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Prepared for the U.S. Department of Energy
Assistant Secretary for Environmental Management

Project Hanford Management Contractor for the
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Status of Portable Non-Destructive Assay at the Plutonium Finishing Plant

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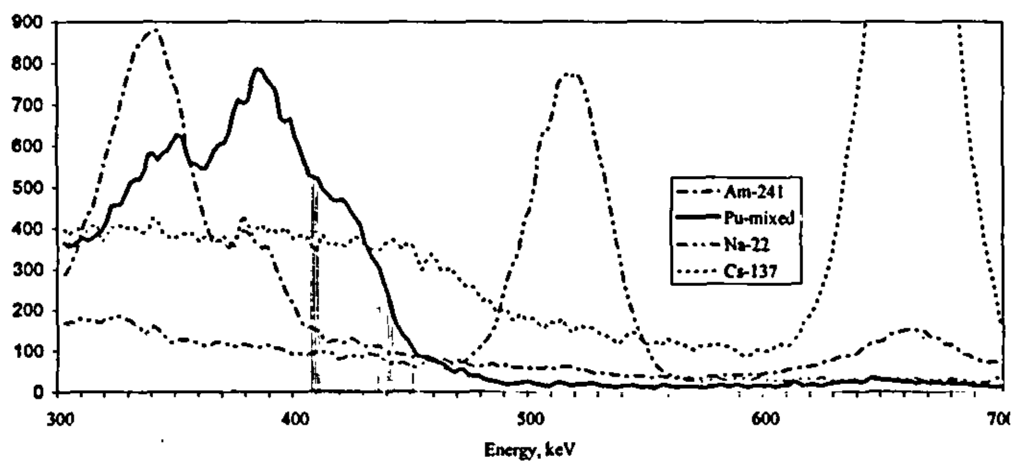
ABSTRACT

Collimated portable gamma-ray detectors are used to quantify the plutonium holdup in support of facility deactivation and decommissioning. The Generalized Geometry Holdup model[1,2,3] recently has been implemented for data reduction to support a new decontamination and decommissioning mission. An approach to assess the total measurement uncertainty (TMU) has been developed.[4] The TMU is added to the assay value for compliance with safety based limits. Details of the measurement techniques and comparisons to assays of materials removed are described.

DETECTION EQUIPMENT

The primary detection systems are 2 by 2-inch (5.1 by 5.1-cm) Sodium Iodide (NaI) detectors. The region-of-interest (ROI) is approximately 405-435 keV. Spectral background is subtracted using an ROI from approximately 440 to 450 keV. ROI limits are determined by counting sources as shown in Figure 1, ROI Limits Used in Analysis. The low-energy limit is set above the extent of the 376 keV ^{241}Am photopeak. The high energy limit is set to just lower than the Compton edge from ^{137}Cs and also below the low-energy extent of the 511 keV annihilation photopeak resulting from the decay of ^{22}Na. An interesting phenomena occurs because the ROI's occupy a portion of the spectrum with a fairly consistent slope, small gain shifts only have slight effects on the net count rate.

Figure 1. ROI Limits Used in Analysis.



§ Currently with Pacific Northwest National Laboratory.
** On contract to Fluor Hanford, Inc.

ASSAY CONVENTIONS

Most gloveboxes are assayed by placing the detector inside gloveports to assess each surface. The floor, sidewalls, and ceiling are assessed by tipping the detector. Measurement shots are selected with the goal to assay the complete surface with adjacent measurements overlapping at near the Effective Length[††] (Eff L). When practical, internal glovebox equipment is avoided during the assay of surfaces and is assessed separately. Otherwise, such equipment is inherently included in the assay of surfaces. Corrections for incident angles are not made, nor are self attenuation corrections made on area source geometry items.

A secondary means is to model the entire glovebox as a single plane of activity down the centerline of the glovebox. The detector is placed back from the front edge at a distance equal to one-half the glovebox depth or greater. The detector is intentionally aligned with the edge and corners of the glovebox to fill one-half and one-fourth of the detector field of view, respectively. Weighting factor corrections are applied for partial field of view measurements. To avoid glovebox frame/structure on the edges and corners of the glovebox, the detector generally is moved in from the edge by approximately 4 in. (10 cm) and aimed slightly outward pointing at the center of the floor, wall, or ceiling.

Linear systems such as vacuum piping, ventilation ducts and some conveyor gloveboxes are assayed from the far field and calculated as line sources. Special items such as valves and elbows are assessed separately as point sources. Spacing between adjacent shots is maintained at near the Eff L. Self attenuation and finite width corrections are made.

TOTAL MEASUREMENT UNCERTAINTY

The TMU includes all identified sources of uncertainty that affect the quality of a final measured value.

Systematic uncertainties are evaluated by segregating individual measurements into distinct populations with similar characteristics. Each distinct population group is assigned an uncertainty that represents each member in the group. Generally, the population group size is an entire surface, line length, or item.

The systematic uncertainty of the measurement is calculated as follows:

$$\sigma_{systematic} = \sqrt{\begin{aligned}&(m_{tot}\times\sigma_f)^2 + (m_{tot}\times\sigma_K)^2 + \sum_p (m_p\times\sigma_{CF(AT)_p})^2 + (m_{tot}\times\sigma_{CF(AT)_{general}})^2 + \\ &\sum_p (m_p\times\sigma_{ICF_p})^2 + \sum_p (m_p\times\sigma_{dist_{Line,p}})^2 + \sum_p (m_p\times\sigma_{glove_p})^2 + \sum_p (m_p\times\sigma_{equip_p})^2 \\ &+ \sum_p (m_p\times\sigma_{ledges_p})^2 + \sum_p (m_p\times\sigma_{mat\ distrib_p})^2 + \sum_p (m_p\times\sigma_{f\ bkg_p})^2 + \sum_p (m_p\times\sigma_{Sorenson_p})^2\end{aligned}}$$

[††] Eff L is a unitless geometrical constant of width/distance approximately equal to the full width at half maximum of the radial response curve.

where:

m_{tot} and m_p	is the calculated total and population mass, respectively.
p	represents the population group.
σ_{f_i}	is the uncertainty for the mass fraction of ^{239}Pu.
σ_K	is the calibration uncertainty.
$\sigma_{CF(AT)}$	is the attenuation uncertainty for each shield.
$\sigma_{CF(AT)general}$	is the general attenuation uncertainty that is judged to be 10 percent, due to additional sources of uncertainty including: measurements at angles, poorly known material densities, use of empirically determined coefficients, and a variable thickness of glove material.
σ_{ICF}	is the uncertainty associated with the item correction factor (ICF)[‡‡]. It is due to both the uncertainty of the position of the deposit within the line width or point source and due to the uncertainty in the width of a line or size of the point source.
σ_{dist}	is the distance uncertainty. It is a systematic uncertainty in relation to line sources, because the detector is held a consistent distance from the surface of an item for multiple measurements.
σ_{glove}	is the contaminated glove uncertainty. Assays made through glove ports, bagout ports and windows are assumed clean, but may be contaminated with plutonium.
σ_{equip}	is the intervening equipment uncertainty. Assays made through glove ports assume the activity is located on the opposite surface. Intervening process equipment may be included in assay measurements from both sides resulting in assaying the item twice.
σ_{ledges}	is the ledges uncertainty. When a glovebox is modeled as a center plane, the assay is through the glovebox wall. Additional steel framework between panels and around edges could represent significant shielding on the near surface, but not on the far surface.
$\sigma_{mat\ distrib}$	is the material distribution uncertainty. Measurements of area and line sources have sensitivity to non-uniform material distributions, there may be a diminished response at the edge of a surface or line, and it is not generally practical to space adjacent measurement shots uniformly at Eff L.
$\sigma_{f\,bkg}$	is the forward background uncertainty due to background interference from plutonium deposits forward of the detector and difficult to account for in a normal background measurement. The uncertainty is assigned based on technical judgment.
$\sigma_{Sorenson}$	is the Sorenson[§§] uncertainty. Measurement assays assume the detector is aiming directly at the object. However, this is not always the case and results in an underestimation of the activity.

The attenuation uncertainty and the ICF uncertainty are estimated from the range of plausible correction factors. The distance uncertainty, σ_{dist}, is estimated from the range of plausible distances. In each case, the range is assumed to represent four standard deviations of a normal distribution.

‡‡ The ICF is the historical Plutonium Finishing Plant name for the Finite Width Correction Factor.

§§ Donald L. Sorenson is a senior NDA technician at the Plutonium Finishing Plant.

The ledges and Sorenson uncertainty are assumed to represent an underestimation by as much as 50 % and 11 %, respectively. The intervening equipment and contaminated glove uncertainty are assumed to represent an overestimation by as much as 50 %. Each case is assumed to represent the boundary at three standard deviations of a one-sided probability distribution. Therefore, σ_{ledges}, $\sigma_{Sorenson}$, σ_{equip} are assigned uncertainties of 17 %, 4 %, and 17 %, respectively. The contaminated glove uncertainty, σ_{glove}, is also estimated to be 17 %, however, a default value of one-half the maximum value, or 9 %, is assigned.

The basis for the material distribution uncertainty is depicted in Figure 2, The Individual Detector and Overall System Response to a Single Point Source Located on the X-axis, showing the effects of un-even detector spacing in relation to counting a single point source located on the x-axis. The x-axis also represents measurement positions. The Y-axis represents the individual detector responses to a point source located accordingly on the X-axis. The thick black line represents the average response of the overall measurement system (measurements made at each detector location).

Figure 2. The Individual Detector and Overall System Response to a Single Point Source Located on the X-axis.

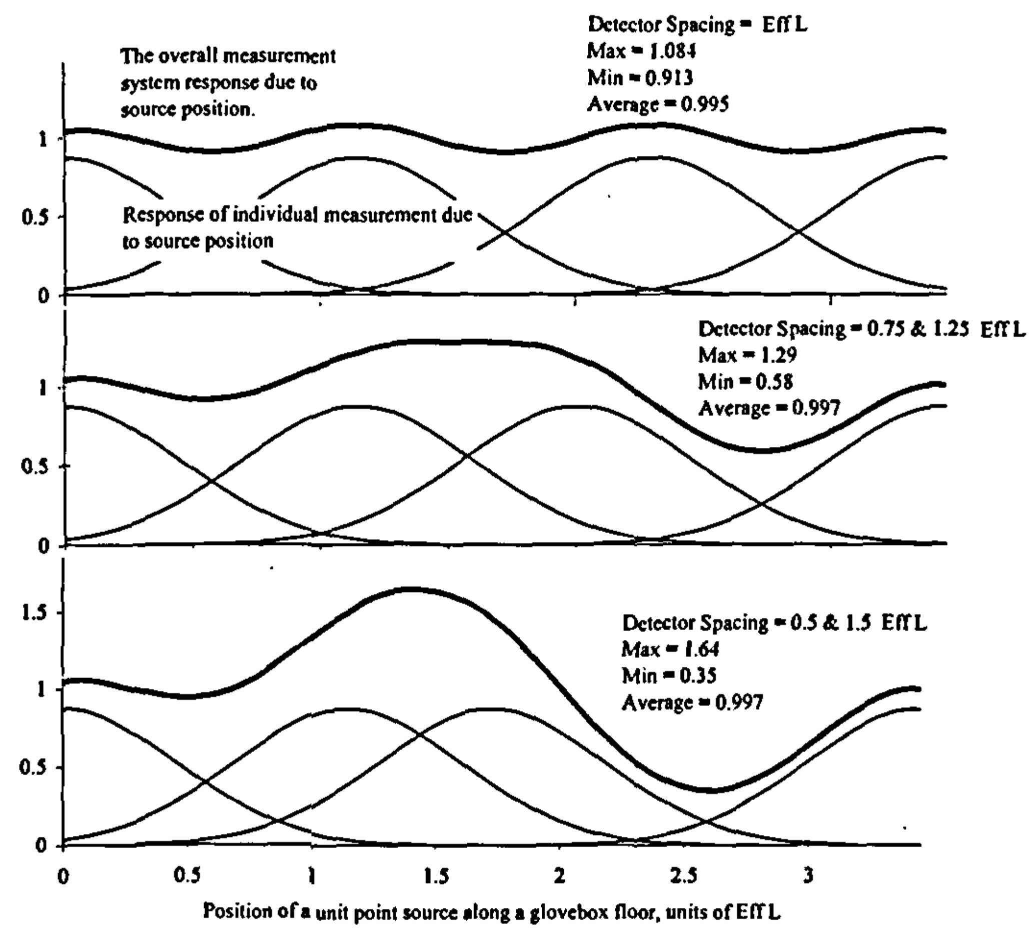


Figure 2 is taken to represent the extreme case of positioning effects due to assaying a localized deposit when the detector placement deviates from normal spacing. In reality, material distributions tend to be spread out. The minimum and maximum values of Figure 2 are taken to be the lower and upper ends of a normal probability distribution at the 99 % confidence level, or six sigma. These curves were used to support a default guidance listed in Table 1, Default Guidelines for $\sigma_{mat\ distrib}$. The two dimensional uncertainty is estimated by root sum square of each dimension.

Table 1. Default Guidelines for $\sigma_{mat\ distrib.}$

Spacing of measurements	One dimensional uncertainty	Two dimensional uncertainty
Near Eff L	3%	4%
Less than +/- 25% of Eff L	12%	16%
Greater than +/- 25% of Eff L	22%	31%
Angled measurements	N/A	31%

The random uncertainty contribution is estimated using the following reduced equation:

$$\sigma(random) = \sqrt{\sum_j \left[\left(\frac{\sqrt{\sum_i \sigma_{self}^2}}{N} \right)^2 + \sigma_{\bar{x}}^2 + \sigma_{area}^2 + \sigma_{length}^2 + 4\sigma_{dist_{point}}^2 \right]}$$

where:

σ_{self}	is the uncertainty due to counting statistics adjusted for self attenuation (reference 2)
$\sigma_{\bar{x}}$	is the standard deviation in the mean
σ_{area}	is the area uncertainty for area source items
σ_{length}	is the length uncertainty for line source items
σ_{dist}	is the distance uncertainty for point source items. It is considered random as the distance bias to the actual deposit is variable depending on the rotation of the object.
i	is each individual measurement
j	is each individual item, line segment or surface in the total system.

The overall TMU is the root sum square of the random and systematic components.

COMPARISONS TO FIXED INSTRUMENTS

At the time of this writing assays of material removed from 16 gloveboxes, 3 demisters and 4 vacuum lines have been compared with differences between initial and post remediation holdup measurements. Table 2, Glovebox, Demister, and Vacuum Line Comparison Data, shows the comparison data.

Residue material from gloveboxes is measured using calorimetry, a small table segmented gamma scanner, an add-a-source neutron counter, or a fixed NaI counter. Waste materials ultimately are assayed by either a large table segmented gamma scanner, an add-a-source neutron counter, or a fixed NaI counter. The Fixed NaI counter is used for small gram items. It uses a wider ROI and more simplistic attenuation and geometry correction than the portable instruments.

Comparison data is confounded by several factors, some of which are listed in the table notes. Several gloveboxes are interconnected with conveyors. Cross contamination can occur either through physically staging packages and equipment in gloveboxes or through a common ventilation system. Glovebox filters are generally external to the glovebox and are not part of this comparison. Lastly, this comparison does not account for uncertainties in the Fixed NDA systems.

It is also noted that some gloveboxes were omitted from this comparison. One omitted glovebox likely was cross contaminated with items not assessed by NDA, two gloveboxes were initially assayed early in the implementation of the Generalized Geometry Holdup method before the described assay conventions were standardized in practice, and several gloveboxes had negligible removals compared to the NDA values.

Table 2. Glovebox, Demister, and Vacuum Line Comparison Data. (2 sheets)

ID	Initial NDA	2 σ	Removals[a]	2nd NDA[a]	2 σ[a]	Removals + 2nd NDA	Within +/- 1 σ[b]	Within +/- 2 σ[b]
Box A	645	250	449	198	102	647	X	X
Box B	129	130	41	132	86	173	X	X
Box C	328	290	31	194	122	225	X	X
Box D	192	148	18	127	80	145	X	X
Box F	768	764	822	145	92	967	X	X
Box H[c]	609	1256	409	241	292	650	X	X
Box I[d]	157	132	99	67	100	166	X	X
Box K	738	592	194	603	438	797	X	X
Box L	1144	906	248	475	304	723	X	X
Box M	45	34	9	0	2	9	-	-
Box N[e]	1167	768	403	916	800	1319	X	X
Box O	1302	1098	734	280	266	1014	X	X
Box P	565	716	245	217	146	462	X	X
Box R[f]	2048	1126	1069	1522	982	2591	X	X
Box S	329	210	55	299	178	354	X	X
Box T	32	24	16	12	14	28	X	X
Sub Total	10198	-	4842	5428	-	10270	-	-

Table 2. Glovebox, Demister, and Vacuum Line Comparison Data. (2 sheets)

ID	Initial NDA	2 σ	Removals[a]	2^{nd} NDA[a]	2 σ[a]	Removals + 2^{nd} NDA	Within +/- 1 σ[b]	Within +/- 2 σ[b]
Demister A	354	220	298	N/A[g]	-	298	X	X
Demister B	357	302	337	N/A[g]	-	337	X	X
Demister C	391	380	331	N/A[g]	-	331	X	X
Sub Total	1102	-	966	N/A[g]	-	966	-	-
Vac A	1019	448	925	67	56	992	X	X
Vac B	507	378	489	45	106	534	X	X
Vac C[h]	696	354	819	183	192	1002		X
Vac D[i]	287	516	282	N/A	-	282	X	X
Sub Total	2509	-	2515	295	-	2810	-	-
Grand Total	13809	-	8323	5723	-	14046	-	-

Notes:

[a] Several gloveboxes had multiple assay and removal cycles. The second NDA represents last assay made at the time of this writing.

[b] The uncertainty for this purpose solely is taken to be the uncertainty reported for the initial measurement.

[c] The first assay of box H was particularly difficult, the measurement was through a one-half inch lead floor. The subsequent assay was through the gloveports.

[d] Box I contains a high background from adjacent gloveboxes.

[e] Box N contains glass tanks, which were calculated as drained in the first NDA, but unknowingly (to NDA) they were not empty. Tanks were drained for the subsequent NDA.

[f] Box R was found to contain materials in which self-attenuation was not accounted for in the first NDA. Material was removed for second NDA.

[g] The item was completely removed. There was no subsequent NDA.

[h] Vacuum line C was calculated as empty, but found to be plugged with a solid material. The additional attenuation was not accounted for in the initial assay.

[i] Vacuum line D was interior to a glovebox and subject to a large uncertainty in differentiating glovebox background from contained plutonium.

DISCUSSION

At the time of this writing, assays of the plutonium removed compares favorably with the difference between initial and subsequent portable measurements using the generalized geometry holdup method. The TMU calculation for gloveboxes appears to be somewhat overstated, except for Box M. There is no attributable reason for the discrepancy in Box M. In general, it is thought that the material distribution uncertainty may be somewhat overstated. It may also be overstating the TMU by including separate terms for the counting statistics adjusted for self attenuation, the standard deviation of the mean, and the material distribution uncertainty. Given the consequences of understating NDA uncertainties, there are no plans to revise uncertainty estimates. More data is needed for comparison purposes. It is planned to report additional comparative data in the future.

A nationally recognized means for determining the TMU, for holdup measurements, is needed. It is hoped that this field work can be a step toward that end.

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