

A Novel and Cost Effective Approach to the Decommissioning and Decontamination of Legacy Glove Boxes – Minimizing TRU Waste and Maximizing LLW Waste – 13634

Daniel Pancake*, Cynthia M. Rock*, Richard Creed*, Tom Donohoue**, E. Ray Martin**, John A. Mason**, Christopher J. Norton***, Daniel Crosby*** and Thomas J. Nachtman****

*Argonne National Laboratory, 9700 South Cass Avenue, Lemont, IL 60439,

**ANTECH Corporation 9050 Marshall Court, Westminster, CO, 80031

*** Environmental Alternatives, Inc., 149 Emerald Street, Suite R, Keene, NH 03431,

****InstaCote, Inc., 160 C. Lavoy Road, Erie, MI, 48133

ABSTRACT

This paper describes the process of decommissioning two gloveboxes at the Argonne National Laboratory (ANL) that were employed for work with plutonium and other radioactive materials. The decommissioning process involved an initial phase of clearing tools and materials from the glove boxes and disconnecting them from the laboratory infrastructure. The removed materials, assessed as Transuranic (TRU) waste, were packaged into 55 gallon (200 litre) drums and prepared for ultimate disposal at the Waste Isolation Pilot Plant (WIPP) at Carlsbad New Mexico. The boxes were then sampled to determine the radioactive contents by means of smears that were counted with alpha and beta detectors to determine the residual surface contamination, especially in terms of alpha particle emitters that are an indicator of TRU activity. Paint chip samples were also collected and sent for laboratory analysis in order to ascertain the radioactive contamination contributing to the TRU activity as a fixed contamination. The investigations predicted that it may be feasible to reduce the residual surface contamination and render the glovebox structure low level waste (LLW) for disposal. In order to reduce the TRU activity a comprehensive decontamination process was initiated using chemical compounds that are particularly effective for lifting and dissolving radionuclides that adhere to the inner surfaces of the gloveboxes. The result of the decontamination process was a reduction in the TRU surface activity on the inner surfaces of the gloveboxes by four orders of magnitude in terms of disintegrations per unit area (DPA). The next phase of the process involved a comprehensive assay of the gloveboxes using a combination of passive neutron and gamma ray scintillation detectors and a shielded and collimated high purity Germanium (HPGe) gamma ray detector. The HPGe detector was used to obtain gamma ray spectra for a variety of measurement positions within the glovebox. The spectra were used to determine the TRU content of the boxes by assessing the activity of Am-241 (59 keV) and Pu-241 (414 keV). Using the data generated it was possible for qualified subject matter experts (SME) to assess that the gloveboxes could be consigned for disposition as LLW and not as TRU. Once this determination was assessed and accepted the gloveboxes were prepared for final disposition to the Nevada National Security Site (NNSS) – formerly the Nevada Test Site (NTS). This preparation involved fixing any remaining radioactive contamination within the gloveboxes by filling them with a foam compound, prior to transportation. Once the remaining contamination was fixed the gloveboxes were removed from the laboratory and prepared for transported by road to NNSS. This successful glovebox decontamination and decommissioning process illustrates the means by which TRU waste generation has been minimized, LLW generation has been maximized, and risk has been effectively managed. The process minimizes the volume of TRU waste and reduced the decommissioning time with significant cost savings as the result.

INTRODUCTION

As part of the effort to address and disposition TRU waste and LLW generated from ongoing facility deinventory activities at the Argonne National Laboratory (ANL), it has become necessary to decontaminate and decommission (D&D) a number of gloveboxes that were employed for work with Americium and other radioactive materials. Without any decontamination, the two gloveboxes located in

Argonne experimental facilities would have been treated as TRU waste. After an initial phase of clearing tools and materials from the gloveboxes and disconnecting them from the laboratory infrastructure the disposal would then involve the costly process of dismantling and cutting the gloveboxes into a large number of pieces that would be placed in 55 gallon (200 litre) drums. This process would create a significant number of waste drums, increase the potential health risks associated with size reduction activities, as well as increase the potential for industrial hazards in the work areas and finally increase worker exposure.

The overall goal of setting the different approach was to control contamination and manage the gloveboxes as intact units, as opposed to the very high risks associated with size reducing these items. As these changes to the Project Plans evolved, reducing both the cost and the time for decommissioning the two gloveboxes became clear priorities for the Project team. The first phase of the work involved taking full advantage of the engineering controls and confinement provided by each glovebox and clearing tools and materials from the gloveboxes and disconnecting the gloveboxes from the laboratory infrastructure, as in the original decommissioning strategy. This phase involved carefully characterizing, certifying and packaging the waste directly out of the gloveboxes in accordance with the WIPP CH TRU packaging instructions.

A decontamination phase was then undertaken. The purpose of this phase was to remove and reduce the residual radioactive material, in particular the TRU component in order for the two gloveboxes to be characterized as low level waste (LLW) at the completion of decontamination activities. As LLW the gloveboxes could be dispositioned to an LLW disposal site such as NNSS. This successful outcome would preclude the need for exhaustive size reduction, additional CH TRU packaging activities and costs for preparing additional CH TRU shipments. The process would segregate and separately manage TRU and LLW while bringing a number of initial benefits including; reduced worker exposure, reduced potential for accidents or contamination events, reduced decommissioning time and finally deliver cost savings.

In order to execute the alternative decommissioning strategy, ANL engaged three companies with relevant capabilities and experience in decommissioning and decontamination. ANTECH Corporation, (ANTECH) which has expertise in radiation surveying and waste assay, was engaged to perform the critical assay measurements once the gloveboxes were decontaminated to demonstrate that the reduction in the residual TRU activity meant that they were now LLW. Environmental Alternatives, Inc., (EAI) which has expertise and technology for chemical decontamination of radioactive materials, was engaged to undertake the decontamination of the two gloveboxes. Finally, Instacote Inc., with expertise in surface coatings for stabilising radioactivity, were approached to fill the gloveboxes with structural foam once the decontamination and assay stages were successfully completed prior to shipment off site.

INITIAL CHARACTERIZATION PHASE

ANL staff needed to determine the initial radiological conditions of the two gloveboxes and perform a preliminary calculation to determine if they would likely be Transuranic waste. Smears were taken from a number of locations inside the gloveboxes to establish the level of radioactivity and prepared for analysis at the ANL Analytical Chemistry Laboratory (ACL). Additionally paint chip samples capturing areas of fixed contamination were analysed. The outcome of the analysis and calculations showed that both gloveboxes had specific activity greater than 100 nanocuries per gram (nCi/g) and that without decontamination were destined to be disposed of as Contact Handled TRU (CH TRU). The investigations also indicated that when the surface contamination was removed, it was likely that the gloveboxes could be characterized as LLW.

DEINVENTORY PHASE

A factor which increased the degree of complication of the glovebox decommissioning process was that the units were located adjacent to operating laboratories. In order to mitigate the potential for contamination events from glove or pouch failures during D&D activities, HEPA (high efficiency particulate air) ventilated tents were erected in the two laboratories. After installing the tents, ANL staff were required to wear full tyvek suits and PAPR (powered air purifying respiratory) protection until the potential for contamination events had been mitigated by removing the majority of the radionuclide activity. Additionally, continuous air monitors were installed to monitor the airborne contamination in the work area inside and outside of the tent.

The deinventory process for both gloveboxes involved bagging out waste while recording a narrated video describing each item as it was placed into a drum. This process, while somewhat slow, proved to be invaluable when information was required for the sentencing of the waste drums. Materials removed consisted largely of hand tools, electrical components, lab ware, metal racks, gloves and pouch material. Contamination levels on these items were up to tens of millions of disintegrations per minute (dpm) per unit area and the radioactivity was almost exclusively from alpha emitters.



Figure 1: Workers spraying fixative in the glovebox

After the gloveboxes were empty of the removable items, smears were taken and pouched out and counted with a mini-fiddler instrument through the pouch material to give a baseline of the remaining contamination levels within each glovebox. With hindsight it was realised that a radiation survey with a far field detector such as ANTECH RadSearch [1] would have assisted the subsequent decontamination effort by locating hot spots. This information would have helped in targeting the decontamination effort on activity concentrations.

DECONTAMINATION PHASE

ANL contracted Environmental Alternatives Incorporated (EAI) to support decontamination of the gloveboxes, with the intent to remove enough contamination to be able to sentence them as LLW and shipped out whole. The decontamination process utilized “Rad Release” solution, developed and patented by Idaho National Labs and licensed to EAI. Rad Release has been used around the world in a variety of decontamination applications. Workers inserted nozzles through gloves and sprayed the solution onto the internal surfaces of the gloveboxes, scrubbed lightly with scotch-brite and allowed the solution a dwell time of 30 minutes. Then using the same nozzles that were used to spray “Rad Release”, a 10% solution of nitric acid in demineralized water was sprayed on all the surfaces to dilute the decontamination solution. This solution was then vacuumed using a perastaltic pump located outside the glovebox and connected by means of a hose inserted through a glove finger into a plastic carboy. After three applications of “Rad Release” in each glovebox over the course of approximately five working days, smears were taken in a number of locations inside each glovebox. The resulting decontamination activity reduction factor was approximately 10^4 or a factor of 10,000.

Calculations were performed based on the new activity levels derived from the sampling smears and it was determined that the new specific activity of each glovebox was below the level of 100 nCi/g which is

the TRU/LLW threshold. This result would be confirmed by the assay measurements performed in the later assay and assessment phase of the project.

The solid rags and scrubbing materials were placed in a pan inside each glovebox, mixed with neutralizing agent and then dried and pouched out of the glovebox as transuranic waste. The corrosive liquid transuranic waste was pumped out of the gloveboxes, collected in poly bottles and transferred to a liquid transuranic waste processing line in the on-site Waste Management Facility where the liquid was neutralized, solidified and packaged in accordance with the CH TRU packaging instructions.

REMOVING VENTILATION FROM THE GLOVEBOXES

After the completion of the deinventory and decontamination phases, the gloveboxes had to be completely stripped of all external piping and ports, including those connected to the HEPA filtered building air supply. The ANL Health Physics Group calculated that the loose contamination inside the gloveboxes had to be below an average of 2000 dpm to mitigate the possible consequences of a contamination event after the removal of building air. In order to get to these activity levels, nozzles were used through gloves to spray a fixative over the surfaces inside



Figure 2: Ventilation ports after plenum removal

the gloveboxes. After spraying the fixative, the resulting removable contamination levels inside the gloveboxes met the 2000 dpm limit as demonstrated by further sampling.

With the removable contamination at this low level, the in-box HEPA filters were removed and bagged out as the last CH TRU waste. The plenums were removed and blank plates were installed and sealed in place with duct tape. The last step in this part of the process was to remove the tents and allow access without PPE.

ASSAY AND ASSESSMENT PHASE

ANTECH employed a variety of measurement technologies in order to perform both a survey for “hot spots” and the assay of the gloveboxes. Initially the ANTECH QuickSort measurement system was deployed. It is comprised of two, broad energy, plastic scintillator detectors and four, polyethylene shielded, neutron detectors. These independent detector systems are arrayed to permit simultaneous scanning of a section of a glovebox. The scanning protocol presents a single system with both passive gamma ray and passive neutron measurement capabilities. For the glovebox measurements, QuickSort was deployed strictly as a qualitative measurement tool.

The plastic scintillator detectors are highly sensitive to gross gamma ray emission and are therefore quite useful in detecting variations in the gamma ray flux for different measurement locations, in this case, different glovebox sections. This qualitative evaluation permitted the operator to determine the relative homogeneity of gamma ray emission for different measurement locations and to distinguish areas where gamma ray emission is disproportionately higher (hot spots) than other measured areas.

The neutron detector modules are sensitive to thermalized neutrons and therefore are effective in identifying isotopic sources that emit neutrons, such as weapons-grade plutonium, uranium and (alpha,n) emitters. Neutron measurements are very effective in establishing the presence of spontaneously emitted neutrons even from sources that are found in highly shielded configurations, such as motors, compressors, and shielded gloveboxes. Both gamma ray and neutron activity are identified by the QuickSort measurement system. The measurement results serve a triage function and provide an overview of the glove box activity distribution in order to facilitate ultimate quantitative isotopic assay.

Following the initial determination of the distribution of radiation-emitting sources within each glovebox, a high-purity germanium (HPGe) detector was used to analyze the resulting spectra from the remaining radioactivity, using the information obtained by the plastic scintillators to locate the areas within the glovebox for detailed measurements. By this means, the high-resolution spectra were used to quantify the amount of radioactive materials remaining in the entire glovebox. Based on detailed calibration of the germanium-based measurement system, the quantity of remaining radioactive materials were determined to be below the TRU threshold when normalized to specific activity, based on the mass of the gloveboxes. The analysis included a total uncertainty determination [2] which demonstrated that the errors in the measurements, when added to the measured values, resulted in specific activities that were significantly less than the TRU/LLW threshold of 100 nCi/g.

FILLING THE GLOVEBOXES WITH STRUCTURAL FOAM

With confidence gained that the sentencing of the gloveboxes would be LLW, the process of filling void space to comply with disposal requirements could be undertaken. Instacote Inc. was contracted by ANL to fill the gloveboxes with a BASF structural foam that would take up the volume and be acceptable in the waste. Instacote brought specially designed equipment to mix the two part foam in a specific ratio. Using a calculated amount for the initial lift, and then manually topping off the boxes, they were filled to approximately 90% and allowed to set. Once set, the gloveboxes were ready to ship.

PREPARATION FOR SHIPMENT

The two gloveboxes are in the process of being shipped as of the date of writing this paper. A Department of Transport (DOT) type A 7A shipping container will be used to transport the gloveboxes to Nevada National Security Site (NNSS) in two separate shipments. Each glovebox will be blocked and braced in position for the trip with lifting hardware permanently attached so that at the disposal site, they can remove the lid of the shipping container and lift the gloveboxes out and place them in their permanent location.



Figure 3: Loading the First Glovebox for On-site Transport

The first glovebox was moved out to the loading area and lifted into the transportation container and readied for transport. The second glovebox had been decontaminated to SC-1 level, which allowed its transport across the ANL site on a flatbed with only plastic shrink wrap. This glovebox will be stored for the short time while the other glovebox is delivered to NNSS.

OVERCOMING ISSUES

A number of issues arose during the project, and as a credit to the workers, a number of innovative solutions were developed in order to keep the project on track. One of the highlights was removal of the transfer port on the first glovebox. The job was planned based on the transfer port being attached by a series of clamps on the exterior of the glovebox and placing sleeving around the exterior of the port. Once the clamps were removed, the transfer port would be separated from the glovebox using a manual fork truck and then “horsetailing” the plastic and cutting through. Unfortunately, the project team was unaware that the transfer port was attached with a further twelve bolts on the inside of the glovebox, which also included a heavy pressure door.

With this emerging issue, the workers came up with a plan to use a battery impact wrench to remove the bolts while supporting the door with a modified scissor jack. Unfortunately, the bolts were degraded and the team were unable to remove them using this method. Being ever resourceful, the field superintendent contacted the ANL fire superintendent and obtained a burn permit so that an angle grinder could be used inside the glovebox to remove the bolt heads. With grinder in hand and the door supported by the jack, the door and latch were removed, which in turn, allowed the transfer port to be removed.



Figure 4: Grinding Seized Bolt Heads to Remove Transfer Port

CONCLUSIONS

This paper describes the process of glovebox decommissioning at the Argonne National Laboratory. An innovative approach to the decommissioning has been adopted involving the use of a chemical decontamination process to reduce the TRU activity of the gloveboxes to LLW levels. This was followed by a comprehensive assay measurement and a detailed assessment of the assay data by subject matter experts (SMEs) in order to provide definitive evidence and data to demonstrate that the gloveboxes, following decontamination, can be dispositioned as LLW.

A key element of the decommissioning process has been the use of a chemical agent to remove radioactivity (and especially alpha particle emitting TRU) that adheres to the inner surfaces of the glovebox. By this means the majority of the TRU content of the gloveboxes has been removed and concentrated in small containers that can be dispositioned as contact handled TRU (CH TRU) waste. As a result the gloveboxes have become LLW waste resulting in a significant reduction in the volume of TRU waste and in disposal costs.

The submitted manuscript has been created by UChicago Argonne, LLC, Operator of Argonne National Laboratory (“Argonne”). Argonne, a U.S. Department of Energy Office of Science laboratory, is operated under Contract No. DE-AC02-06CH11357. The U.S. Government retains for itself, and others acting on its behalf, a paid-up nonexclusive, irrevocable worldwide license in said article to reproduce, prepare derivative works, distribute copies to the public, and perform publicly and display publicly, by or on behalf of the Government.

WM2013 Conference, February 24 – 28, 2013 Phoenix, Arizona, USA

This work is supported by the U.S. Department of Energy, Basic Energy Sciences, Office of Science, under contract # DE-AC02-06CH11357.

REFERENCES

1. John A. Mason, Marc R. Looman, Adam J. Poundall, Daniel Pancake and Richard Creed, *Development and Testing of a Novel Gamma ray Camera for Radiation Surveying, Contamination Measurement and Radiation Detection*, Proceedings of INMM12, Orlando, Florida, July 2012. (12-A-409-INMM)
2. Currie, L. A. (1968). "Limits for Qualitative Detection and Quantitative Determination, Application to Radiochemistry", *Analytical Chemistry* 40(3):586-593.

ACKNOWLEDGEMENTS

The authors wish to thank the Argonne staff who undertook the deinventory work and the staff of the Argonne Analytical Chemistry Laboratory who analysed the smears.

They also wish to thank Mr R. DelPizzo and Mr M. Piotrowski of ANTECH for their assistance with the preparation of the assay equipment prior to the assay measurements.