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Title: Final Radiological Assessment of External Exposure for CLEAR-Line Americium Recovery Operations

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Final Radiological Assessment of External Exposure for CLEAR-Line Americium Recovery Operations

Los Alamos National Laboratory

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This evaluation was conducted in accordance with RP-3-06-PR-05.2, "Radiological Design Review Procedure." This evaluation meets the requirements of P121, "Radiation Protection, Chapter 12 – Radiological Design" and ISD 341-2, "Engineering Standards Manual Chapter 11, Radiation Protection."

EXECUTIVE SUMMARY

Los Alamos National Laboratory is currently planning to implement an americium recovery program. The americium, ordinarily isotopically pure ^{241}Am, would be extracted from existing Pu materials, converted to an oxide and shipped to support fabrication of americium oxide-beryllium neutron sources. These operations would occur in the currently proposed Chloride Extraction and Actinide Recovery (CLEAR) line of gloveboxes. This glovebox line would be collocated with the currently-operational Experimental Chloride Extraction Line (EXCEL).

The focus of this document is to provide an in-depth assessment of the currently planned radiation protection measures and to determine whether or not further design work is required to satisfy design-goal and ALARA requirements. Further, this document presents a history of americium recovery operations in the Department of Energy and high-level descriptions of the CLEAR line operations to provide a basis of comparison.

Under the working assumptions adopted by this study, it was found that the evaluated design appears to mitigate doses to a level that satisfies the ALARA-in-design requirements of 10 CFR 835 as implemented by the Los Alamos National Laboratory procedure P121. The analyses indicate that extremity doses would also meet design requirements. Dose-rate calculations were performed using the radiation transport code MCNP5 and doses were estimated using a time-motion study developed in consort with the subject matter expert. A copy of this report and all supporting documentation are located on the Radiological Engineering server at Y:\Rad Engineering\2013 PROJECTS\TA-55 Clear Line.

BACKGROUND

There is a continual national need for a domestic supply of americium-241 (^{241}Am), for which the largest customers are oil and natural gas well-logging companies. There are quite a number of additional industrial uses for ^{241}Am including smoke detectors, thickness-gauging measurements; and in nondestructive elemental analysis applications. The available domestic supply of purified $^{241}AmO_2$ was depleted in 2005, and there are many customers who are interested in obtaining these sources. The most common and most easily extractable source of ^{241}Am is reprocessed plutonium-based materials.

Well-logging is critical to assessing the production potential of a well. In fact, most financial institutions only accept data derived from AmBe (americium-beryllium) sources. The Department of Energy (DOE) supports the oil and gas industry in many ways to help ensure the overall security of the petroleum supply. The Rocky Flats Plant (RFP) and Hanford produced ^{241}Am in the 1960s and 1970s, which was primarily extracted from weapons-grade plutonium. In the early 1980s, Los Alamos and the Savannah River Site (SRS) produced 14 kg of ^{241}Am as AmO_2, which when added to the already existing supply of the isotope resulted in quantities of material significantly in excess of industry demands. This supply has been depleted, and currently companies are purchasing ^{241}Am oxide material originating from Russia. The DOE, in concert with industrial partners, has identified a need for re-establishing a domestic supply of ^{241}Am as AmO_2 at Los Alamos.

Both weapons-grade plutonium and reactor grade plutonium isotopic mixtures include a fraction of ^{241}Pu, with reactor-grade having a greater fraction of the heavier plutonium isotopes produced by successive neutron capture. The beta decay of ^{241}Pu (half-life = 14.4 years) to ^{241}Am (half life of 432.7 years) provides a path to high isotopic purity ^{241}Am material in separated plutonium. Thus, the ^{241}Am net in-growth is substantial over time because the ^{241}Am builds up faster than it decays away. A simple view of the pertinent decay chain is shown below.

$$^{241}_{94}Pu \rightarrow {}^{0}_{-1}\beta + {}^{241}_{95}Am \rightarrow {}^{4}_{2}\alpha + {}^{237}_{93}Np \rightarrow \ldots$$

The radioactivity of the source plutonium and resultant americium is such that significant quantities of these materials are amenable to processing in glovebox facilities. Gloveboxes provide contamination control for alpha emitting isotopes along with some gamma shielding, both of which protect the workers. One significantly viable source of americium would be spent salts that were or are produced during molten-salt extraction (MSE), which is used to extract americium from plutonium to help provide pure plutonium feed to other programs like Pit Manufacturing. These MSE residues are unique feed materials for americium recovery, as they have Am/Pu enrichment some 20 fold higher that other residues and also have significant chloride content.

Motivation for Detailed Preliminary Dose Study

^{241}Am is an in-growth daughter product from ^{241}Pu, which is a normal, yet minor, component of plutonium isotopic mixtures. ^{241}Pu (half life of 14.4 years) beta-decays to ^{241}Am. Thus, the ^{241}Am in-growth is relatively rapid. The key exposure concern with unshielded ^{241}Am (half life

of 432.7 years) is that as it decays (alpha decay to ^{237}Np), it produces an intense (high yield) 59.5 keV gamma ray as the excited ^{237}Np daughter nucleus transitions to the ground state. For the shielded ^{241}Am contribution to the dose from 59.5 keV photons is low compare to the contribution from higher energy photons The low-energy gamma ray is relatively easy to mitigate given sufficient shielding. However, if there is little gamma shielding, as in a glovebox window or glovebox glove without incorporation of high Z elements, the dose rates can become a problem. Further, because ^{241}Am decays by alpha emission, (α,n) reactions with light elements make americium oxalate and oxide low grade neutron sources.

The goal of this report is to follow up on the preliminary dose analysis performed in 2012 for the americium recovery project[1] with a more detailed analysis of the source terms and unit operations involved in the project.

Review of Historical ^{241}Am separations efforts

Review of large scale ^{241}Am separations efforts provides some historical background for the CLEAR line operations. A comprehensive review of all ^{241}Am separations efforts is beyond the scope of this paper. Interested readers are encouraged to seek earlier reviews for development and application of the earliest separations efforts to produce mg and g quantities of $^{241}AmO_2$ for R&D purposes.[2,3] Early efforts in separations of kg quantities of $^{241}AmO_2$ have also been reviewed elsewhere.[4]

Rocky Flats Plant

A large number of flow sheets have been run at the Rocky Flats Plant (RFP) over the years. RFP work utilized aged, weapons grade plutonium metal as feedstock, particularly the ^{241}Am rich feedstocks from Am extraction residues associated with purification of Pu metal by chloride-based pyrochemistry. The similarity of feedstocks and separations approaches make the RFP experience particularly important to planned LANL efforts. From 1960 to 1980, kilograms of ^{241}Am were purified at RFP by thiocyanate anion exchange.[5,6] RFP produced the first 200 g of purified $^{241}AmO_2$ to be offered for sale by the ORNL Isotope Sales office.[7] RFP operated an ammonium thiocyanate anion exchange process for ^{241}Am recovery/purification until 1975.

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[1] A. C. Davis and D. A. Gonzalez "Preliminary Radiological Assessment of CLEAR-line Operations", Los Alamos National Laboratory LA-UR-12-21725, 2012

[2] G. T. Seaborg. *"Chapter 2 Transuranium Elements: A Half Century"*, Transuranium Elements: A Half Century American Chemical Society, pp 10-49, 1992. L. R. Morss and J. Fugar Editors.

[3] R. A. Penneman. *"Americium, Its Early History and Gram-Scale Separation"* Americium and Curium Chemistry and Technology, Topics in f-Element Chemistry, D. Reidel Publishing Co, pp 25-33, 1985. N. Edelstein, J. Navratil and W. Schulz Editors.

[4] W. Schulz. *"Chemistry of Americium"* Atlantic Richfield Hanford Co., TID-26971, Technical Information Center Energy Research and Development Administration, 1976.

[5] V. A. Ryan and J. W. Pringle. *"Preparation of Pure Americium"* USAEC Report RFP-130, Rocky Flats Plant, Dow Chemical Company, January, 1960.

[6] L. J. Beach and C. C. Perry. *"Production Scale Americium Recovery at Rocky Flats, 1953 - 1980"* Internal Report CRD 80-023, Rocky Flats Plant, Rockwell International, March, 1980.

[7] J. D. Navratil, W. W. Schulz, G. T. Seaborg. *"The Most Useful Actinide Isotope: Americium-241"* Journal of Chemical Education, Volume 67, Number 1, pp 15-16, January, 1990.

This process was used for recovery of many kg of ^{241}Am , but suffered from large Am losses to waste streams, production of large waste volumes, and dose to personnel. In 1976, simpler and more efficient cation exchange processes (in both chloride and nitrate media) were adopted to replace the thiocyanate process at RFP. Evolution of Am separations flow sheets at RFP progressed towards developing extraction chromatography recovery using bidentate organophosphorous ligands.[8,9,10,11] The RFP specification for purity of AmO_2 was >95 wt. % AmO_2, with < 1 wt. % individual contaminant elements. Pu and Pb were contaminants often carried with the recovered Am in quantities greater than the specification, meaning reprocessing was required.

Hanford

The Hanford site was heavily engaged in separation of Pu and Am isotopic mixtures from irradiated reactor fuel for many years, primarily by the PUREX process. Hanford produced isotopically pure ^{241}Am in significant quantities in the 1960s and 1970s, with work essentially stopping after an accident with ^{241}Am loaded cation exchange resin in 1976.[12] Quantities of pure ^{241}Am were typically about one half RFP production over the same time period. A number of recovery flow sheets were developed and utilized at Hanford, with the later ^{241}Am recovery flow sheet similar to that utilized at SRS (described below).

ORNL

Separations efforts at ORNL have historically been engaged with separations efforts to recover the ^{243}Am isotope, in particular separations of ^{243}Am and ^{244}Cm from irradiated High Flux Isotope Reactor (HFIR) fuels. The ORNL efforts are based on oxalate precipitation of $^{244}Cm^{+3}$ from carbonate solutions with ^{243}Am oxidized to higher oxidation states. Good work was done, but will not be reproduced here due to the significant differences between feedstreams, recovery scale, and general approaches between ^{243}Am and ^{241}Am.

Savannah River Site

Over the years from 1978 to 1981, approximately 8 kg of ^{241}Am were purified (as AmO_2) at Savannah River Site (SRS).[13,14,15] Similar to LANL work, this campaign utilized aged, reactor

[8] A. C. Muscatello and J. D. Navrtil. *"Americium Removal from Nitric Acid Waste Streams"*. RFP Report R04008, 1986.

[9] L. L. Martella and J. D. Navrtil. *"A Combined Anion Exchange-Bidentate Organophosphorous Extraction Process for the Recovery and Purification of Americium and Plutonium from Molten Salt Extraction Residues"*. RFP Report A-4041, 1980.

[10] J. D. Navrtil. *"Plutonium and Americium Processing Chemistry and Technology"*. Inorganica Chimica Acta, 94 (1984) 263-269.

[11] H. Siddall, III, *"Bidentate Organophosphorus Compounds as Extractants I. Extraction of Cerium, Promethium, and Americium Nitrates",* J. Inorg. Nucl. Chem., Vol. 25. pp. 883-892, , October 1962.

[12] For a more complete discussion and references associated with this accident, see the "Resin Safety" section of this paper.

[13] Tracy Rudisill (SRNL), Leonard Gray (LLNL), Bob McQuinn (LANL) Personal Communication.

[14] L. W. Gray, G. A. Burney, T. A. Reilly, T. W. Wilson, J. M. McKibben. *"Recovery of Americium-241 from Aged Plutonium Metal"*. U.S. DOE Research and Development Report DP-1577, E.I.duPont de Nemours & Co., Savannah River Laboratory, Aiken, South Carolina, December 1980. ~5 kg of SRS-recovered ^{241}Am is noted, but this report is likely not comprehensive for all Am produced.

grade plutonium metal as feedstock, and also produced PuO_2 for the Fast Flux Test Facility in Hanford. In addition, recovered PuO_2 was utilized for fabrication of Mark-42 targets to be utilized for production of heavier actinide elements. Also similar to Los Alamos work, this Pu recovery scheme was based on HNO_3 recovery of Pu. The SRS flow sheet differed in that it utilized PUREX separations chemistry to remove the bulk of the Pu, multiple concentration steps, and cation exchange for final concentration and purification of the ^{241}Am prior to precipitation as americium oxalate: about 8 kg total $^{241}AmO_2$ in two campaigns in F Canyon and the MPPF (Multi-Purpose Processing Facility).

The SRS specification for purity was noted as >95 wt. % AmO_2. Cr, Ni and Pb were contaminants often carried with the recovered Am, and blending was sometimes used to meet the specification. The final product $^{241}AmO_2$ average purity was 98%, and recovery efficiency was 98.5%.

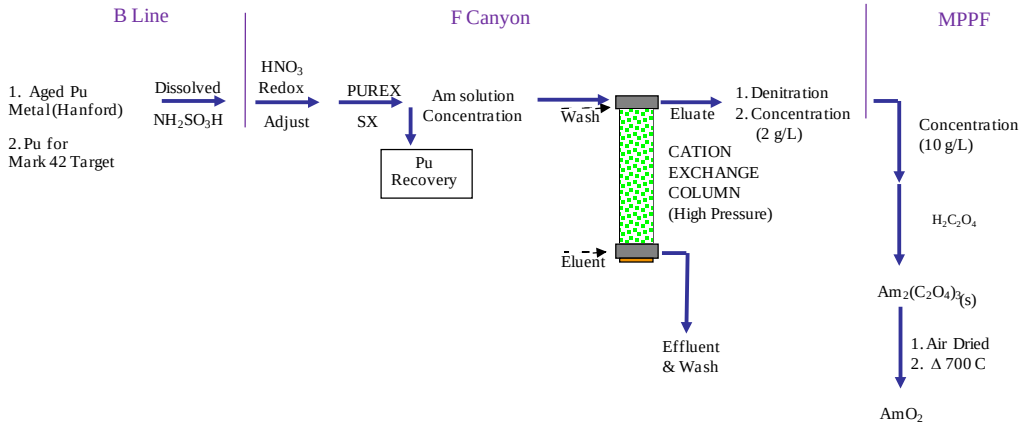

Figure 1. SRNL Flow sheet (simplified) 1979-1981

LANL-Historical

Over the years from 1979 to 1984, 6.5 kg of ^{241}Am were purified (as AmO_2) at Los Alamos National Laboratory.[16,17] This historical work utilized aged, reactor grade plutonium metal as feedstock, and produced PuO_2 for the Fast Flux Test Facility (FFTF) in Hanford, as did much of the SRS reprocessing effort. The FFTF required hundreds of kg of reactor grade PuO_2 product, and a large inventory of "old" weapons grade Pu metal existed from Pu recovery campaigns at Hanford. The LANL flow sheet removed the majority of Pu by peroxide precipitation, and an additional fraction of Pu by anion exchange. Multiple ^{241}Am concentration steps of hydroxide precipitation followed by redissolution, or concentration using an evaporator were utilized

[15] 7.9 kg of SRS-recovered ^{241}Am is noted in: C. A. Mangeng, G. R. Thayer. *"Beneficial Uses of ^{241}Am"*. LA-10074-MS, 1984.

[16] T. Blum, J. Parker, M. Romero, D. Yearwood. *"Americium Oxide Production at LANL: Recovery Up – Exposures Down",* LAUR-83-2962.

[17] H. Ramsey, D. Clifton, S. Hayter, R. Penneman, E. Christensen. *"Status of Americium-241 Recovery and Purification at the Los Alamos National Laboratory"* Transplutonium Elements Production and Recovery, ACS Symposium Series 161, pp 75-91, 1980. J. Navratil and W. Schulz Editors.

prior to precipitation of the ^{241}Am as americium oxalate. No LANL specification for purity was noted in reports, and it is assumed that specifications similar to those in use at RFP and SRS about the same time were in place. LANL observed some difficulty with contamination from silicates originating from glassware exposed to HF/HNO_3 dissolutions. Contaminants in the product $^{241}AmO_2$ were primarily Pb (from shielding) and Pu with occasional high content of Ca and Si. The final product $^{241}AmO_2$ purity was 98 to 99%, and average recovery efficiency was above 90%.

The dosimetry data discussed in the earlier LANL work is of particular value for a comparison to the modeling information presented here. For the final full year (1983) of $^{241}AmO_2$ production at LANL the received dose by glovebox workers was about 10 mrem extremity exposure, and 1 mrem whole body exposure per gram of ^{241}Am produced.[16] The results showed a gradual ~8 fold reduction in extremity exposure, and ~3 fold reduction in whole body exposure per gram of ^{241}Am produced over the years from 1979 to 1983. Attention to operational details, shielding, and worker skill all can clearly contribute to reducing exposure in glovebox operations.

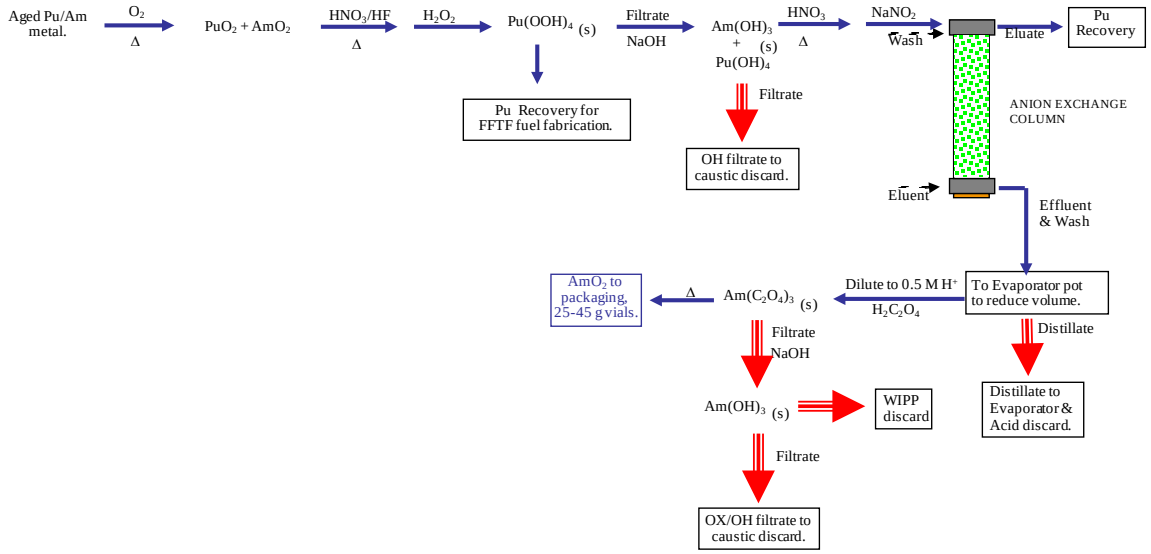

Figure 2. LANL Flowsheet 1979-1984

LANL – Current Plan

The CLEAR glovebox line is comprised of three large gloveboxes, interconnected by square tunnels. The glovebox shells are welded, 7-gauge, 316L stainless steel and include a sandwiched lead layer for improved gamma shielding. The gloveboxes extensively utilize specialty materials to reduce corrosion problems. All glovebox inner surfaces are covered in heavy-sheet PVDF (polyvinylidene fluoride) or ECTFE (ethylene chlorotrifluoroethylene) linings welded in place to provide enclosures that are capable of housing aqueous HCl processing operations. These linings are integral to the inner surface of the push-through HEPA housings, the connection tunnels, and wrap fully around windows and gloveports. Local zone 1 exhaust duct connected to the HEPA housings is PVDF powder coated.

The three CLEAR gloveboxes contain a large capacity (by standards of aqueous chloride processing) of tanks/vessels/columns for staging and handling process solutions more than all four gloveboxes of EXCEL.[18] Specialty materials are utilized in construction of tanks and columns with cylindrical vessels constructed of glass with PVDF endcaps, and 4" slab tanks fabricated of PVDF. Specialty shielding material of either 0.25 g GdO/cm^2 in PVDF and/or 1/8" metal lead encapsulated in PVDF was utilized for some tanks/vessels. All tank capacity for radiological HCl solutions are internal to the glovebox, unlike the large volume tank farms utilized in HNO_3 process areas.

CLEAR Glovebox 1

CLEAR Glovebox 1 was originally designed for "Pu^{+3} column" operations to improve efficiency in solvent extraction operations for feeds unusually high in Fe. With the discontinuation of the mission to reprocess PuBe neutron sources for Pu recovery, the problems with Fe separation in HCl process operations were alleviated. Typical pyrochemistry feeds into HCl process operations contain only small quantities of Fe which have been processed by TBP based solvent extraction without difficulty. Fe does follow Pu in this separations scheme, but significant separation of Pu from small amounts of Fe occurs in the oxalate precipitation step.

CLEAR glovebox 1 can be reconfigured for Pu^{+4} anion exchange operations for direct Pu recovery. Current anion exchange column capacity is 4 columns at 1.25 L each for a total of 5 L of resin. With a nominal capacity of 100 g of Pu per liter of resin volume, the current column configuration is compatible with recovery of Pu at 500 g scale, compatible with a 520 g CSLA (Criticality Safety Limit Analysis). Increasing column capacity by installation of a single large column, or by replacing the 2" diameter (1.25 L volume) columns with 3" (2.8 L volume) would increase column capacity for efficient Pu^{+4} anion exchange operations with an approved 1040 g CSLA for this glovebox.

Reconfiguration of glovebox 1 for Pu^{+4} anion exchange operations would be particularly important for Pu feeds high in ^{241}Am and suitable for ^{241}Am production operations. The anion exchange operations can be performed largely hands-off, reducing dose to personnel for these feeds. Dissolved feed solutions would be transferred from EXCEL Dissolution to glovebox 1 using overhead transfer lines. Purified Pu solutions from IX recovery in glovebox 1 would be transferred to EXCEL Oxalate using overhead transfer lines. The Am-rich anion exchange effluents would be transferred to adjacent glovebox 2 for additional Pu and Am recovery. This study has led to a better understanding of the importance of additional gamma shielding on a number of tanks in CLEAR glovebox 1, primarily to reduce extremity exposure. The change in feedstock required for an ^{241}Am production operation is a significant driver in this case.

[18] **Ex**perimental **C**hloride **E**xtraction **L**ine is a bit of a misnomer. As this glovebox has been in use for nearly 20 years, the solvent extraction technology deployed is now well proven rather than "experimental" as the name would suggest. CLEAR total process tank space is nominally 1400 L, EXCEL 1200 L.

CLEAR Glovebox 2

CLEAR glovebox 2 was designed for secondary Am and Pu recovery from process effluents associated with Pu recovery of electrorefining salts, failed DOR runs and crucibles of all kinds. These effluents contained up to 3 g of Am, and in the case of occasional process upsets, 10s of g of Pu. Pyrochemistry Am extraction residues were not given large consideration as a significant feed in design of this glovebox.

Each side of CLEAR glovebox 2 has space for 4 columns of 1.25 L volume capacity, with a plan that 2 columns would be filled with anion exchange or TRU resin (for Pu recovery) and 2 columns would be filled with resin utilizing $(4\text{-t-Bu}C_6H_4)_2P(O)CH_2C(O)N(i\text{-Bu})_2$ as an extractant for trivalent actinides (Pu^{+3} and Am^{+3} recovery). A practical loading capacity of about 10 g of Am per liter of resin column volume without breakthrough has been observed. Each resin $(4\text{-t-Bu}C_6H_4)_2P(O)CH_2C(O)N(i\text{-Bu})_2$ column of 1.25 L volume represents a nomination capacity of about 12 g Am. Observed flow rates for process solutions in tandem column arrangements with these columns and materials were 1-2 L per hour. No additional acid molarity or REDOX adjustment was typically utilized in demonstration runs or planned for solutions in CLEAR glovebox 2.

CLEAR Glovebox 2 can be reconfigured and operations optimized for better efficiency in larger scale americium recovery. The columns on each side of CLEAR glovebox 2 will be changed to provide a single column filled with anion exchange resin (for Pu recovery) and 3 columns filled with resin utilizing $(4\text{-t-Bu}C_6H_4)_2P(O)CH_2C(O)N(i\text{-Bu})_2$ as an extractant for trivalent actinides (now primarily for Am^{+3} recovery). Process upsets containing more Pu than the capacity of a single 1.25 L anion exchange column will be handled in glovebox 1. Additional REDOX adjustment will be performed on process effluents to hold Pu in the tetravalent state prior to flow through the tandem column arrangement. Each side of CLEAR glovebox 2 will have a maximum Am capacity of about 36 g Am (3 columns at 12 g Am capacity each).

Purified Pu solutions from anion exchange columns in glovebox 2 would be transferred to EXCEL Oxalate using overhead transfer lines. Purified Am solutions from extraction chromatography columns in glovebox 2 would be transferred to a glass 6” diameter cylindrical tank (CLEAR GB 2-OXPPT-C9) to be used for Am oxalate precipitation.

CLEAR glovebox 2 offers opportunity for flexibility in operations. For example, all the EC columns on both sides could be utilized for simultaneous treatment of a very rich (above 35 g Am) feed solution split between the two sides. Americium oxalate filtrates may be treated by hydroxide precipitation or recycled on extraction chromatography columns as desired. A CSLA limit of 520 g for the whole glovebox is anticipated. Additional shielding will likely be utilized in this glovebox.

CLEAR glovebox 3

CLEAR Glovebox 3 was designed for hydroxide precipitation and filtration operations. No need for reconfiguration of existing design has become apparent during this review for larger scale Am recovery. Solutions to glovebox 3 are anticipated to be primarily from glovebox 2. A CSLA

limit of 520 g for the whole glovebox is anticipated. No upgrade to shielding is anticipated for glovebox 3, as column effluent solutions are anticipated to be low in both Pu and Am before receipt in this glovebox.[19] However, the secondary background radiation in the vicinity of this box is of enough concern that the time spent working in it must be taken into account. Thus, a gross estimate of the time spent will be multiplied by the background dose rate at the face of the box. The secondary background dose rate is higher than that for the other boxes because glovebox 3 is located near a safe containing several hot materials.

CLEAR Glovebox 4

Glovebox 4 will house the ^{241}Am oxalate-to-oxide-calcination furnace as well as oxide weighing and blending operations. This box is fabricated entirely of type 316 stainless steel (with 3/16" wall thickness) and 1/4" lead shielding in the wall encapsulated by 1/16" type-316 stainless steel. It is assumed also in this glovebox that ¼" thick leaded glass windows are at the bottom viewport. The box will contain an Inconel crucible for use in the furnace with 0.5" wall thickness, an open top and a nominal outer diameter of 4.5" long, 3" wide and 2.25" deep.

Assumptions

As a reference, this section presents a list of assumptions that were made to perform this analysis.

1) Yearly dose is assumed to be based on the processing of 25 batches per year.
2) The production of 25 batches per year (750 g/y) will require 3.75 workers, at a minimum.
3) From assumptions 1 and 2, yearly dose is calculated based on an assumption of 3.75 exposed workers per year.
4) Maximum yearly throughput of americium-241 is 750 g.
5) All CLEAR processes are collocated in the same room.
6) Exposure time for secondary background dose is considered to be the time in the room for the whole-body calculation.
7) Exposure time for secondary background dose is considered to be the time out of gloves for extremity.
8) The time-in-room estimate used for the whole body calculation is the sum of extremity distances 1 and 2 and the time out of gloves.
9) The production of 17 batches per year (500 g/y) will require 2.75 workers, at a minimum.
10) All americium sources contain 30 g americium regardless of the mass of the product form (oxalate, oxide, etc).
11) The final CLEAR-line glovebox configuration is similar to the configuration analyzed in this study.
12) The time and distances estimated in the time-motion study are assumed to accurately reflect the average times and distances involved in the actual operations performed by average operators.
13) Whole-body dose at the glovebox surface is assumed to be the personal dose equivalent averaged over a 30x30 cm area 1 inch from the glovebox surface.
14) Material transfer from the PF-4 vault to the work area is considered beyond the scope of the CLEAR line operations.
15) For Glovebox 1, there are two exposure cases. In case 1, the entire source is contained by the slab tank. In case 2 the source is split between the Slab tank (18.1L) and the filter boat (1.9L).
16) The dissolution process is considered beyond the scope of CLEAR line operations.

[19] Americium oxalate filtrate solution may be either recycled through extraction chromatography columns or neutralized. If neutralized, it is anticipated that one 6" cylindrical precipitation vessel will be kept dedicated for this purpose.

17) Dose rate associated with the transfer of Pu solution is assumed to be equal to that associated with am solution transfer. (Conservative)
18) Source term in GB 2 is 5 g Pu and 30 g Am. This refers to 30 g of americium, not 30 g of product (oxalate or oxide) Columns and cylindrical tanks are glass with PVDF endcaps. Thickness of PVDF column endcaps 1/2" PVDF.
19) Nominal 6" glass columns are 6.073" ID, 6.698" OD, and wall thickness 0.312" in field measurements.
20) Nominal 2" glass columns are 2.010" ID, 2.335" OD, and wall thickness 0.162" in field measurements.
21) Shielding on the glass cylindrical tanks and columns (where used) are 1/8" Pb, sandwiched between two layers of PVDF of 0.06" thickness.
22) The borosilicate glass (brand-name Pyrex) has a density of 2.23 g/cm^3 at 20 C and a composition of :

Constituent	% by weight
SiO_2	81%
B_2O_3	13%
Na_2O	4%
Al_2O_3	2%

23) Slab tanks are PVDF with wall thicknesses of 0.5 inches.
24) Shielding on the PVDF slab tanks (where used) are PVDF with 0.25 g/cm^2 Gd_2O_3; In GB 1 slab tank shielding is 3/16" tin.
25) PVDF/GdO_2 layer is 0.30" thickness, sandwiched between two layers of PVDF of 0.06" thickness.
26) PFVD density at 20C is 1.77 g/cm3
27) Assume installed 5/16" thick, 2mm Pb equivalent glass windows cover 3 lower levels of glovebox windows.
28) 30 mil leaded hypalon gloves have 0.1 mm Pb equivalent.
29) For conservatism, 30 mil gloves are used in the assessment of extremity doses for boxes 1 and 3.
30) 65 mil gloves were assumed for the calculation of extremity doses during the oxalate handling and transfer operation.
31) Inconel Crucible is shielded with 304 stainless, wall 0.0625" wall thickness, 12" ht, 4.88" OD diameter.
32) High-fired porcelain is $Si_3Al_4O_{12}$ with density of 2.40 g/cm^3, 0.25 inch wall thickness, 7.3 inch inner diameter and is 5 inches tall
33) Glovebox shell sheet lining is PVDF of minimum thickness of 90 mil.[20]
34) Lining will match the shape of the 7 gauge Type 316 stainless steel glovebox shell inner surface.
35) Gloveboxes 1 have the following properties:

Component	Value
Shell – Type 304 SS	3/16" = (0.47625 cm)
Pb Shield core	1/4" = (0.635 cm)
Shield surface Type 304SS Layer (surrounding ¼" lead layer)	1/8" = (0.3175 cm)
Top Type 304SS Layer	3/16" = (0.47625 cm)
Bottom Type 304SS Layer	3/16" = (0.47625 cm)
Inside GB Kynar lining	0.090 mil = (0.23 cm)
Neutron Shielding	None
Viewing windows	5/16" leaded glass = (0.79375 cm)
Viewport Covers on lower level only	1/4" lead encapsulated by 1/8" Stainless Steel
Slab Tank Shielding	1/8" Tin
Glove thickness	30 mil

[20] Specification No NMT8-PS-11617-R00

36) Glovebox 2 has the following properties

Component	Value
Shell – Type 316 SS	3/16” = (0.47625 cm)
Pb Shield core	1/4” = (0.635 cm)
Shield surface Type 304SS Layer (surrounding ¼” lead layer)	1/8” = (03175 cm)
Top Type 316SS Layer	3/16” = (0.47625 cm)
Bottom Type 316SS Layer	3/16” = (0.47625 cm)
Inside GB Kynar lining (not modeled)	0.090 mil = (0.23 cm)
Neutron Shielding	None
Viewing windows	5/16” leaded glass = (0.79375 cm)
Viewport Covers on lower level only	1/4” lead encapsulated by 1/8” Stainless Steel
Glove thickness	30 mil

Neutron shielding (PVDF lining) was omitted from the glovebox 2 model. This assumption provides a conservative assessment of the neutron dose. The actual clear-line gloveboxes 1 and 2 will be lined with PVDF.

Assumptions for Calcination-furnace Glovebox

37) Glovebox 4 is location for ^{241}Am oxalate to ^{241}Am oxide calcination furnace, and oxide weighing/blending.
38) Glovebox 4 is fabricated entirely of 316 stainless steel, wall thickness of 3/16" thick (7 gauge).
39) GB wall includes 1/4" Pb shielding to bend line above top of large viewing window, encapsulated by 1/16" 316 stainless steel.
40) Assume installed 1/4" thick "standard" Pb glass windows cover the lower viewports.
41) 30 mil leaded hypalon gloves have 0.1 mm Pb equivalent used for extremity dose assessment.
42) Design of Inconel 625 crucible for calcination is 0.5" wall thickness; open top, nominal OD 4.5" long, 3" wide, 2.25" deep.
43) Chemical resistant shielding of future tanks and radiation source shielding in CLEAR gloveboxes will be 1/4" pewter inside 1/8" PVDF.
44) Analytical Chemistry operations and the transfer of sample are assumed beyond the scope of this operation.
45) Whole-body dose from blending operations is assumed to be equivalent to the dose from 30 g of americium in a closed inner container in the calcination glovebox.

Validity of selected assumptions

Most of the assumptions made in this analysis are based on equipment specifications and values quoted by the subject matter expert. This section examines the validity of the broader assumptions underlying this study.

One of the fundamental assumptions used in calculating the yearly dose is that 25 batches will be processed each year. Under the further assumption that each batch contains 30 g of ^{241}Am this would lead to the production of 750 grams of ^{241}Am per year, which exceeds the expected 500 g/year output. Individual batch size will vary depending on the source material from the vault. Assuming 25 30-g batches per year allows for variation in total output without danger of exceeding the dose limit.

In defining the scope of this analysis, it was decided that both the transfer of the source material from the PF-4 vault to the work area and the dissolution process were beyond the scope of the CLEAR-line radiological analysis. This is because material transfer to the EXCEL line (where dissolution occurs) is currently in practice and is unlikely to be modified to accommodate CLEAR-line operations.

For the purposes of this assessment, the dose rates from gloveboxes 1 and 2 are assumed to be equal. The glove boxes differ primarily in size and source term. Glovebox 1 is larger than glovebox 2 and the source in glovebox 1 is contained in slab tanks vs. extraction columns in glovebox 2. Both gloveboxes 1 and 2 process 30 g of americium, but 1 processes 1 kg of plutonium as opposed to 5 g of plutonium through 2. Because the plutonium is expected to be predominantly ^{239}Pu, it is expected to contribute little to the total source term and the additional material in GB 1 will likely provide additional self-shielding relative to the source in GB 2. Further, as discussed in the assumptions section, the structural materials and radiation shields are assumed to be equivalent for these two boxes. Given all of these considerations, equating the dose rates from these two boxes was considered a reasonable assumption.

The time-motion study is assumed to be an accurate reflection of the workers' average actions in these processes. Considering that these processes are similar to other processes that are currently in practice, these data are likely to be realistic. Though the time-motion data quotes two distances for use in whole body calculations, the assumption was made that all work time (the sum of times spent at distances 1 and 2) in gloveboxes 2 and 4 is spent at about 1" from the glovebox face. This is a conservative estimate because time spent at a further distance would reduce the worker's dose. Credit was taken for the time spent at distance two in the analysis of glovebox 1 in an attempt to keep the primary dose from the three modeled processes less than 1 rem/year.

PROCESS STEPS

The primary process steps in the CLEAR line are described below. EXCEL-line processes are considered, for radiological engineering purposes, to be beyond the scope of the americium recovery project and thus, this study. Figure 3 presents a flow sheet describing the integration of the CLEAR and EXCEL glovebox lines.

Primary Plutonium Separation

This process occurs in CLEAR glovebox 1 and is fed by the EXCEL dissolution process.

+4 Pu Feed Treatment for Plutonium Separation: This process consists of the addition of HCl to achieve an 8 M solution and the addition of sodium nitrite ($NaNO_2$) or sodium chlorite ($NaClO_2$) to place the plutonium in solution into the +4 valence state. The process will occur in feed tanks.

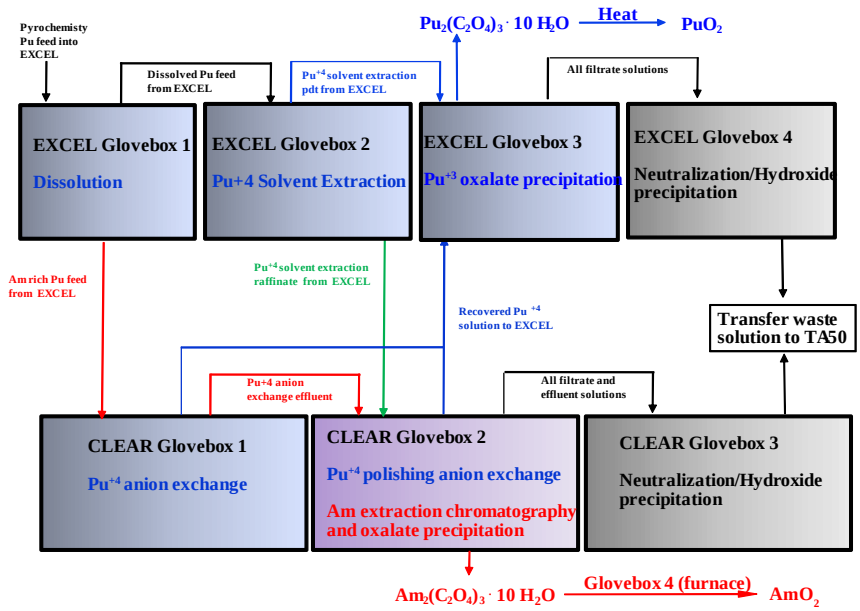

Figure 3. Flowsheet showing the integration of the CLEAR and EXCEL glovebox lines. Glovebox 4 is referenced in red at the bottom of the flow diagram

+4 Pu Chloride Anion Exchange: This process consists of the ion exchange process whereby the plutonium in the +4 valence state is loaded onto the column and the americium passes through into the effluent. The column is washed, which is collected with the effluent, and then eluted to remove the plutonium from the column, which is then sent to EXCEL. The process will occur in anion exchange resin columns. This process separates the bulk of the plutonium (99%) from the americium.

Secondary Plutonium Separation and Americium Extraction

+4 Pu Feed Treatment for Americium Purification: This process is essentially the same as describe above for plutonium separation, although it occurs in a different location. The process will occur in feed tanks before the anion exchange/extraction chromatography operation.

Pu Polishing Anion Exchange/Am Extraction Chromatography: The anion exchange and extraction chromatography columns are connected in series for the load and wash cycles, but are eluted separately. The first process is essentially the same as the chloride anion exchange describe above (with smaller volumes of eluent and wash) and essentially removes the last remnants of plutonium from the solution, which are then sent to EXCEL. All of the above processes are either already performed in some other fashion in PF-4 or are minor variants of similar processes already performed in PF-4. In addition, the processes described above have been previously used with existing operational equipment to achieve ~99% separation of alpha activity.[21]

[21] Schulte, L. D., R. R. Salazar, and S. D. McKee, "Use of Extraction Chromatography to Recover Americium and Plutonium from Hydrochloric Acid Effluent Streams," Los Alamos National Laboratory, LA-UR-09-7664, 2009.

Americium Precipitation

Am Oxalate Precipitation: The eluate from extraction chromatography is combined with oxalic acid dihydrate, which precipitates the americium as americium oxalate. The oxalate filtrate containing a small amount of americium is sent to hydroxide precipitation or recycled onto extraction chromatography columns, and the filtered americium oxalate is sent to calcination. The oxalate precipitation occurs in the same glovebox as the anion exchange and extraction chromatography operations.

Am Hydroxide Precipitation: The effluent and wash from extraction chromatography and filtrate from oxalate precipitation are combined with potassium hydroxide (KOH), which precipitates the trace americium contents which are sent to waste handling. The filtrate is send to liquid waste treatment. The hydroxide precipitation occurs in a dedicated glovebox for CLEAR waste processing. Because of the small amounts of plutonium and americium in this solution, it has been deemed to be negligible from a radiological standpoint and has therefore not been modeled in this analysis.

Americium Oxalate Calcination

Am Oxalate Calcination: The americium oxalate cake is placed in a calcination furnace that is ramped to ~650ºC and held for 6 h (assume 24 h cycle time for ramp up and cool down) and converted to a pure oxide. The calcination will occur in a small muffle furnace in a dedicated glovebox. This box also includes the unit operations of weighing and blending the oxide after it has been removed from the furnace.

DOSE MODELS

Methods

Several computer software packages were used in the analysis of the various exposure cases. Source terms were calculated using Sources4C for neutron sources and RadSrc for photons. Transport calculations were performed using the Monte Carlo N-particle transport code (MCNP5). Each of these software packages are discussed briefly in the following sections.

Source Term Calculations

Sources 4C

Neutron source terms were calculated using Sources 4C. The Sources 4C code calculates neutron production rates and spectra due to spontaneous fission, delayed neutron emission and (α,n) for four different problem configurations (interface, homogeneous, beam, and three-region interface problems). Spontaneous fission spectra are calculated with evaluated half-life, spontaneous fission branching, and Watt spectrum parameters for 44 actinides. The (α,n) spectra are calculated using an assumed isotropic angular distribution in the center-of-mass system with a library of 107 nuclide decay a-particle spectra, 24 sets of measured and/or evaluated (α,n) cross sections and product nuclide level branching fractions, and functional a-particle stopping cross sections for Z<106 . The delayed neutron spectra are taken from an evaluated library of 105 precursors. The code provides the magnitude and spectra, if desired,

of the resultant neutron source in addition to an analysis of the contributions by each nuclide in the problem. [22]

RadSrc

Photon source terms were calculated using the RadSrc application. The RadSrc application is based on a "library" file that is suited for implementation into Monte Carlo codes. The library code computes the concentrations of decay products given an initial concentration and age, as well as the photon source spectrum and strength due to the continuing decay of these products. The standalone application exists as a user interface to the library file.[23]

Transport Calculations

MCNP5

The shielding calculations for the individual exposure cases were performed in the Monte Carlo N–Particle code, MCNP5. MCNP is a general-purpose Monte Carlo transport code that can be used for neutron, photon, electron, or coupled neutron/photon/electron transport. The code indirectly solves the modified Boltzmann transport equation (known as the neutron or particle transport equation) by the Monte Carlo method of simulating individual particles and recording some aspects (tallies) of their average behavior. The average behavior of particles in the physical system is then inferred (using the central limit theorem) from the average behavior of the simulated particles. [24]

For dose rates and fluence calculations, F4 and F6 tallies were used. F4 and F6 tally calculate dose rate averaged over a defined volume of interest. MCNP initially calculates fluence of particles. Fluence is a stream of particles crossing a unit area, and is expressed as the number of particles per second per 1 cm2. In order to convert particle fluence to dose to humans, fluence to dose conversion factors (DCF) are used in conjunction with F4 tally results. Radiological Engineering adopted dose conversion factors from ICRP Publication 74, "Conversion Coefficients for use in Radiation Protection against External Radiation" for estimating worker doses.

The generation of the photon DCFs for transport code input requires use of two separate tables in ICRP 74. The first table (Table A1) converts fluence to Air KERMA and the second table converts Air KERMA to dose. Table A24 was selected to be used in conjunction with Table A1 for dose rate calculations at RP-3. For neutron DCFs, Neutron Fluence-to-Personal Dose Equivalent dose conversion factors for monoenergetic neutrons from ICRP 74 Table A.42 were used.

[22] Wilson, W.B., et al., "SOURCES 4C: A Code for Calculating (alpha,n), Spontaneous Fission, and Delayed Neutron Sources and Spectra," Los Alamos National Laboratory, LA-UR-02-1839, 2002.
[23] Hiller, L., Gosnell, T., Gronberg, J., Wright, D., "RadSrc Library and Application Manual," Lawrence Livermore National Laboratory, UCRL-TM-229497, 2007.
[24] X-5 Monte Carlo Team, "MCNP- A General Monte Carlo N-Particle Transport Code, Version 5: Volume 1: Overview and Theory," Los Alamos National Laboratory, LAUR-03-1987, 2003 (Rev. 2008).

For calculation of the absorbed dose a heating tally F6 was used. This tally converts the fluence in the tally cell to absorbed energy by multiplying each scoring fluence track by the cross section and heating number in MeV per collision (KERMA factor). The raw tally units in MeV g-1 were converted to give the absorbed dose rate in rad hr-1

Case Development

Once the unit operations of a given process are defined by the subject matter expert, one of the first tasks facing the radiological engineer is to prioritize the unit operations in terms of hazard. In some cases, such as with hydroxide precipitation, the activity of the material involved in the process is so small that the hazard is negligible. In cases using higher-activity or for which simple shielding implementation is difficult, the radiological engineer is able to develop new tools or practices that will reduce dose to workers. In the present study, initial models of extremity and whole body dose led to the several modifications of current tools as well as development of new tools.

Hand Shield for Americium Oxalate Handling

Americium oxalate precipitation requiring the operator to handle the porcelain filter boat containing 30 g of americium (in the form of americium oxalate) was studied with respect to extremity dose. Figure 4 shows a picture of the filter boat and its representation in MCNP.

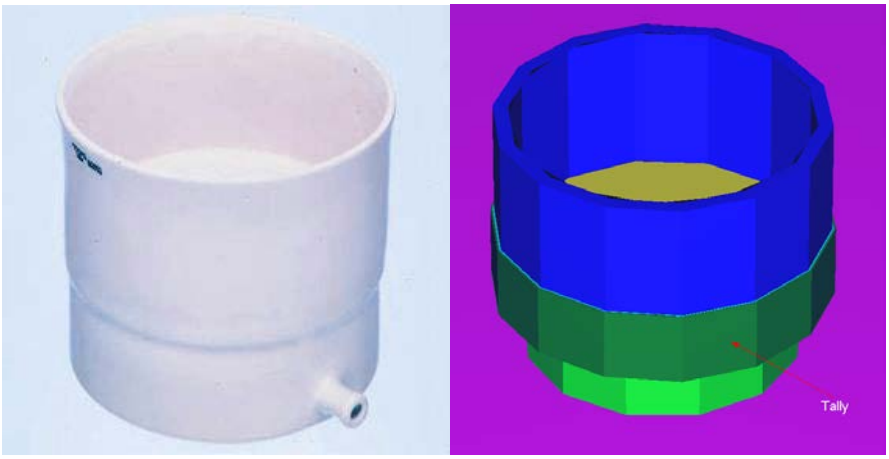

Figure 4. Porcelain filter boat (left) and MCNP model of porcelain filter boat (right)

The dose rates at the surface of this container were calculated for four cases: no shielding with no leaded gloves, no shielding with 30 mil leaded gloves, no shielding with 65 mil gloves and 1/8” lead shielding with 30 mil leaded gloves. These results are shown in Table 1. The actual CLEAR line gloveboxes will use 65 mil gloves; the use of 30 mil gloves represents a limiting case.

Table 1. Dose rates in mrem/hr on surface of filter boat containing 30 g americium (in oxalate form)

Exposure Case	Dose rate (mrem/hr)
1/8"Pb shielding, 30 mil leaded gloves	5.2×10^{1}

NO shielding, 65 mil gloves	9.3×10^3
NO shielding, 30 mil leaded gloves	2.1×10^4
NO shielding, NO leaded gloves	4.4×10^4

The dose rates were worse directly above the filter boat; with 30 mil gloves, the dose rate was 20 rem/hr at 5 cm from the top of the filter boat and 8.9 rem/hr at 10 cm above the boat. To mediate the dose rate when the operator must have his hand directly over the top, a stainless steel hand-shield was modeled and studied for its dose-reducing properties. Ultimately, a titanium hand shield was also developed and modeled. An MCNP model of the hand shield and the filter boat is shown in Figure 5.

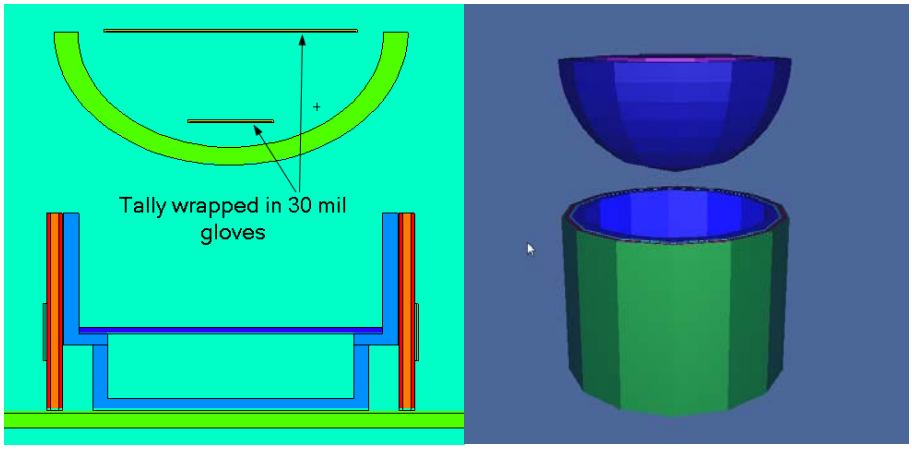

Figure 5. Steel hand shield implemented with 30 mil leaded gloves (shown as tallies in the leftmost plot)

The effect of various thicknesses of hand shields for stainless steel is presented in Table 2.

Table 2. Effect of stainless steel hand shield and 30-mil leaded gloves on extremity dose above Am oxalate filter boat

With 30 mil leaded gloves				
Dose rate, mrem/hr	No shield	0.5 cm shield	0.35 cm shield	0.15 cm shield
5 cm from top	2.0×10^4	3.80×10^3	7.60×10^3	1.60×10^4
10 cm from top	8.90×10^3	1.40×10^3	2.95×10^3	6.10×10^3
weight of shield, grams		718	299	151

The dose rate on top of the 0.9525 cm thick hand shield, 11 cm from the source, is 319 mrem/hr. Analyses of this type will allow for the implementation of tools and practices that minimize worker dose.

Exposure Cases

As previously discussed, the modeling effort focused on dissolution, anion exchange, extraction chromatography/americium precipitation and oxide calcinations. Hydroxide precipitation was omitted from the analysis because the hydroxide solution contains less than 1% of plutonium or americium and is thus of negligible concern.

Dissolution unit Operation

Anion Exchange Unit Operation

Anion exchange occurs in glovebox 1 shown in Figure 7.

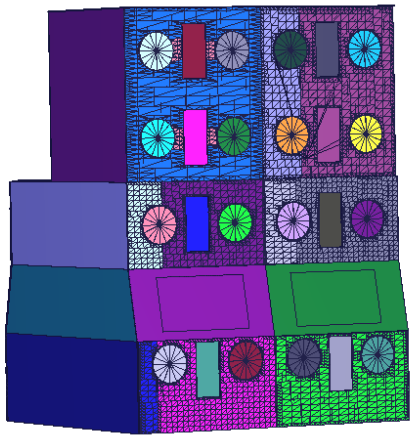

Figure 6. 3D image of the MCNP model representation of CLEAR glovebox 2

The source term for this case is 20 L of a solution containing 7 M hydrochloric acid (HCl), 1000 g of plutonium and 30 g of americium. Two broad exposure cases were selected for this unit operation:

Case 1: The total volume (20 L) of the solution in a slab tank (CLEAR GB 1-IX-S1).

Case 1A: The volume split between the slab tank and a high-fired porcelain filter boat in front of this tank with 9.5% of the material in a boat and 90.5% of the material in a tank. The boat was modeled at 30 cm from the front panel of the glovebox. The second case is modeled with and without a cylinder of 1/8” lead sandwiched between two layers of PVDF of 0.06” thickness, 12” tall. Figure shows an elevation view of the MCNP model of this case.

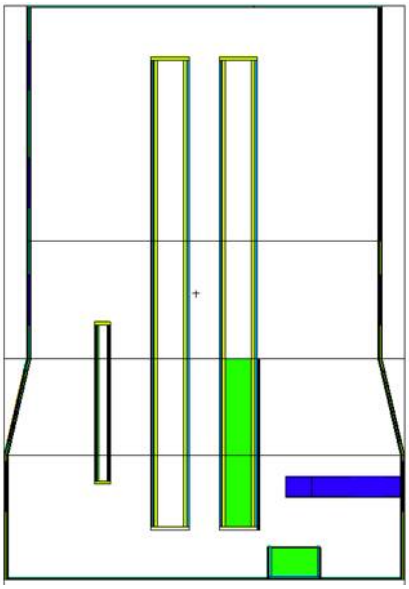

Figure 7. Elevation view of Glovebox 1 model for Case 1A. The blue rectangle represents an arm tally inside the glovebox at 10 cm from the slab tank

Extraction Chromatography and Americium Precipitation

These processes take place in CLEAR glovebox 2. The MCNP model of CLEAR glovebox 2 is represented in Figure 8.

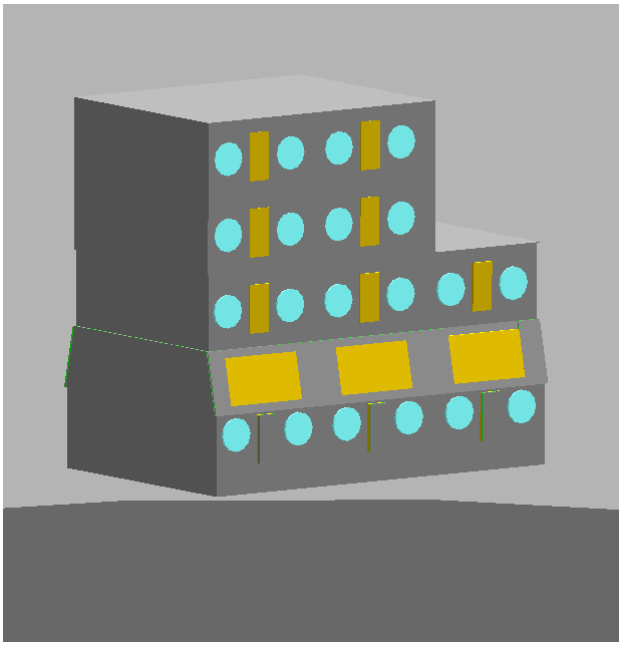

Figure 8. 3D image of the MCNP model representation of CLEAR glovebox 2

There are several separate source terms for this unit operation describing the column before and after precipitation, the case when the americium oxalate solids are in the filter boat, and when the solids are loaded into the extraction chromatography columns.

Case 2: Americium loaded onto 2 extraction chromatography columns with each containing 15 g of americium and 5 g of plutonium in 7 M HCl.

Case 3: The total volume (10 L) of the solution is in the precipitation column (CLEAR GB 2-OXPPT-C9). The source in this case is 30 g of americium distributed uniformly through the 10 L solution of 1 M HCl.

Case 4: Similar to Case 3, the total volume of the solution is in the precipitation column. However, the americium oxalate precipitate has settled to a 0.5 inch layer at the bottom of the column with the supernatant above.

Case 5: The americium oxalate solids are in the high-fired porcelain filter boat (shown on the left side of Figure 4) in front of the column CLEAR GB 2-OXPPT-C9. This case is expected to be of most concern with respect to extremity dose. Therefore, doses at several additional extremity distances were calculated.

Case 5A: Exactly the same as case 5 except with the assumption of no shielding on the filter boat to model the extremity exposure resulting from the transfer of the oxalate solids.
Case 6: Americium solids in an Inconel crucible with shielding consisting of a ¼ inch cylinder of pewter sandwiched between two 0.06 inch layers of PVDF. This case is similar to case 5 with the filter boat substituted with the Inconel crucible.

Calcination and Packaging
These cases model americium oxide in the Inconel crucible as well as shipping containers in various configurations. Figure 9 shows the MCNP model of the crucible inside the calcination furnace in glovebox 4.

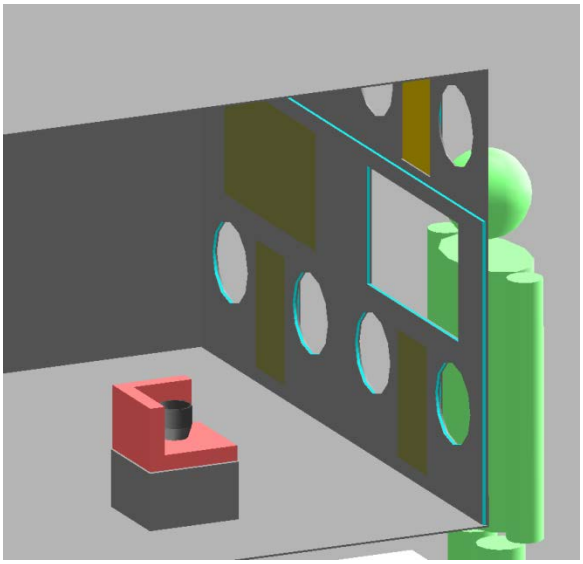

Figure 9. Inconel Crucible inside calcination furnace in glovebox 4. Two of the furnace walls and the furnace top are cut away for illustration

Case 7: 30 g of americium (in the form of AmO_2) in an Inconel crucible. This case models the step between removing the oxide from the calcination furnace and transferring it to the inner shipping container. This step was modeled in MCNP as shown in Figure 10.

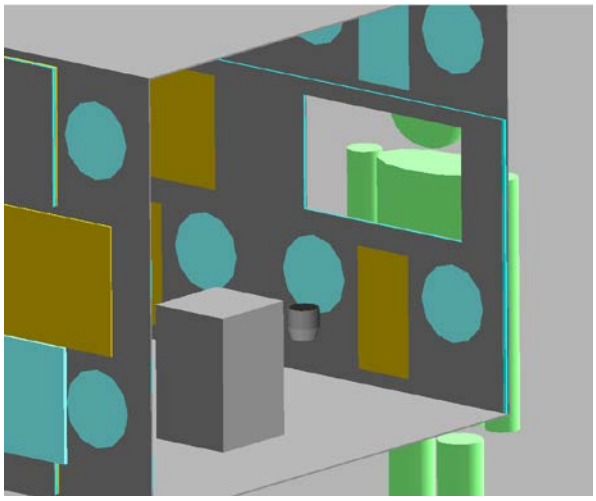

Figure 10. Model of glovebox 4 with Inconel crucible outside of the calcination furnace

Case 8: 30 g of americium (in the form of AmO_2) in the innermost shipping container with lid on. The inner shipping container is shown in Figure 11.

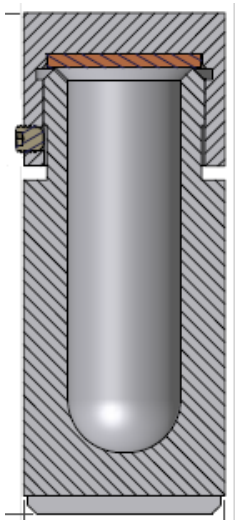

Figure 11. Innermost shipping container

For the transfer of the oxide from the crucible to the inner container, the inner container will be held stable in a temporary glovebox fixture referred to as a "pig that's a jig" (or pig/jig). Figure 12 shows an MCNP model of this pig/jig in front of the calcination furnace in glovebox 4.

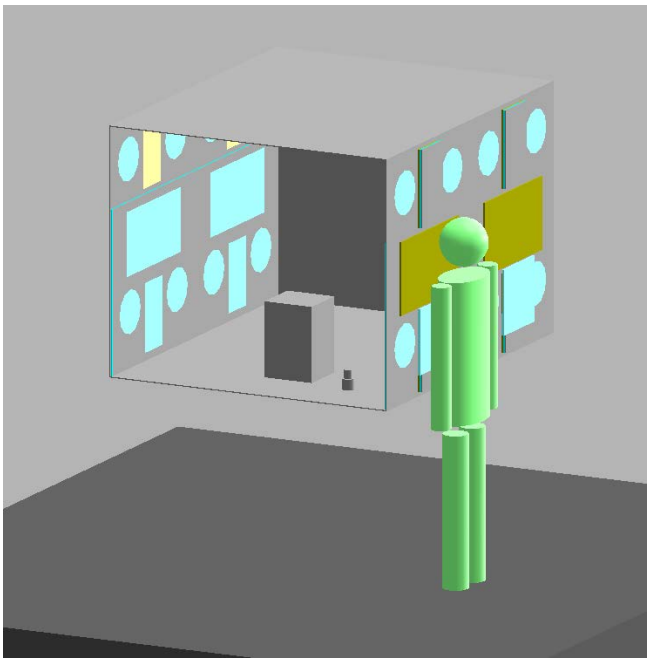

Figure 12. MCNP model of the "pig that's a jig" holding the inner container in front of the calcination furnace in glovebox 4

Case 9A: 30 g of americium (in the form of AmO_2) in the innermost shipping container with additional shielding and lid off.

Case 9B: 30 g of americium (in the form of AmO_2) in the innermost shipping container with additional shielding and lid on.

Case 9C: 30 g of americium (in the form of AmO_2) in the innermost and middle shipping containers

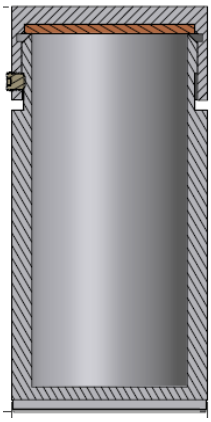

Figure 13. Middle shipping container. Innermost container fits inside

Case 9D: 30 g of americium (in the form of AmO_2) in the innermost and middle shipping containers in SAVY container. SAVY containers are used to temporarily store and transfer objects, such as the AmO_2, internally.

Case 9: Blending Operations. 30 g of americium (in the form of AmO_2) in the innermost container in the calcination glovebox.

TIME-MOTION STUDY

Motivation for Time-Motion Study

An important piece in the development of an occupational radiological assessment is the development of a time-motion study. Time-motion studies are based on experts' estimation of the amount of time required to perform the unit operations comprising a larger process (henceforth "time data") as well as and the actual, physical location of the operator's trunk and extremities ("motion data"). These data can be used to estimate production rates or can be combined with dose calculations to estimate the total dose incurred by performing an entire cycle of operations. Time-motion data is also instrumental in initial phases of the analysis as the information provided by the subject matter experts often aids the analyst in the development of exposure cases.

Development of the Time-Motion Study

In many cases, time-motion studies are performed for operations which have been in practice for some time. In these cases, the study-development process typically consists of successive interviews with multiple subject matter experts involved with each step of a given process. This is an iterative process where the analyst and subject matter expert continue to modify the data until a consensus is reached. An example flow sheet for the development of a typical time-motion study is shown in 14.

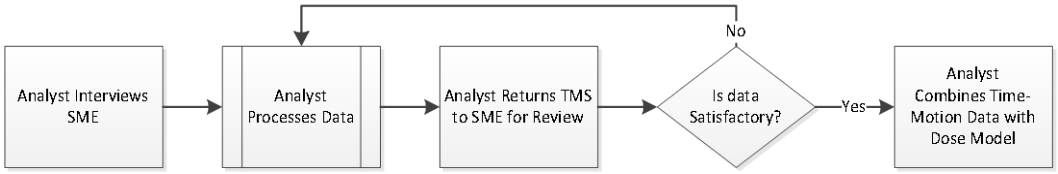

Figure 14. Example flow sheet for the development of a time-motion study for operations currently in practice

With proposed operations such as those to be performed in the CLEAR line, typically there are fewer individuals with the experience to estimate the time and motion data. Because these processes are still at various phases of development, time-motion studies serve an important role in the engineering of the total system. For these cases, it is important that close contact be maintained between the radiological engineers and the subject matter expert. An example flow sheet for this case is shown in Figure 15.

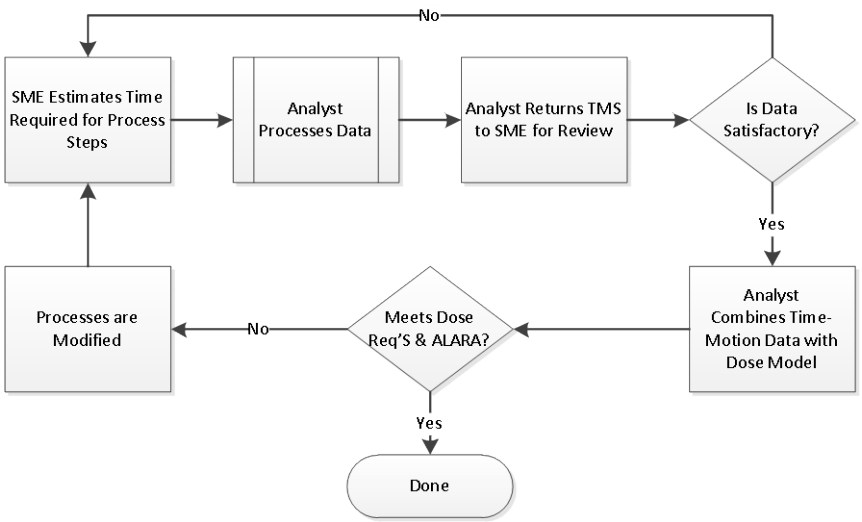

Figure 15. Example flow sheet for the development of a time-motion study for proposed operations

In the current project several unit operations intended for the CLEAR line are currently operational, though in other contexts. Thus estimates of time and motion data for these processes are subject to less uncertainty in estimation than if these operations where purely notional.

Inputs for time-motion study

The time and motion data describe time spent at various distances from either the source (for extremity dose) or the glove box face (for whole body exposure). The extremity data present times spent at two and ten inches from the source performing different manual tasks. Two inches from the source is taken to be the surface of any applicable container. In the case of glovebox 1, when the source was split between the slab tank and the filter boat in glove box 1, the distances were taken as 2 and 10 inches from the slab-tank face at the height of the glove ports. The whole body data present times spent at one inch and one foot from the source. Times estimated at one inch from the glovebox face represent the time the worker spends with hands in the glovebox while times estimated at two feet from the glovebox typically represent times spent observing the glovebox with hands out of gloves. For some unit operations, a large majority of the operator's time is spent away from the glovebox. These times are also estimated as part of the time-motion study and the dose rates during these times (both whole body and extremity) are estimated using the typical background dose rate in the room. The conservative assumption was made that dose rates at the one foot from the glove box face were the same as those at the surface. For glovebox 1, in the case where the source was split between the tank and the filter boat, the one inch dose rate was taken as the dose rate at the gloveport nearest to the filter boat and the two feet dose rate was taken as the dose rate between the gloveports.

Results of Time-Motion Study

Introduction and Dissolution

The first activity performed is the transfer of the feed material from the PF-4 vault to the area where the processes will occur. The material is removed from the vault by cart, and is moved to the room. The material is then placed on the trolley by way of the introduction hood. The trolley transfers the material from the intro hood to the work environment. Because these processes are currently in practice and are beyond the scope of the CLEAR line, they are not analyzed from a time-motion perspective. With the exception of the analysis of glovebox 1, the conservative assumption is made that, in calculating whole-body dose, all time is spent at the closest distance defined in the time motion study. The various distances are, however, used for the extremity calculations for all gloveboxes.

Primary Plutonium Separation

Primary plutonium separation takes place in glovebox 1 and consists of feed treatment to put the plutonium into the +4 valence state followed by two anion exchange processes to remove plutonium from the solution and purify the americium. The time and motion data for primary plutonium processing is shown in Table 3.

Table 3. Time and motion data for primary plutonium processing. Times listed are in hours

	Extremity			Whole Body		
Unit Operation	2 inch	10 inches	Out of gloves	1 inch	1 foot	Room background
Chemistry Adjust	0.05	0.5	1	0.55	0.5	1.55
Filtration	0.05	0.5	1	0.55	0.5	1.55
Column Prep	0.05	0.5	1	0.55	0.5	1.55
Column Load	0.1	1	3	1.1	1	4.1
Column Wash	0.05	0.5	2	0.55	0.5	2.55
Column Elution	0.05	0.5	2	0.55	0.5	2.55
Pu Soln. Transfer	0.05	0.1	1	0.15	0.1	1.15
Am Soln. Transfer	0.05	0.1	1	0.15	0.1	1.15

Secondary Plutonium Separation and Americium Extraction Chromatography

Further purification of actinides from the process effluent solution is performed in glovebox 2 by plutonium anion exchange followed by extraction chromatography which removes and purifies the vast majority of americium (>99%). The time and motion data for these processes are shown in Table 4.

Table 4. Time and motion data for secondary plutonium processing and americium extraction chromatography

	Extremity			Whole Body		
Unit Operation	2 inch	10 inches	Out of gloves	1 inch	1 foot	Room

						background
Chemistry Adjust	0.1	0.1	1	0.2	0.1	1.2
Column Prep	0.1	0.1	1	0.2	0.1	1.2
Column Load	0.2	0.4	3	0.6	0.4	3.6
Column Wash	0.1	0.1	2	0.2	0.1	2.2
Pu Column Elution	0.1	0.1	2	0.2	0.1	2.2
Am Column Elution	included with above operation					0
Pu Soln. Transfer	0.1	0.1	1	0.2	0.1	1.2

Americium Oxalate Precipitation

After extraction chromatography, the purified americium eluate solution is combined with oxalic acid dihydrate. This precipitates the americium from the eluate as americium oxalate. This process takes place in the same glovebox as secondary anion exchange and extraction chromatography (GB 2). The time and motion data for the americium oxalate precipitation unit operation is shown in Table 5.

Table 5. Time and motion data for americium oxalate precipitation. Times shown are in hours

	Extremity			Whole Body		
Unit Operation	2 inch	10 inches	Out of gloves	1 inch	1 foot	Room background
Chemistry Adjust	0.05	0.1	2	0.15	0.1	1.15
Filter Boat Prep	included with above operation					
Reagent Addition	0.05	0.1	1	0.15	0.1	1.15
Digestion	0.05	0.1	1	0.15	0.1	1.15
Filtration	0.05	0.1	1	0.15	0.1	1.15
Washing	included with above operation					
Air Dry	0.01	0.1	3	0.11	0.1	2.11
Am oxalate handling	0.01	0.1	0	0.11	0.1	0.11
Am oxalate transfer	0.1	1	2	1.1	1	3.1

Americium Calcination and Oxide Handling

The americium oxalate cake precipitated from the precipitation unit operation is transferred to a separate glovebox containing a calcination furnace where it is converted to americium oxide. After calcinations, the oxide is weighed, blended and sampled. The time and motion data for calcination and oxide handling is shown in Table 6.

Table 6. Time and motion data for calcination and oxide handling processes. Times shown are in hours

	Extremity			Whole Body		
Unit Operation	2	10	Out of	1	1	Room

	inch	inches	gloves	inch	foot	background
Oxalate Calcination						
Loading Furnace	0.01	0.1	0	0.11	0.1	0.11
Furnace Cycle	0	0.05	2	0.05	0.05	2.05
Unloading Furnace	0.05	0.1	1	0.15	0.1	1.15
Am_2O_3 handling						
Weighing in crucible	0.01	0.1	1	0.11	0.1	1.11
Weighing in innermost C.	0.05	0.1	1	0.15	0.1	1.15
Blending	0.01	0.1	2	0.11	0.1	2.11
Combine/Split	0.05	0.2	2	0.25	0.2	2.25
Sampling	0.01	0.1	1	0.11	0.1	1.11

Americium Hydroxide Precipitation and Calcination

The effluent and wash from extraction chromatography and filtrate from oxalate precipitation are combined with potassium hydroxide (KOH), which precipitates the trace americium contents which are sent to waste handling for calcination. The filtrate is send to liquid waste treatment. The hydroxide precipitation occurs in a dedicated glovebox for CLEAR waste processing. The times and distances for the hydroxide precipitation operations are shown in Table 7.

Table 7. Time and motion data for americium hydroxide precipitation. Times shown are in hours

	Extremity			Whole Body		
Unit Operation	2 inch	10 inches	Out of gloves	1 inch	1 foot	Room background
Reagent Addition	0.05	0.1	1	0.15	0.1	1.15
Filter Boat Prep	included with above operation					
Digestion	0.05	0.1	0.5	0.15	0.1	0.65
Filtration	0.05	0.1	1	0.15	0.1	1.15
Washing	included with above operation					
Air Dry	0.01			0.1	1	0.11
Am hydroxide handling	0.01	0.1	0	0.11	0.1	0.11
Am hydroxide transfer	0.1	1	0.5	1.1	1	1.6

The times and distances for the hydroxide calcination unit operation are shown in Table 8.

Table 8. Time and motion data for calcination and oxide handling processes. Times shown are in hours

	Extremity			Whole Body		
Unit Operation	2 inch	10 inches	Out of gloves	1 inch	1 foot	Room background

Loading Furnace	0.01	0.1	0	0.11	0.1	0.11
Loading Furnace	0.01	0.1	0	0.11	0.1	0.11
Furnace Cycle	0	0.05	1	0.05	0.05	1.05
Unloading Furnace	0.05	0.1	1	0.15	0.1	1.15

Material Out

After the oxide has been handled, it is removed from the glovebox work environment and returned to the vault to await shipping. This consists of placing the oxide batch in the trolley, bagging the batch out, and carting it back to the vault. The time and motion data for these steps is shown in Table 9. The whole-body dose for the case when the source is outside of the glove box was taken as the personal dose equivalent at 13.5" from the inner-container surface.

Table 9. Time and motion data for removing the batch from the glovebox line. Times shown are in hours

	Extremity			Whole Body		
Unit Operation	2 inch	10 inches	Out of gloves	1 inch	1 foot	Room background
Trolley	0.1	0.2	1	0.3	1	1.3
Bagout	0.05	0.1	0.5	0.15	-	0.65
Bagout	0.05	0.1	0.5	-	-	0.65
Cart	-	-	0.5	-	-	0.5
Vault	-	-	0.25	-	-	0.25

Results

This analysis finds that appropriate radiation protection measures are in place to maintain occupational doses to workers in the CLEAR line below current dose design objectives. This analysis is considered to be conservative, so fluctuations in yearly production are unlikely to be of concern from a radiological perspective.

Several assumptions were made when calculating the doses and comparing them to the time-motion data. For the GB2 and GB 4, the whole-body doses at both distances from the gloveports were taken to be the dose at one inch from the viewport between the glove ports with the cover off. For GB 1, the doses were calculated at 1 inch and one foot from the maximally affected port. For GB 2 this was taken as the viewport between the glove ports with the viewport cover off. For the calcination Glovebox, doses were taken at one inch from the leaded view port in front of the source. Secondary background dose was assumed to be equal to 1 mrem/hr. The time motion study indicates that 81.82 hours will be spent per operation in the room with the americium process. If 25 batches are to be made, this results in 2045.5 hours per year and a background dose of 2045.5 mrem/year whole body. When the 8 hrs per operation (200 per 25-operation-year) from glovebox 3 is considered, this number jumps to 2245.5 mrem/year. For secondary background dose to the extremities, the time out of gloves was taken as the exposure time. The exposure time for glovebox 3 is higher for whole body

than for extremities because it is assumed that the 4 hours spent working in the box will shield the arms from the background, but not the trunk.

Whole-body results

Table 10 shows the contact times and calculated whole-body doses for each process in the CLEAR line. It is clear that the background dose is the largest concern with respect to whole body. The total dose from all operations, including background dose is 3,325.5 mrem/year. The glovebox doses represent the conservative case of doses at the central viewport without a viewport cover. This assumption is conservative because, during normal operations, the viewport will have a cover installed. Time and dose per operation in the table refer to the time spent and the dose incurred in processing one batch. The "yearly" values are the times and doses under the assumption of 25 batches per year.

To illustrate the relative demand of each operation in terms of contact time and dose, Figure 16 and Figure 17 show pie charts of the worker time per operation and whole-body dose per operation respectively.

Note that though americium oxalate calcination and americium oxide handling are relatively short operations from a contact-time perspective, they are two of the highest-dose operations and demand the most attention from radiological engineering. Conversely, the "material out" operation is relatively lengthy but results in one of the lowest doses.

It is here noted that glovebox 3 is located in an area of locally higher secondary background due to a material safe in close proximity. This report recommends that this safe be moved to a location to reduce the secondary background to workers in this area. The relocation of this safe would also reduce the secondary background to workers at the other gloveboxes.

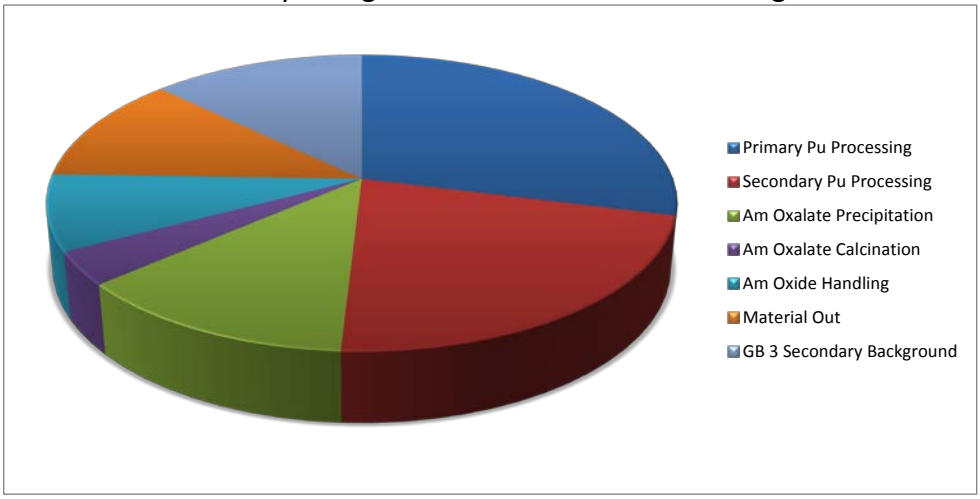

Figure 16. Worker Time (in hours) per operation for a single batch

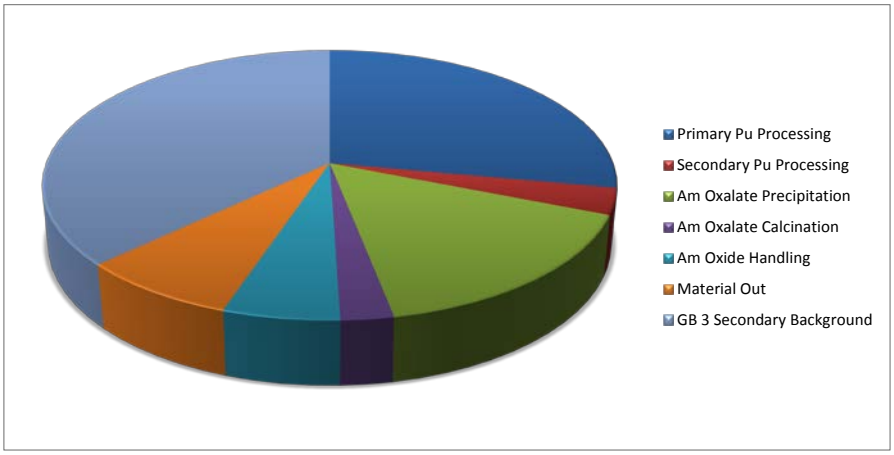

Figure 17. Whole Body Dose per operation for a single batch

Table 10. Total times and whole-body doses for CLEAR line operations broken down by glovebox. "Yearly" values assume 25 operations per year. Time includes all time spent in the room, not just contact time

Glovebox	Operation	Time per batch (h)	Yearly time (h)	Dose per batch (mrem)	Yearly dose (mrem)
Glovebox 1	Primary Pu Processing	16.15	403.75	17.632	440.8
	Glovebox 1 Total	**16.15**	**403.75**	**17.632**	**440.8**
Glovebox 2	Secondary Pu Processing	11.6	290	1.935	48.38
	Am Oxalate Precipitation	9.92	248	9.764	244.09
	Glovebox 2 Total	**21.52**	**538**	**11.699**	**292.47**
Glovebox 4	Am Oxalate Calcination	3.42	85.50	1.575	39.38
	Am Oxide Handling	7.73	193.25	3.484	87.104
	Material Out	3.35	83.75	4.788	119.7
	Glovebox 4 Total	**14.5**	**362.5**	**9.847**	**246.184**
	Sum of all Gloveboxes	**81.82**	**2045.5**	**39.18**	**979.5**

Glovebox 3	Secondary Background	12[25]	300	26.8	**670**
	Total Secondary Background	**93.82**	**2346**	**93.82**	**2,346**
	Sum with background	**-**	**-**	**133.0**	**3,325.5**

Extremity Dose

Extremity dose is of most concern in these operations. For extremity dose from slab tanks, detectors were placed inside the glovebox at the surface and 10" from the tank face. For cylinder tanks, cylindrical detectors were placed at similar locations surrounding the tank. For the "material out" operation, MCNP calculations were performed at the surface of and 10" from the inner container. The other cases were assumed to have dose rates equal to those associated with the filter boat shielded by 1/8" lead and 30 mil leaded gloves. Dose rates were calculated at the surface of the filter boat and at a distance of 10 cm at a height equal to the center of the source inside the boat.

Discussion and Conclusions

The purpose of this study was to expand upon the preliminary dose assessments performed for the Americium Recovery Project in 2012, both in scope and complexity of analysis. By combining detailed geometry and source term models of unit operations with extensive time-motion data, this report presents the most accurate estimation of the Americium Recovery Project's expected doses to date.

Table 11 reports the sum of the yearly doses from all CLEAR line gloveboxes to the extremities as 13,877mrem/year. LANL dose design objective for extremities is 10 rem/year. When the actual number of people participating in a project is considered, the annual dose to the extremities remains below P121 limits.

Table 11 presents the extremity doses per batch and per year (assuming 25 batches per year) for each set of operations in the CLEAR line. Under the current assumptions, it is estimated that the total extremity dose associated with 1 batch would be 543 mrem leading to a yearly dose of 13,877 mrem under the conservative assumption of 25 batches per year. As discussed before, the 30-mil thick leaded gloves were assumed for operations in gloveboxes 1 and 3. Operations in glovebox 2 were modeled using 30-mil gloves, except for the americium oxalate handling operation, which uses 65-mil gloves. The use of 65-mil gloves is being discussed in the other gloveboxes. However, more detailed information is required to assess whether or not the use of 65-mil gloves will significantly affect process times; the dexterity of the operators will be

[25] 4 hours is spent in a 4.7 mrem/hr dose field (3.5 gamma, 1.2 neutron) and 8 hours is spent in a 1 mrem/hr dose field (0.5 mrem/hr gamma and 0.5 mrem /hr neutron)

compromised by the use of lead-lined gloves that are over twice as thick. Longer process times and decreased dexterity will increase exposure time and thus dose. It remains to be seen whether or not this increase in exposure time will lead to doses comparable with 30-mil gloves.

Table 11. Doses to the extremities per batch and per year for each operation broken down by glovebox. Yearly values assume 25 batches per year. Times presented in "Sum with Background" row represent the time spent out of gloves

Glovebox	Operation	Time per batch (h)	Yearly time (h)	Dose per batch (mrem)	Yearly dose (mrem)
Glovebox 1	Primary Pu Processing	4.15	103	101.5	2536
	Glovebox 1 Total	**4.15**	**103**	**101.5**	**2536**
Glovebox 2	Secondary Pu Processing	1.6	40	8.106	202.6
	Am Oxalate Precipitation	1.92	48	157.5	3938
	Glovebox 2 Total	**3.52**	**88**	**165.606**	**4141**
Glovebox 4	Am Oxalate Calcination	0.42	10.5	23.87	596.7
	Am Oxide Handling	0.73	18.25	94.52	2363
	Material Out	0.6	15	94.13	2353
	Glovebox 4 Total	**1.75**	**43.75**	**212.5**	**5312**
	Sum of all Gloveboxes	9.42	234.75	479.606	11,989
Glovebox 3	**Secondary Background**	**8**	**200**	**8**	**200**
All Processes	**Total Secondary Background**	**75.5**[26,27]	**1888**	**75.5**	**1888**
	Sum with background	**93**	**2325**	**543**	**13,877**

[26] This is derived from 67.5 hours out of gloves (from the time motion study) for gloveboxes 1, 2 and 4 added to 8 hours out of gloves for glovebox 3.
[27] System Functions and Requirements Analysis for Am-241 Recovery Operations, Los Alamos National Laboratory (2010) LA-CP-10-1109

Compliance with design goal/administrative control limits

P121 Radiation Protection states that, "For a continuously occupied area, the design must maintain the average radiation exposure levels As Low As Reasonably Achievable (ALARA) and below an average of 0.5 mrem per hour. Continuous occupancy (2000 hr/yr) must be assumed unless the occupancy for the activity and area has been well established, documented, and committed to by line management and Project Design Team." This is tantamount to limiting the design to 1000 mrem per year for dose to the whole body. The total yearly dose presented in Table 10 (which assumes processing of 25 batches per year) shows that, alone, the background dose rates meet or exceed this. It is suggested that facility management investigate ways to reduce the ambient dose to the glovebox worker to below 1000 mrem per year. This report's conservative estimate of the yearly dose to the whole body from operations in all gloveboxes, taking secondary background radiation into account, to be around 3,325.5 mrem/year. The dose from the processes, ignoring secondary background, is collectively less than the limit, at around 979.5 mrem/year. When the actual number of people participating in a project is considered (3.75), the annual whole body dose remains below P121 limits.

Additional Workers

The estimated worker time required to process 25 batches per year (2045.5) exceeds the length of a working year (2,000 hours). Further, a study of workplace efficiency at TA-55 has demonstrated that employees are only engaged in productive work 42% of their time engaged in productive work[28]. This would suggest, that, based on a 2,000 hour work year for employees engaged solely with the CLEAR-line operations, only 840 hours would be spent on the clear line and thus:

$$\frac{2045.5\ \frac{hours}{year}}{840\ \frac{hours}{employee-year}} = 2.44\ employees$$

2.44 employees would be necessary to complete 25 batches. According to the Cost estimate justification for WBS 01.03.02.02.02 *$^{241}AmO_2$ Production Glovebox Work*, revision 1.1[29], a minimum of 2.75 employees are required to achieve a production level of 500 g per year (17 operations/year). The subject matter expert also extrapolated the work-force requirement to a minimum necessary 3.75 employees to support a production basis of 750 g/year (25 operations per year) [29]. Thus, for the calculation of person-mrem for this project, **3.75 employees are assumed to be exposed per year.**

If the sum of the each CLEAR-line glovebox's contributions to whole body dose for 25 operations per year is divided by the required number of employees, this results in a yearly

[28] Kornreich, D.E.; Burnside, M.R.; Burnside, R.J.; Demuth, N.S.; Gonzales-Lujan, J.M.; Hadden, B.M.; Lier, K.; Jackson, J.W.; Parker, R.Y.; Rising, T.L., "TA-55 Pit Manufacturing Responsive Infrastructure and Capapcity Study (U)" Los Alamos National Laboratory (2005), LA-CP-05-0256

[29] Schulte, Louis D. Personal Communication (3/6/2014)

whole body dose of 886.7 mrem/year. The same figure for **dose to the extremities is 3,701 mrem/year**.[30]

ALARA considerations

ALARA is based on three principals: keeping doses below regulatory limits, justifying dose-limitation practices by demonstrating a net benefit and optimizing the radiation protection schemes by adjusting the worker's exposure time, their physical distance from the source and the thickness and composition of the shielding.

10 CFR 835 specifies that ALARA is an approach to radiological control to manage and control exposures (individual and collective) to the work force and to the general public at levels that are as low as is reasonable, taking into account social, technical, economic, practical, and public policy considerations. ALARA is not a dose limit but a process which has the objective of attaining doses as far below the applicable limits as is reasonably achievable. ALARA requirements state that further reductions in dose are not indicated where the costs of reducing exposure become disproportionate to the benefit achieved.

Regulations and Laboratory policy require new design to be ALARA and further that optimization methods be used to assure that occupational exposure is maintained ALARA in developing and justifying facility design and physical controls. This project team has designed engineering controls consisting of shielding for the box and has spent additional time contemplating shielding for hand tools and shielded containers that will be used within the glovebox. The project team has spent effort to balance the controls with competing factors that influence the efficiency. Optimizing exposures includes taking relevant factors along with the estimated doses into account. The objective of attaining doses as far below the applicable limits as is reasonably achievable includes weighting all of these inputs along with social, technical, economic, practical, and public policy considerations. At this point LANL upper management is in discussions regarding the quantification of the non-health related detriment of exposure to ionizing radiation such as the social, technical, economic, practical, and public policy considerations. Therefore at this time quantifying an optimization is not possible; however, it is recommended that once decisions in management are made regarding values for non-health related detriments that a formal quantified ALARA optimization be documented.

Conclusions

Table 12 summarizes the results for whole-body dose due to clear line operations from this analysis. Results for dose to the extremities are shown in Table 13. The analysis in this

[30] Dividing the calculated doses for a system by the number of employees is not common as both P121 and 10 CFR Part 835.202 mandate a design objective of 0.5 mrem/hr for a 2,000 hour year. However, because the 25 operations per year will require more than 2,000 hours to complete, because the dose from whole body from the sum of doses over all the gloveboxes is less than 1 rem /year, and because the doses from the individual gloveboxes each maintained far below any design goal, the technique of dose-spreading was used in this analysis.

document serves to evaluate personnel dose for selected unit operations of highest concern associated with planned americium-241 recovery. Of particular concern is the fact that the secondary background dose in the room is greater than the design criterion (an average of 0.5 mrem/hr) for the development of new processes. Thus, it is impossible to design a process that satisfies the radiation protection design criterion in this room for a single individual (the primary dose to the whole body from CLEAR-line operations considering the current radiation protection design satisfies this design criterion). The current estimate of the minimum number of workers provided by the subject matter expert (3.75 workers at a production level of 750 g/year) satisfies the Dose Design Objectives.

Attention to operational details, shielding, and improving worker skill all can clearly contribute to reducing exposure in glovebox operations. As in the case of earlier work at LANL, operational experience and improvements are likely to provide better understanding of high dose operations and the methods to optimize/reduce personnel dose.

Table 12. Summary of whole-body doses from CLEAR-line gloveboxes

Operation	Time per batch (h)	Yearly time (h)	Dose per batch (mrem)	Yearly dose (mrem)
Glovebox 1 Total	27.15	678.75	17.632	440.8
Glovebox 2 Total	32.52	813	11.699	292.47
Glovebox 4 Total	22.15	553.75	9.847	246.184
Sum of all Gloveboxes	81.82	2045.5	38.73	979.5
Total Secondary Background	93.82	2346	93.82	2,346
Sum with background	**-**	**-**	**133.0**	**3,325.5**

Because some operations involved in these processes are still under development, this report represents a conservative view of the americium recovery process in its current state of development. The goal of this study was to determine the feasibility of the americium recovery project as it stands. Several conservative simplifying assumptions made in this assessment will be improved upon as the process develops. Methods for handling the Inconel crucible and the filter boat are still in development; as these actions become more specified better estimates of their associated doses can be calculated. The blending operation is also in the development though, due to the conservative assumptions made in this analysis, the actual operation will likely result in a lower dose.

Table 13. Summary of extremity doses from CLEAR-line gloveboxes

Operation	Time per batch (h)	Yearly time (h)	Dose per batch (mrem)	Yearly dose (mrem)
Glovebox 1 Total	4.15	103	101.5	2536
Glovebox 2 Total	3.52	88	165.606	4141
Glovebox 4 Total	1.75	43.75	212.5	5312
Sum of all Gloveboxes	9.42	234.75	479.606	11,989
Total Secondary Background	75.5	1888	75.5	1888
Sum with background	**93**	**2325**	**543**	**13,877**

The dose per worker for this project, assuming a minimum of 3.75 workers is 886.8 mrem/year whole body and 3700 mrem/year to the extremities.

Glovebox 3 is located in an area of locally higher secondary background due to a safe with radioactive material in close proximity. This report recommends that this safe be moved to a different location to reduce the secondary background to workers in this area. The relocation of this safe would also reduce the secondary background to workers at the other gloveboxes.

This analysis shows that, for the estimated minimum number of workers, the dose satisfies the design objectives set in P121. Specifically, radiation protection measures are in place such that doses to the worker's whole-body will not exceed 1,000 mrem in a year and that doses to the worker's extremities will not exceed 10 rem in a year.

This evaluation was conducted in accordance with RP-3-06-PR-05.2, "Radiological Design Review Procedure." This evaluation meets the requirements of P121, "Radiation Protection, Chapter 12 – Radiological Design" and ISD 341-2, "Engineering Standards Manual Chapter 11, Radiation Protection."