted in any particular case. Selection of a suitable decontamination system will depend on several factors including the type and extent of contamination, physical form of the lead, volume of lead requiring treatment, cost of disposal vs. treatment cost/recycling credit, and secondary waste generation. In some instances, it will be considered very feasible to decontaminate to a level of activity that will permit reuse or recycle of the lead. In other cases, it will be apparent that such a level can only be achieved at great cost, both in financial terms and in increased exposure risks. Recycle/reuse will not be a viable option in these cases, requiring macroencapsulation prior to disposal.

Based on the findings of this review and the resources available for this effort, a list of chemical and physical decontamination options was developed for further study. Feasibility testing was conducted and results are presented in Section 4.

3. CHARACTERIZATION OF ACTIVATION PRODUCTS IN LEAD

In order to develop a screening technique to sort surface contaminated lead from activated lead, a review was conducted and model calculations were run to characterize activation products that are likely to be present. The bulk activation may be the result of neutron induced activation from a reactor, accelerator, or experimental environment, or it may contain spallation products if it was used in an accelerator. The presence of impurities in the material will also determine the quantity and character of the activation products.

This section outlines possible activation products and the corresponding prominent decay photons from several different sources. These include neutron-induced activation of lead and impurities from a reactor or low-to-intermediate energy neutron source experiment, and the consideration of spallation activation products from an accelerator environment. The resulting decay photons are gamma-rays and x-rays that result from beta, positron, electron capture, alpha, or internal transition decays. These emissions are characterized by discrete energies and intensities that are uniquely identified with the isotope involved. This information on photon signatures may then be used to sort surface contaminated lead from activated lead.

3.1 Neutron-Induced Activation of Pure Lead

Activated lead produced by interactions with low-to-intermediate energy neutrons may have been generated at several BNL facilities, including the High Flux Beam Reactor (HFBR), Brookhaven Medical Research Reactor (BMRR), and a myriad of fixed neutron source experiments. These systems may be characterized by neutron spectra ranging from low (thermal) to intermediate energy (several MeV).

In order to characterize the activation products generated by neutron-induced activation of lead, several calculations using the ORIGEN2 depletion and radioactive decay code were performed.[34,35] This is a one neutron-energy group code that is widely used throughout the nuclear industry to simulate reactor fuel cycles. The spatial and energy dependent effects in the simulation are included in the nuclear cross section data used in the analysis. The cross sections are weighted over a specific geometric configuration and the associated energy spectrum. The calculations performed correspond to the following neutron spectra:

- Thermal spectrum
- Candu spectrum: Heavy water moderated reactor with enriched uranium (The HFBR is a heavy water moderated system utilizing 93% enriched uranium. This is a slightly undermoderated system with the neutron flux peaking in the reflector region. The average neutron flux in this region is approximately 2×10^{14} neutrons/cm²-sec.)

- Fast Flux Test Facility (FFTF) spectrum: maximum neutron energy is 10-14 MeV. (This was used to represent various fixed neutron source experiments having a fast energy spectrum.)

The ORIGEN2 calculations utilized a point neutron flux of 2×10^{14} neutrons/cm²-sec and a 200 day irradiation period followed by decay cooling periods of one and two years. A one kilogram sample of lead was used without the inclusion of impurities. The results using the thermal spectrum cross sections yielded the highest overall concentration of neutron-induced activation products in lead. Table 1 gives the principle neutron induced activation products produced in lead with half-lives greater than 1 day. Of these radionuclides, the most significant contributor for a one year decay period is Po-210 (138.4 day half-life) with an emission of a 0.8 MeV photon. The presence of certain impurities discussed in Section 3.2, may dominate the decay photon emissions.

Table 1. Principle Neutron-Induced Activation Products Produced in Pb with Half-Lives > 10 Days^(a)

Isotope	Half-life	Decay mode ^(b)	Prominent Photopeak Energies (MeV) ^(c)
Hg-203	46.6 days	β^-	0.28
Bi-208	3.7 yr	EC	2.61
Bi-210m	3.0×10^5 yr	α	0.27, 0.31
Bi-210	5.01 d	α, β^-	0.31, 0.27
Po-210	138.4 d	α	0.8

^{a)} Energy spectra: Thermal to FFTF (10-14 MeV). Only activation products with half-lives > 1 day are presented.

^{b)} Decay Modes: β^- : beta emission; IT: internal conversion; EC: electron capture; β^+ : positron emission; α : alpha particle emission

^{c)} Multiple photons are listed in order of decreasing intensity

^{d)} Isomeric state

3.2 Neutron-Induced Activation of Impurities

The presence of impurities in the lead shielding material may significantly contribute to the neutron-induced activation. Representative elemental analysis of typical impurities are available from suppliers of shielding materials. One supplier, Reactor Experiments Inc., indicated that typical impurities may include antimony (Sb), silver (Ag), tin (Sn), copper (Cu), arsenic (As), zinc (Zn), iron (Fe), and bismuth (Bi).

Table 2 summarizes the effects of neutron activation of typical impurities contained in lead. Calculations were performed using the ORIGEN2 code for a point neutron flux of 2×10^{14} neutrons/cm²-sec and a 200 day irradiation period followed by a one year decay cooling time. The neutron cross sections were averaged over a thermal spectrum. A one kilogram sample of lead was used with 0.05% of the aforementioned list of impurity elements.[36]

Table 2. Neutron-Induced Activation of Typical Impurities in Lead with Half-Lives > 10 Days

Isotope (MeV)	Half-life	Radioactivity Level (Curies) (1 yr decay)	Prominent Photopeak Energies (Intensities)
Co-60	5.3 yr	2.08×10^{-4}	1.173 (100%), 1.332 (100%)
Zn-65	243.8 d	1.45	1.116 (50.8%)
Ag-108m	127.7 yr	6.53×10^{-2}	0.43(91%), 0.61, 0.72(90%)
Ag-109m ^(a)	39.8 sec	4.09×10^{-2}	0.088 (3.6%)
Ag-110m	252.2 d	3.28	0.66 (94%), 0.89 (73%)
Cd-109	462.0 d	4.09×10^{-2}	0.088 (3.8%)
Sn-113	115.1 d	8.49×10^{-3}	0.392 (64%)
Sn-119m	293.0 d	8.21×10^{-3}	0.024 (16.2%)
Sn-123	129.2 d	1.06×10^{-2}	1.089 (0.6%)
Sb-124	60.2 d	3.25×10^{-1}	0.60 (98%), 1.69(49%)
Sb-125	2.8 yr	2.49×10^{-2}	0.428 (30%), 0.601 (18%)
Te-123m	119.7 d	8.48×10^{-3}	0.159 (83.6%)
Te-125m	58.0 d	6.04×10^{-3}	0.109 (0.27%)
Po-210	138.4 d	1.56×10^{-2}	0.80

^{a)} Produced from the decay of Cd-109 (by electron capture)

3.3 Activation Products from High Energy Particle Interactions

When a high energy proton (GeV) interacts with a nucleus, neutrons, protons and other nuclear fragments may be emitted, converting the struck nucleus to that of a different isotope, which most probably is radioactive. Some of the secondary particles emitted in an interaction may have sufficient energy to cause further activation by spallation reactions or be captured by nearby nuclei. This series of inelastic scattering reactions results in a nucleonic cascade process. The residual target nucleus is the spallation radioactive product. The emitted neutrons can have a maximum energy up to the energy of the incident high energy particle and may range in number up to 20 neutrons per incident proton. The overall radioactive inventory induced depends on the amount of primary beam loss. The probability of producing a particular isotope depends on the composition of the material struck and the presence of impurities, the spectrum of secondaries produced, and the production cross section of the isotope concerned. The amount of a radioactive isotope present at a given time also depends on the half-life and the time that the accelerator has been in operation as well as the time that the activity has had to decay since operation stopped. Furthermore, the mass distribution of the radionuclide inventory will also be a function of the thickness of the target material. Therefore, the complexity of the processes governing the amount of radioactivity at any one time makes it very difficult to quantify the activity in any detail. Table 3 gives some of the principal radioactive isotopes produced by spallation reactions in accelerator structures.[37,38]

A series of experiments were performed at BNL and at Eastern Kentucky University in order to obtain specific information on the isotopic identification and yields of the spallation products produced in an accelerator reaction.[39] Ten activation experiments of lead and tungsten target materials were carried out in the beam line of the AGS at BNL using protons of 0.84, 1.0, and 1.42 GeV. Results from one experiment are summarized in Table 4, illustrating the complexity and difficulty in providing specific identification signatures for activated lead in an accelerator environment. In this experiment, a beam of 1.0 GeV protons irradiated a lead target assembly for 210 minutes. The assembly configuration was a 10.2 cm diameter cylinder, 61 cm long, mounted to a U-shaped rail composed of aluminum. Measurements were taken at the surface of the target and at depths up to 35 cm within the material. The counting of the activated target commenced at 1.5 hours after cessation of the beam and continued to 112 days afterward. The results of the isotopic yields were extrapolated to the beginning of the counting time. Forty eight isotopes with mass numbers in the range of 161 to 206 were detected. Table 4 lists the abundance of those with half lives more than 1 day.

Table 3. Principal radioactive isotopes produced in accelerator structures by spallation reactions .

Isotope	Half-life	Decay mode	Energy of photons (MeV)
Be-7	53 d	EC	0.478
C-11	20 min	β^+	0.511
F-18	1.8 h	β^+	0.511
Na-22	2.6 y	β^+	1.27, 0.511
Na-24	15 h	β^-	1.37, 2.75
Sc-46	84 d	β^-	1.12, 0.89
Sc-48	1.8 d	β^-	0.98, 1.3, 1.04
V-48	16 d	β^+	0.98, 1.3, 0.511
Cr-51	28 d	EC	0.32
Mn-52	5.7 d	β^+	1.4, 0.94, 0.74, 0.511
Co-56	77 d	β^+	0.85, 1.24, 0.511
Co-60	5.3 y	β^-	1.33, 1.17
Zn-65	245 d	EC	1.11, 0.511

Decay Modes: EC: electron capture; β^+ : positron emission; β^- : beta emission

In order to obtain information on the specific characterization of the radiation environment of the lead shielding materials used in the accelerator facilities onsite, several meetings were held with AGS engineers and physicists. The objectives of the discussion were to ascertain the probability of a high energy proton (currently 24 GeV) would directly impact a lead shielding object. This would only occur when a dipole magnet goes off and the proton beam sweeps into a fault mode. It is estimated that this has occurred with a frequency of less than 10 hours per year for a total 20 week annual operating period. It was stated that a beam spill on the lead has not been detected within the last one-to-two years, due to the implementation of a new searching procedure whereby the lead is not exposed to a beam spill. It was also mentioned that there may be a physical signature from the collision of a 24 GeV proton on lead: a hole melted through the length of the shielding object.[40] Therefore, most of the lead shielding material in the AGS is exposed to neutrons, photons, and other charged particles, primarily neutrons and photons. In the absence of further information, the neutron spectra could range from thermal to 24 GeV.

Table 4. Radioisotope Production

1.0 GeV Protons on Lead Irradiation period: 210 minute Counting time after end of beam: 1.5 hrs to 112 days (Yields at surface of lead target)			
Isotope	Half-life	Average Yield (At start of counting) (nuclei/proton/cm ³)	Prominent Photopeak Energies (MeV) (*)
Bi-206	6.24 d	1.489×10^{-6}	0.516, 1.719, 0.537
Bi-205	15.31 d	2.391×10^{-7}	1.861, 1.764, 0.703
Bi-204	11.2 d	1.971×10^{-6}	0.984, 0.912, 0.918
Hg-203	46.61 d	1.215×10^{-6}	0.279 (81.5%)
Pb-202m	3.53 hr	2.864×10^{-6}	0.961, 0.422, 0.787
Tl-201	3.04 hr	1.982×10^{-5}	0.168, 0.135
Au-200m	18.7 hr	7.252×10^{-7}	0.256
Pb-200	21.5 hr	1.358×10^{-5}	0.236, 0.269, 0.451
Tl-199	7.4 hr	1.197×10^{-5}	0.247, 0.158, 1.013
Au-198m	2.3 d	1.354×10^{-6}	0.097
Tl-198	5.3 hr	9.802×10^{-6}	1.435
Pb-198	2.4 hr	1.795×10^{-6}	0.290, 0.173, 0.865
Tl-197	2.83 hr	5.380×10^{-6}	0.426, 0.152, 1.411
Hg-197m	24.0 hr	1.757×10^{-6}	0.134
Au-196	6.18 d	1.106×10^{-6}	0.333, 0.426
Hg-195	9.5 hr	2.891×10^{-5}	0.585
Au-194	1.64 d	1.442×10^{-6}	0.328
Hg-193	3.8 hr	1.090×10^{-5}	0.186
Hg-193m	11.8 hr	4.739×10^{-6}	0.408, 0.573
Hg-192	5.0 hr	5.755×10^{-6}	0.275, 0.186
Au-191	3.2 hr	1.177×10^{-5}	0.587
Pt-191	2.96 d	1.113×10^{-5}	0.097, 0.135

Isotope	Half-life	Average Yield (At start of counting) (nuclei/proton/cm ³)	Prominent Photopeak Energies (MeV) (°)
Ir-189	13.2 d	1.939×10^{-5}	0.245
Pt-189	10.9 hr	1.490×10^{-5}	0.721, 0.608, 0.300
Re-189	24.0 hr	4.840×10^{-6}	0.219
Pt-188	10.2 d	1.026×10^{-5}	0.188, 0.195, 0.382
Ir-187	10.5 hr	4.060×10^{-7}	0.978, 0.502
Pt-186	2.0 hr	1.658×10^{-6}	0.689, 0.611
Ir-186m	1.7 hr	5.284×10^{-6}	0.137, 0.435
Ir-185	14.0 hr	9.253×10^{-6}	1.829, 0.137
Os-185	93.6 d	1.038×10^{-5}	0.646 (81%)
Hf-184	4.1 hr	1.779×10^{-6}	0.480, 0.451
Ir-184	3.0 hr	4.227×10^{-6}	1.105, 2.063
Os-183m	9.9 hr	5.140×10^{-6}	1.102, 1.108, 1.035
Os-182	21.5 hr	9.702×10^{-6}	0.180
Re-181	20.0 hr	1.272×10^{-5}	0.953, 0.662
Yb-177	1.9 hr	2.967×10^{-4}	0.899
W-177	2.2 hr	2.884×10^{-6}	0.116, 0.427, 1.037
Lu-176m	3.66 hr	1.035×10^{-6}	0.413, 0.172
Ta-176	8.1 hr	5.372×10^{-6}	1.824, 1.863
Ta-175	10.5 hr	8.470×10^{-6}	0.348, 0.126, 1.793
Hf-173	23.6 hr	9.115×10^{-6}	0.124, 0.311, 0.162
Ta-173	3.6 hr	3.308×10^{-6}	0.172, 1.208, 0.206
Er-171	7.52 hr	1.021×10^{-5}	0.296
Hf-171	12.2 hr	5.992×10^{-6}	0.662, 0.347
Lu-171	8.24 d	8.283×10^{-6}	0.740, 0.667, 0.781

Isotope	Half-life	Average Yield (At start of counting) (nuclei/proton/cm ³)	Prominent Photopeak Energies (MeV) (a)
Hf-170	16.0 hr	7.708×10^{-6}	0.165, 0.621, 0.573
Lu-169	1.4 d	5.322×10^{-6}	0.191, 1.185
Yb-169	32.03 d	6.977×10^{-6}	0.198 (35.6%), 0.110 (17.4%), 0.131 (11.4%)
Tm-167	9.24 d	5.559×10^{-6}	0.208
Yb-164	1.26 hr	9.799×10^{-7}	0.106
Er-163	1.25 hr	9.008×10^{-7}	0.564
Tm-163	1.81 hr	1.421×10^{-6}	1.265
Er-161	3.21 hr	2.113×10^{-6}	0.826

a) Multiple photons are listed in order of decreasing intensity.

4. SCREENING OF CONTAMINANTS

Based on the identification of characteristic gamma spectra associated with activated lead and lead impurities, testing was conducted on actual lead sampled from Building 650 to identify those radioactive species deposited either as surface contaminants or the result of specific activation of the bulk material.

Ten relatively small, randomly chosen lead pieces were removed from two auxiliary storage facilities located adjacent to Bldg 650. The test specimens were in the shape of bricks, sheets, lead wool and other geometric configurations with the largest piece measuring approximately 25.4 cm x 10.2 cm x 7.6 cm (10" x 4" x 3"), and weighing ~15.9 kg (35 lbs.).

A GeLi gamma detection system (using Canberra System 100 software) was used to qualitatively identify the radionuclides present on or within the lead test specimens. This particular GeLi detection system was designed to analyze light weight liquid solutions and solids and therefore, a modification to the detector holder assembly was necessary to support the increased mass of the lead pieces. An adjustable support rack was built to accommodate not only this mass increase, but the variation in size and configuration of each lead piece. In addition, the maximum size of the lead specimens was restricted due to the limited space available within the shielding cave which surrounds the detector (maximum dimensions 28 cm x 28 cm, 11" x 11").

Each piece of lead was double wrapped with polyethylene bags prior to entering the counting room to avoid inadvertent contamination of the detector and the surrounding areas. Before gamma-scanning, each piece of lead was surveyed using a Ludlum Model 3 survey meter for determining radioactive hot spots. The lead was then placed on the support rack with the radioactive hot spot positioned over the detector, the cave door was closed and a full spectrum, gamma-ray analysis was initiated. Each piece of lead was initially counted for 15 minutes to determine if radionuclide identification was possible. If counting statistics were poor, but minor photopeaks identified, the lead pieces were counted again for 2 hours.

Only five of the ten lead pieces randomly chosen from the two storage facilities were identified as being radioactively contaminated or activated. The five lead pieces that generated significant photopeaks during gamma scanning were further identified by their centroid photopeak energies. In some cases, a particular radionuclide may possess multiple photopeaks (at different energies) with different lines of intensity. For example, Co-60 has two photopeaks at 1.173 and 1.332 MeV, each with an abundance of 100%, thereby making their identification relatively easy. Other radionuclides may have multiple peaks in which one energy peak is present in greater abundance than the others. In either case, the identification of an unknown radioactive species can be accomplished with a greater degree of certainty if it is characterized by multiple peaks.

After the energy identification of the strongest photopeaks was completed, two methods of radionuclide verification were employed. The first method of radionuclide identification was

performed by a manual search using two reference books.[41,42] All short lived isotopes (half-lives <100 days) were disregarded as the energy search to match the unknown radionuclides proceeded. Following completion of the manual search, a confirmatory identification was employed using two Canberra software programs (Spectrum-AT and Peak Search and Iso Id.). These software routines automatically locate peaks in the spectrum, compare the peak's centroid energy with isotope energies located in the software library which then identifies the isotope. A mixed gamma standard source was counted and calibrated prior to identifying any unknown radionuclides. The software isotope identification analysis quoted errors at one sigma with the peak confidence level at 95% (two sigma). One limitation to the software is that the library does not contain a complete listing of all the gamma-ray emitting radionuclides.

Three positive (Cs-137, Co-60 and Sr-85) and two possible (Cs-134 and U-235) radioactive contaminants were identified on at least one of the five pieces of lead that were analyzed. Isotopes of Cs, Co, and Sr are produced during the fission process and thus, are evidence of surface contamination. The U-235 is probably the result of natural uranium contamination, but further testing would be required to verify this with certainty. Two radioactive isotopes produced by deuteron activation (Bi-207 and Pb-202) were also positively identified on one piece of lead. These activation products were most likely generated from the impurities present in the lead. The identification of Bismuth-207 was confirmed by both isotope table reference and Canberra isotope library software. Lead-202 could only be identified by isotope table reference because the Canberra isotope library software did not include this gamma-ray emitting radionuclide.

The following four major photopeaks energies (and their accompanying abundance) were observed in counting one test specimen: 72.8 KeV(23%), 74.97 KeV(38.6%), 569.7 KeV(98%) and 1063.6 KeV(77%) enabled Bismuth-207 (Half-life 38 yrs) identification. Once the radionuclide identification was confirmed, reference to the isotope tables verified that Bi-207 is produced principally by deuteron activation of the impurities in the stable form, Pb-208. Lead-202 (Half-life $\sim 3 \times 10^5$ yrs) was also identified as a deuteron activation product with distinguishing photopeaks located at the following energies (abundance): 70.8 KeV(23%), 72.9 KeV(41%), 82.5 KeV(15%) and 84.9 KeV(4%). Referring to the isotope tables revealed that the principal means of production is by deuteron activation of the stable form of Thallium-203 in the lead.

5. BENCH-SCALE DECONTAMINATION AND TREATMENT TECHNOLOGY EVALUATION

5.1 Wet Chemical Methods

Based on the literature review and evaluation of existing/emerging decontamination technologies (Section 2) and other sources, many potential wet chemical decontamination solutions were identified. In order to conduct bench-scale testing and determine the effectiveness of potential solutions, the list was narrowed to the seven candidates indicated below:

Detergents

- Alconox

Chemical Solutions

- 10% acetic acid
- 5% nitric acid
- 5% nitric acid with 10% sodium nitrate
- 10% hydrogen peroxide with 10% sodium nitrate
- 10% hydrogen peroxide with 10% acetic acid
- Quick -off

Effectiveness of the decon solutions was measured by quantifying the mass of material removed from the surface during immersion. All the initial tests were performed by immersing non-radioactive lead coupons in 200mL of each of the chemical solutions at room temperature for five minutes. Depending on the results, other techniques were tested including longer immersion times, elevated solution temperatures, and various methods of agitation. Chemical solution immersion tests with agitation were identical to the 5 minute static tests, except mixing was employed. Elevated temperature (50°C) immersion tests were static in nature and performed over a 24 hour period.

Uncontaminated, 28.6 mm dia. x 3.2 mm thick (1 1/8 in. dia. x 1/8 in. thick) lead coupons were fabricated using an Arch punch and a Carver hydraulic laboratory press. Prior to immersion, nominal dimensions and precise mass measurements were obtained for all lead specimens. Each coupon was identified by a number that was stamped into the lead surface, and a 1.6 mm (1/16 in.) hole was punched near the edge of the coupon using a scratch awl, thus enabling the entire disc to be suspended in the chemical solution. The coupons were immersed in solutions contained in a 400 mL beaker, supported by a stainless steel wire. Following the test, lead specimens were dried, re-weighed and visually inspected. The ability of each solution in removing the surface layer was quantified in terms of g/cm² and percent loss in mass.

Following the five minute static tests, additional immersion tests were performed using two different methods of agitation. Immersion solutions were agitated by means of a rotary stirrer and ultrasonic bath. Rotary stirring was accomplished by using a magnetic stir bar, while agitation by high frequency sound waves (ultrasonic) was accomplished by placing the 400mL

beaker and its contents into an ultrasonic water bath. Elevated temperature testing was conducted at 50°C using a vertical neck distillation flask and heating mantle with an in-line reflux condenser. All the elevated immersion tests were performed for a 24 hour period. Testing and measurement procedures were similar to previous protocol.

The mass of material removed from the surface as a function of surface area and the resulting percent weight loss for lead coupons during five minute static and dynamic immersions are presented in Table 5 and are shown graphically in Figure 1. In almost every case (regardless of chemical solution), the quantity of the surface removed by chemical treatment was directly proportional to the aggressiveness of agitation. For example, the most effective method was ultrasonic agitation, followed by magnetic stirring (less vigorous agitation), with static immersion being the least effective. Only two exceptions were observed. The 5% nitric acid + 10% sodium nitrate and the 10% hydrogen peroxide + 10% sodium nitrate were slightly more effective in removal of the surface layer during static tests compared with the magnetically stirred test specimens. However, this anomaly does not appear to be statistically significant. All the coupons tested revealed lead losses of < 0.3 wt%, with many of the values significantly lower, except for those coupons placed in the hydrogen peroxide + 10% acetic acid. The dissolution losses for the lead specimens immersed in hydrogen peroxide + 10% acetic acid were calculated at 5.1, 6.5, and 18.5%, for the static, magnetically stirred and ultrasonic values respectively.

Table 5. Weight Loss Following Five Minute Static, Magnetic Stirrer Agitation and Ultrasonic Agitation in the Seven Different Chemical Decontamination Solutions.

Decontamination Solution	Weight Loss Following Chemical Treatment					
	Method of Agitation					
	Static		Magnetic Stirrer		Ultrasonic	
	g/cm ² (1 x 10 ⁻³)	% Loss	g/cm ² (1 x 10 ⁻³)	% Loss	g/cm ² (1 x 10 ⁻³)	% Loss ^d
10% Alconox	0.39	0.027	1.08	0.074	1.60	0.11 ± 0.03
10% Acetic Acid	1.39	0.095	1.77	0.12	2.72	0.19 ± 0.08
5% HNO ₃ ^a	2.34	0.16	2.61	0.18	2.95	0.20 ± 0.01
5% HNO ₃ + 10% NaNO ₃ ^b	3.15	0.21	2.39	0.16	4.10	0.28 ± 0.07
10% H ₂ O ₂ ^c + 10% NaNO ₃	0.21	0.01	0.17	0.011	0.39	0.03 ± 0.01
H ₂ O ₂ + 10% Acetic Acid	74.0	5.1	95.8	6.5	242	16.8 ± 2.4
Quick-Off (conc.)	0.80	0.05	1.17	0.085	2.67	0.18 ± 0.05

^a Nitric Acid

^c Hydrogen peroxide

^b Sodium Nitrate

^d Mean value for 3 replicates with 2σ errors.

Figure 1. Percentage of lead removed following chemical treatment as a function of the type of agitation.

Table 6 presents the results of static chemical solution immersion tests performed for a longer duration (24 hr) at elevated temperatures (50°C). The aggressive nature of both the 5% nitric acid and the 5% nitric acid + 10% sodium nitrate solutions under these conditions are demonstrated by percent weight loss in lead coupons exceeding 50%. The percent loss of lead was more than 200 times greater for the longer duration, elevated temperature immersion tests. For the remaining decon solutions tested, the improvement in lead removal efficiency for longer duration, elevated temperature conditions was less dramatic, ranging between 2 and 10 times greater than short duration, ambient temperature conditions. Consequently, the cost-effectiveness of using elevated temperatures is questionable, especially over long time periods. It should be noted that the 10% hydrogen peroxide + 10% sodium nitrate solution was not heated during the 24 hour immersion tests because of the energetic reaction that occurred during the 5 minute tests at room temperature. However, it would appear that this observed reaction had little to do with the actual removal of lead because during all three tests (24 hour and 5 minute static and magnetically stirred immersion tests) reveal the same low percentage of lead removal. The hydrogen peroxide + 10% acetic acid decon solution was not immersion tested at elevated temperatures because of the violent reaction observed during the five minute testing.

Table 6. The Percentage of Lead Coupon Dissolved Following 24 Hours of Static Immersion at 50°C.

Decontamination Solution	Weight Loss Following 24 Hours of Chemical Treatment at 50°C ^(a)	
	g/cm ²	% Loss
10% Alconox	0.018	1.25
10% Acetic Acid	0.008	0.57
5% HNO ₃ ^(b)	0.760	50.9
5% HNO ₃ + 10% NaNO ₃ ^(c)	0.835	56.2
10% H ₂ O ₂ ^(d) + 10% NaNO ₃	0.001	0.09 ^(e)
Quick-Off (conc.)	0.005	0.33

^(a) Reported in terms of % loss and loss per cm² of surface area.

^(b) Nitric Acid

^(c) Sodium Nitrate

^(d) Hydrogen peroxide

^(e) This immersion test was performed at room temperature, not heated to 50°C.

Based on the preliminary investigations (five minute and 24 hour elevated temperature chemical solution immersion tests with varying levels of agitation), the most effective and efficient method for removing surface contamination from lead is by ultrasonic agitation. Therefore, one additional test was performed on lead coupons using each decon solution ultrasonically agitated at two additional immersion time intervals of 120 and 480 minutes. The results of these immersion tests at 5, 120, and 480 minutes are presented in Table 3 and are graphically shown in Figure 2 as percent weight loss of lead. In all cases, more lead was removed with each successive increase in immersion time. However, the increased dissolution with time under ultrasonic agitation was more pronounced for some solutions. For example, the percent weight loss for lead using 5% nitric acid increased from 0.20 wt% at 5 min. to 7.0 wt% at 120 min.

Table 3. The Percent Lead Coupon Dissolution Using a Ultrasonic Bath and the Seven Different Chemical Decontamination Solutions at Three Different Time Intervals.

Decontamination Solution	Weight Loss Following Chemical Treatment ^(a)					
	Time (minutes)					
	5		120		480	
	g/cm ²	% Loss	g/cm ²	% Loss	g/cm ²	% Loss
10% Alconox	0.002	0.11	0.011	0.75	0.051	3.4
10% Acetic Acid	0.003	0.19	0.005	0.34	0.013	0.9
5% HNO ₃ ^(b)	0.003	0.20	0.104	7.0	na	na
5% HNO ₃ + 10% NaNO ₃ ^(c)	0.004	0.28	0.157	10.8	na	na
10% H ₂ O ₂ ^(d) + 10% NaNO ₃	0.0004	0.03	0.004	0.31	0.022	1.5
H ₂ O ₂ + 10% Acetic Acid	0.242	16.8	na	100	na	na
Quick-Off (conc.)	0.003	0.18	0.003	0.21	0.006	0.44

^(a) Reported in terms of % loss and loss per cm² of surface area.

^(b) Nitric Acid

^(c) Sodium Nitrate

^(d) Hydrogen Peroxide

na: not applicable (no further testing was performed because of the significant amounts of lead dissolved following 120 minutes in these particular chemical solutions and ultrasonic bath tests).

The hydrogen peroxide + 10% acetic acid dissolved 18.5% of one lead coupon after 5 minutes and dissolved 100% of a second lead coupon when left in the ultrasonic bath for 120 min. The percent weight loss data for four of the chemical solutions are plotted verses time in Figure 3.

Figure 2. Percent weight loss of lead vs. time.

Figure 3. Percent weight loss for lead vs. time for four of the chemical solutions.

In evaluating the effectiveness of a particular chemical solution in removing contamination from the surface of lead several key issues need to be addressed before choosing the best candidate. These include determining:

- the depth that lead must be removed to ensure complete contamination removal,
- the most advantageous rate of lead removal,
- the methods for treating the secondary waste residuals and at what additional cost.

Once these issues have been addressed, the cost-effectiveness of decontaminating lead using a particular chemical solution can be determined. Based on preliminary testing, the chemical decon solution that was most effective was the hydrogen peroxide + 10% acetic acid solution using ultrasonic agitation. In fact, in order to use effectively, this solution would require dilution to moderate attack on the lead matrix.

5.2 Dry Abrasion Methods

Mechanical decontamination of lead bricks by two variations of high pressure abrasion were attempted. A conventional sandblaster was modified to accommodate polyethylene beads in place of the granular silicate (sand). In this way, solidification of the secondary waste (lead oxide combined with polyethylene beads) could be simply processed by extrusion. However, at 80 psig (the maximum output) there was no apparent oxide removal. When the sandblaster was restored to its normal operation and sand was used as an abrasion medium, a shiny surface resulted, indicating successful oxide removal from the lead brick. Although no quantitative measurements were made during initial feasibility testing, the brick did not appear excessively abraded. Thus, sandblasting may prove to be an effective means of decontaminating lead materials, especially those with large, flat surfaces, e.g., bricks and sheets. Subsequently, a similar process was identified that uses thermoplastic acrylic blasting for removing hazardous paints from aircraft and other delicate surfaces. The acrylic is harder and tougher than polyethylene (Barcol/Shore hardness of 46 to 54 for acrylic compared with 44 to 48 for polyethylene) and thus is better suited for pressure blasting applications. Residual mixtures of acrylic and lead can similarly be processed by melting the plastic to form a monolithic waste form.

5.3 Macroencapsulation

For radioactive lead solids and other hazardous (or mixed waste) debris, EPA has identified two treatment options as the Best Demonstrated Available Technology (BDAT) under 40 CFR 268.45, "Treatment Standards for Hazardous Debris." These include surface deactivation or removal of radioactive lead portions followed by encapsulation of residuals or direct encapsulation of radioactive lead solids by macroencapsulation. Lead waste that has been identified as activated or contains contamination that is not cost-effectively decontaminated can be treated for disposal by macroencapsulation. This process involves surrounding the lead waste

in a clean layer of polymer such as low-density polyethylene to reduce potential for dispersion and leaching. Polyethylene macroencapsulation is currently being commercialized at Envirocare of Utah with support from the DOE Office of Technology Development. Approximately 500,000 lbs of lead from throughout the DOE complex (including 10,000 lbs from BNL) will be treated by polyethylene macroencapsulation at Envirocare's commercially licensed mixed waste facility in Clive, Utah. BNL's Environmental and Waste Technology Center has provided technical support to Envirocare for start-up and licensing of the process.

Polyethylene macroencapsulation consists of compactly packaging large pieces of waste not suitable for processing through the extruder (i.e., microencapsulation) and extruding a clean layer of plastic around it. Pieces of lead are placed in a mesh cage that is suspended inside a drum. Clean polyethylene is extruded into the annulus between the cage and drum. When cooled, the lead is macroencapsulated within a plastic "cocoon" that isolates the waste from the environment, provides structural stability and is resistant to leaching.

Preliminary macroencapsulation processing of lead in polyethylene was conducted. A one quart friction top paint can was used as the outer mold (drum). The friction top portion of the can was cut away to increase the diameter of the can's opening. A metallic mesh basket was filled with lead wool and/or lead turnings and suspended on the inside of the can. Samples of non-radioactive lead supplied by the BNL Safety & Environmental Protection Division were used for macroencapsulation testing. The basket (4"dia. x 4"ht.) was formed from galvanized mesh (6mm x 6mm), creating a 3/4" annulus between the inside of the can and the basket. The height of the basket was 1 1/2" less than the height of the can. This basket dimension allows for an approximately 3/4" outer layer of polyethylene. Later runs were conducted using a slightly larger scale 1 gallon can.

Polyethylene was introduced into the annulus between the basket and the can via a Davis-Standard 1.5" diameter extruder. The temperature profile throughout the extruder was 300°F and the process was operated at a constant screw speed of 20 rpm. The can and lead filled basket were placed on a turntable which rotated the annulus at slightly more than 2 rpm. A rotating drum ensures even delivery of the polyethylene around the annulus.

Several initial runs were conducted to investigate methods of suspending the waste cage to ensure even distribution of the polyethylene layer. The first test was performed using a 3/4" layer of Epoline wax beads lining the bottom of the can to support the lead filled basket. Since the lower molecular weight wax has a lower melting point than polyethylene, the objective was to melt the wax beads by means of the heat transferred from the hot polyethylene, creating a monolithic encapsulation. When this proved unsuccessful, the outside of the can was insulated with woven fiberglass tape and a second encapsulation test was performed. This too, failed to melt the Epoline wax. Additional runs were attempted using pre-molded polyethylene support strips. Strips of varying thickness and geometries were attempted. Vertical supports allowed successful flow under the cage, but residual heat was not sufficient to melt the supports and form a continuous plastic layer. A "cold joint" formed between the polyethylene supports and the newly poured polyethylene layer. Increasing extruder profile temperatures so that Zones 1 - 4

were at 300°F. Zone 5 (closest to the outlet) was 350°F and the outlet die temperature was 375°F improved flow but was still not sufficient to melt the polyethylene supports. In some cases, the molten polyethylene solidified prior to completely covering the bottom of the can.

A different approach was attempted by suspending the basket from above with a swivel joint that allowed the basket to rotate with the drum. A 3/4" annulus was maintained between the bottom of the basket and the can. Using the same conditions as above, this method completely encapsulated the lead filled basket with no insulation on the outside of the can. Other trials were conducted using a polyethylene with a higher melt index.

Following each test run, samples were cross-sectioned to examine the polyethylene outer layer and the degree of polyethylene penetration into the cage and lead. Figure 4 is a photograph of a typical bench-scale sample of lead wool macroencapsulated in polyethylene. In all of the tests conducted, there was some penetration of the basket and lead with polyethylene. Relative effectiveness of the polyethylene in penetrating internal void spaces was estimated by filling the void with water, measuring its volume, and comparing with the original void space. For a one gallon specimen encapsulated with a 50 g/10 min melt index polyethylene, and a pre-formed polyethylene plastic support for the waste cage, the void space was reduced from 85% prior to treatment to an estimated 42% following macroencapsulation, for an overall improvement of 51%. Because the bottom of the cage was supported by pre-formed polyethylene, no penetration of the bottom of the cage occurred. Another sample was tested with 50 g/10 min melt index polyethylene, suspending the cage by a swivel from above. In this case the bottom of the cage was penetrated and the void space was reduced 64%, from 92% prior to treatment to 33% following macroencapsulation. A third sample prepared using a 168 g/10 min melt index polyethylene, suspended from above resulted in a similar reduction in void space of 64%, indicating little benefit achieved by the lower viscosity polymer. These differences can vary depending on the compaction of the lead turnings/wool and the melt flow characteristics of the polyethylene which, in turn depend on the type of polymer and extrusion processing temperatures. Improvements in void penetration may be attained by optimizing polymer melt viscosity and the use of auxiliary heating.

Figure 4. Photograph of a typical bench-scale sample of lead wool macroencapsulated in polyethylene.

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6. SUMMARY AND CONCLUSIONS

A review and evaluation of the literature indicates that a number of physical and chemical methods are available for decontamination of lead. Many of these techniques have been applied for this purpose with varying degrees of success. Methods that apply mechanical techniques are more appropriate for lead bricks and sheet which contain large smooth surfaces amenable to physical abrasion. Lead wool, turnings, and small irregularly shaped pieces would be treated more effectively by chemical decontamination techniques. Either dry abrasion or wet chemical methods result in production of a secondary mixed waste stream that requires treatment prior to disposal.

Evaluation of potential activation products and a radiological survey of random lead samples stored in Building 650 revealed that, in some cases, a unique gamma spectroscopy "fingerprint" can be used to identify lead that has been activated. For example, lead samples that contained isotopes of Cs, Sr, and Co are considered contaminated and thus, are subject to potential decontamination processing. Samples containing isotopes of the activation product Bi-207, however are poor candidates for potential decontamination. Based on preliminary findings, a gamma-scanning survey of lead would be useful for screening and identifying materials that are either clean, contaminated, or activated.

Results of bench-scale investigation of decontamination methods resulted in identification of several wet chemical methods that were very effective in removing layers of lead oxide from the surface, and thus would be likely candidates for decontamination processing. Most effective was the hydrogen peroxide + 10% acetic acid solution. For all solutions tested, considerable removal efficiency was achieved using ultrasonic agitation. Attempts at dry abrasion decontamination were not successful using low-density polyethylene beads, but silica sand blasting was effective in removing lead oxide from the surface of bricks. Thermoplastic acrylic has been used effectively to remove paint from aircraft and other delicate surfaces. Dry abrasion methods are applicable to lead bricks and sheet which have large flat surface areas.

Polyethylene macroencapsulation was successfully demonstrated for the treatment of contaminated lead as an alternative to decontamination, or for cases where decontamination is either not technically or economically feasible. This process is being commercialized by Envirocare and will be available for disposal of contaminated DOE lead. BNL is currently considering shipping of 10,000 lbs of contaminated lead to Envirocare under this DOE sponsored initiative.

It is readily apparent from this evaluation that various wet chemical and dry abrasion can be used to remove radioactive contamination from lead, thus enabling potential recycling of the material or disposal as hazardous rather than mixed waste. However, the ultimate decision on whether or how to decontaminate lead currently stored at BNL or treat it for disposal will require consideration of process economics which were beyond the scope of this study.

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