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A NONDESTRUCTIVE ASSAY SYSTEM FOR USE IN DECOMMISSIONING A PLUTONIUM-HANDLING FACILITY

by

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Nondestructive Assay Section
Special Materials Division

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ABSTRACT

Argonne National Laboratory is decommissioning a facility used to fabricate reactor fuel elements. The equipment is contaminated with alpha emitters at levels up to 10^{12} dpm/100 cm^2. The objective of decontamination is to reduce the TRU concentrations below 10 nCi/g of waste. A portable NDA procedure using NaI(Tl) gamma-spectrometric techniques was selected to measure the residual Pu and ^{241}Am in the glove boxes. Assays were performed at different stages in the decontamination process to estimate the detection system sensitivity and the effectiveness of the cleaning efforts.

I. BACKGROUND

The Reclamation Service of Argonne National Laboratory is in the process of decommissioning a facility which was used to develop manufacturing procedures for plutonium and uranium reactor fuels.[1] This facility, #350, was completed in 1959 and was operated for approximately 15 years, producing fuels for various loadings of the EBR-1, EBR-II, ZPPR, and ZPR critical assemblies. The nuclear materials processed included Pu-metal alloys, mixed-oxide (MOX) powders, and highly enriched uranium. During the operating life of the installation, hundreds of kilograms of plutonium were processed.[2] The fabrication equipment, including items such as lathes, extrusion presses, and furnaces, is located in large glove boxes which are connected by enclosed conveyor lines. The glove boxes are constructed from .93-cm-thick aluminum plate with .93-cm-thick CR-39 (allyl polycarbonate) windows. They are assembled in subsections which are 91 cm long x 122 cm deep

x 91-244 cm high (Fig. 1). The total contaminated surface area is of the order of 2.3×10^3 m^2 (Fig. 2). Preliminary reclamation efforts have removed the major concentrations of SNM; however, the area is still highly contaminated.[3] Typical contamination levels are in the mCi/cm^2 range. These levels may vary significantly, depending upon the type of work conducted within the individual glove box. The objective of the reclamation effort is to reduce the glove-box surface contamination below the 10 nCi TRU/g of waste mandated by DOE as the limit for irretrievable disposal.[4,5] Transuranic wastes (TRU) include materials containing any alpha-emitting isotope with an atomic number greater than 92, and ^{233}U. The nuclides of interest in this project are $^{238-242}Pu$ and ^{241}Am. In the course of the decommissioning, accurate estimates of the levels of plutonium and americium contamination must be obtained. This information is important to ensure the safety of the personnel involved, to measure the effectiveness of successive cleaning efforts, and to decide on final disposition of the material.[6,7,8]

Shortly after facility #350 ceased operation, experiments were performed on a sample glove box to determine the relative effectiveness of various cleaning procedures and solvents.[3] The results of this study were used to formulate a decontamination plan which called for a limited number of surface washings with a caustic solution. However, a measurement procedure which can be used while large-scale decontamination efforts are progressing is necessary. This would permit an assessment of the effectiveness of the cleaning efforts at specific locations and determine the practicality of attempting to reach the TRU limit.

DOE waste-handling procedures require that contaminated material be segregated by the kind and amount of contamination present. Packaging and transport requirements are much more stringent for wastes which exceed the TRU limits. Since the ultimate cost of disposal will be dependent on the level of plutonium contamination, it would be beneficial to be able to identify material which has been decontaminated below the 10 nCi TRU/g limit.

II. GLOVE-BOX ASSAY TECHNIQUE

In order for a measurement system to be useful during the various phases

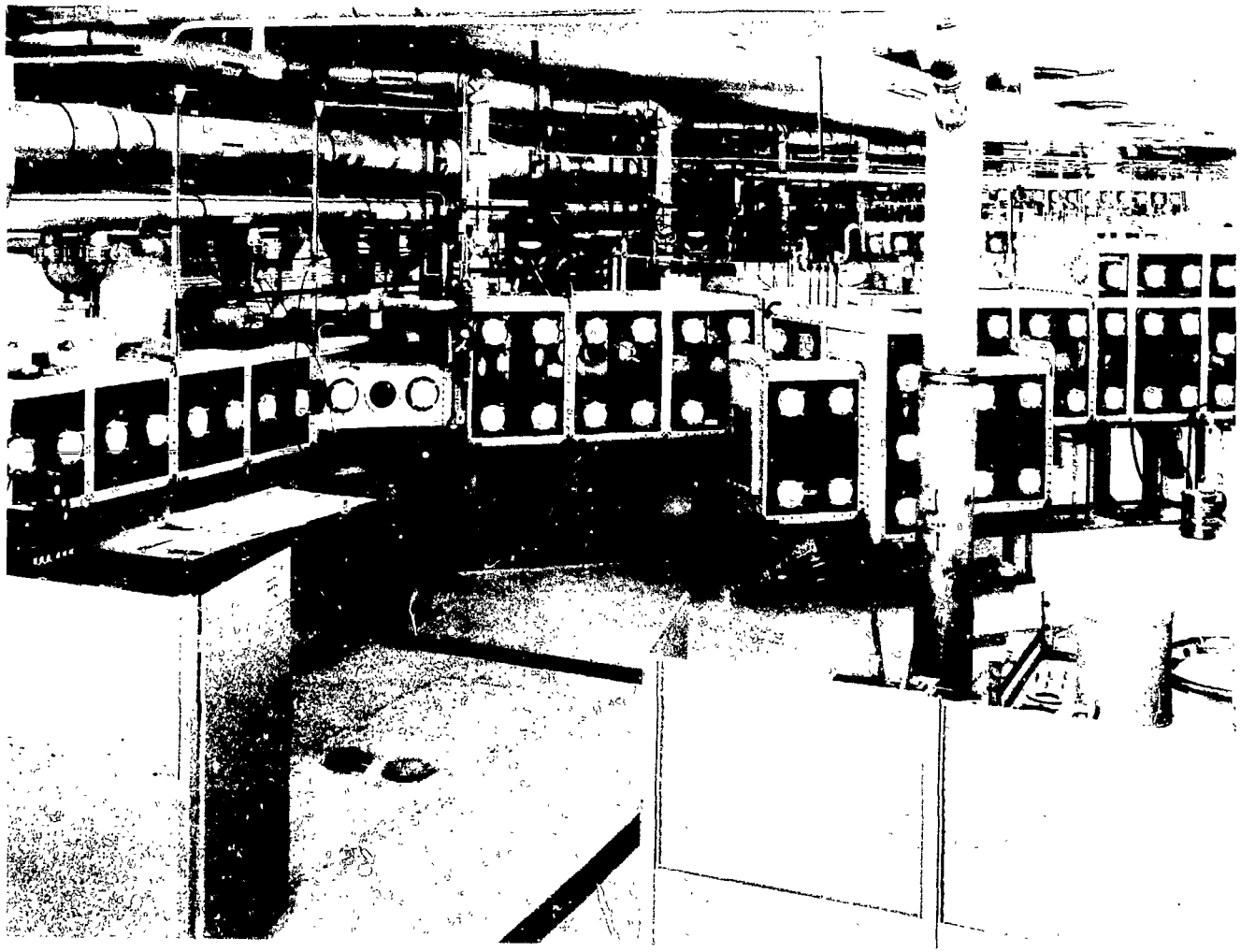

Fig. 1. Facility #350 glove boxes. Glove box PF-5, the plutonium machine shop, is in the foreground. ANL Neg. No. 206-78-8.

FUEL FABRICATION FACILITY

BUILDING 350

First Floor Plan

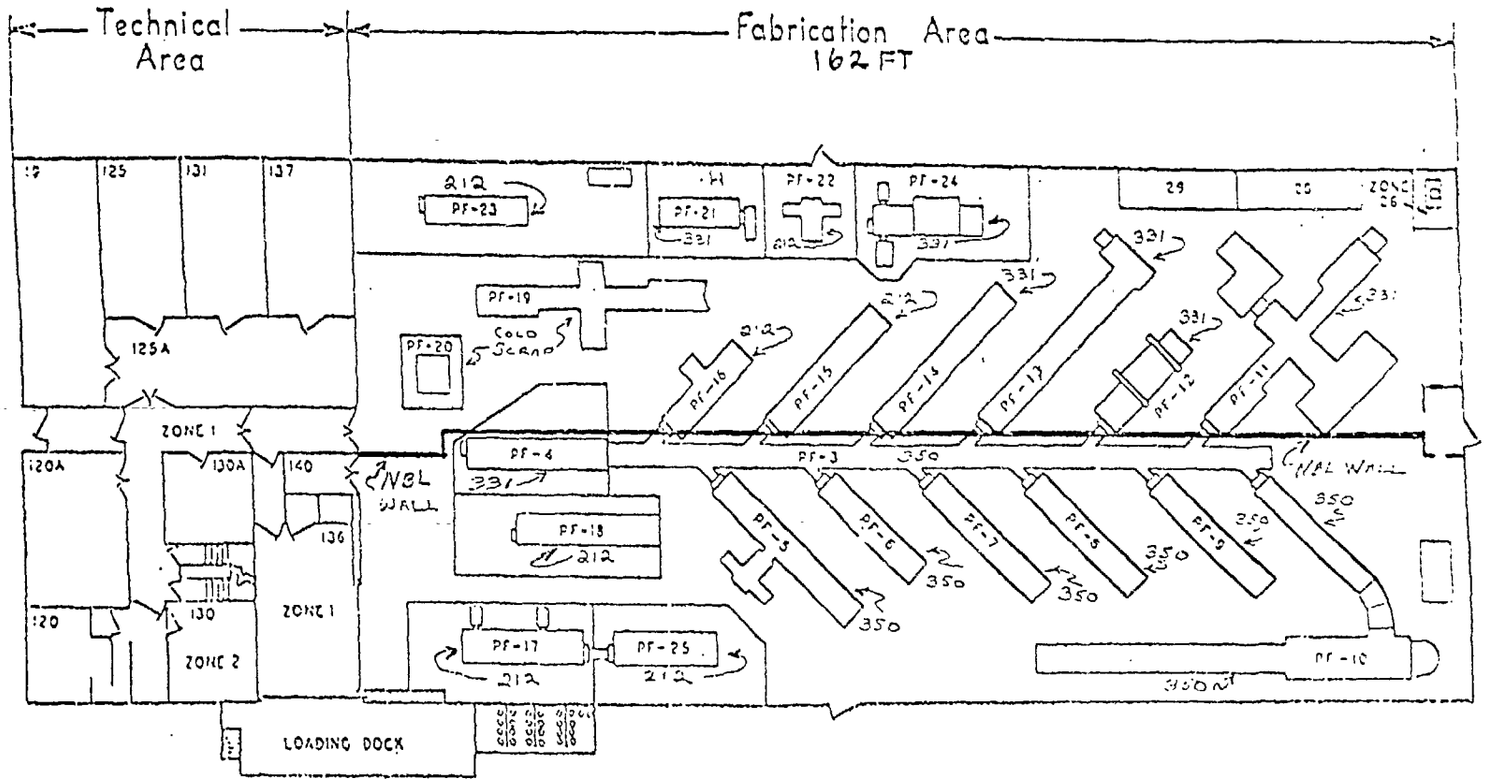

Fig. 2. Floor plan for Facility #350.

of the decontamination process, it must be applicable over a wide range of Pu concentrations. The level of contamination in the #350 glove boxes prior to clean-up has been estimated at 10^9-10^{12} dpm/100 cm^2.[3] The final goal of decontamination is approximately 2 x 10^6 dpm/100 cm^2. A gamma-ray survey procedure was selected to measure Pu and ^{241}Am contamination on the interior surfaces of the boxes. The principal difficulties encountered in these assays result from the nonuniformity and nonstandard geometries of the "samples." In general the contents of the boxes will be removed prior to decontamination efforts. This equipment may then be assayed separately. Except in cases where a specific use exists for an item, the equipment will be classified as TRU waste and packaged accordingly. The primary focus of the assay technique will be the glove boxes which will undergo decontamination treatment. The conditions under which these assays will be performed require that the equipment be portable, operationally simple, and geometrically versatile. A set of NaI(Tl) scintillation detectors, selected to cover the expected count-rate range and coupled with a simple, signal-processing package, best fit these stipulations.

The regions of the gamma-ray spectrum used in this analysis are the 60-keV transition for ^{241}Am and either the 375-450 keV window or the 13-22 keV L X-ray window for plutonium (Fig. 3). Specialized detector holders and collimators are used to assure reproducible positioning of the detector relative to the contaminated surface and to shield against external background radiation. The counting electronics consists of a single, lightweight package containing the detector high-voltage supply, the stabilized amplifier circuitry, and two single-channel analyzers with a scaler. The unit was modified to allow processing of the amplifier signals by a multichannel analyzer (Fig. 4).

High-Level Contamination

During the early stages of decontamination, the objective of the assay procedure is to provide estimates of the $^{241}Am/^{239}Pu$ ratio and the total ^{239}Pu content without entering the glove box. This necessitates the choice of gamma-lines with sufficient energy and intensity to have a reasonable count rate after passing through an 0.93-cm Al plate (Table 1). The 60-keV transi-

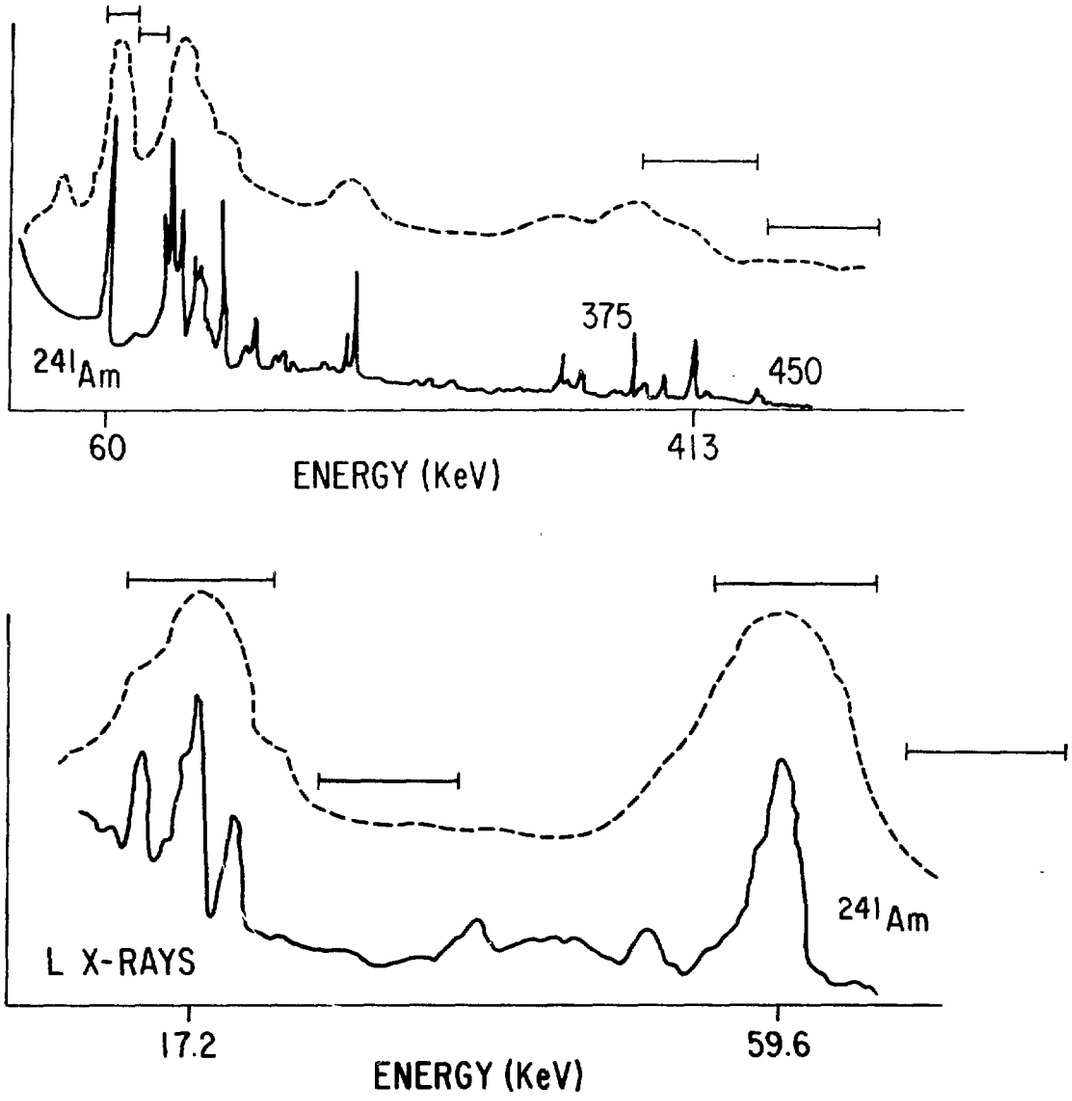

Fig. 3. Plutonium, Americium photon spectrum.

Upper Spectrum: High-energy gamma-ray spectrum (20-550 keV) observed during an external assay of a glove box. The dashed and solid lines show the resolution of a NaI(Tl) detector vs a Ge(Li) spectrometer. The brackets delineate the regions of interest in the assay (60 keV for ^{241}Am, 375-450 keV for ^{239}Pu).

Lower Spectrum: Low-energy gamma-ray and X-ray spectrum (10-70 keV) observed during an internal assay of a glove box. The regions of interest are 12-22 keV for L X-rays and 60 keV for ^{241}Am.

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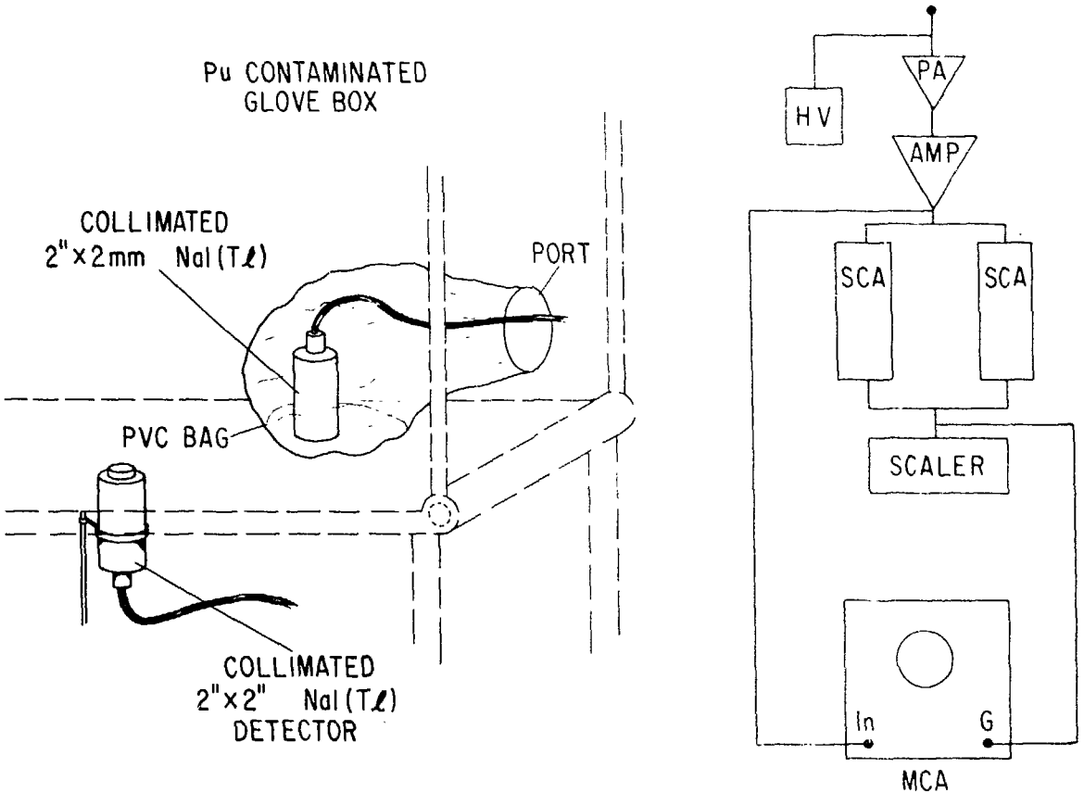

Fig. 4. Schematic representation of assay procedure and hardware.

The right-hand portion of the figure is a block diagram of the signal-processing electronics. The equipment includes a HIGH VOLTAGE SUPPLY (HV), a PREAMPLIFIER (PA), a GAIN-STABILIZED AMPLIFIER (AMP), two SINGLE CHANNEL ANALYZERS (SCA), a SCALER, and a MULTICHANNEL ANALYZER (MCA). All the equipment except the MCA is enclosed in a single package (Eberline SAM-II).

The left portion of the figure shows the configuration of the detectors for the internal and external assay.

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TABLE 1. Intensities of photon transitions of interest in glove-box assay.

PHOTON INTENSITIES[a]

I. Gamma-Ray Intensities

Nuclide	Gamma-ray Energy (keV)	dpm/ng	I (gamma/decay)
^{239}Pu	375.0	1.4×10^{2}	1.5×10^{-3}
	413.7		1.5×10^{-3}
	451.5		1.8×10^{-4}
	sum (375 to 451)		4.5×10^{-3}
^{241}Am	60	7.2×10^{3}	3.5×10^{-1}

II. L X-Ray Intensities[b]

Nuclide	dpm/ng	I (gamma/decay)	(X/min)/ng	X-ray %[c]
^{238}Pu	3.7×10^{4}	0.142	5.3×10^{3}	9
^{239}Pu	1.4×10^{2}	0.047	6.4	20
^{240}Pu	5.0×10^{2}	0.118	5.9×10^{1}	23
^{242}Pu	8.7	0.120	1.1	--
^{241}Am	7.2×10^{3}	0.382	2.8×10^{3}	48

(a) See references 12, 13.
(b) Principal photon energies: Pu (13.6, 17.2, 20.2), Am (13.9, 12.5, 21.3) keV
(c) Typical fuel composition:
^{238}Pu - 0.05%, ^{239}Pu - 87%, ^{240}Pu - 11.5%, ^{241}Am - 0.5%

tion is used to assay ^{241}Am. Even after a 50% reduction in intensity due to the Al floor, the high emission probability (0.35/decay) is sufficient to ensure a usable count rate at low levels. The limited energy resolution of the NaI(Tl) crystals restricts the ^{239}Pu assay to the 375-450 keV region. These are low probability transitions and will limit the sensitivity of plutonium detection. Since it would be prohibitively time-consuming to assay the entire inner surface of the structure with sufficient detail to provide the desired statistical precision, a two-step sampling procedure is used. The objective of this method is to first locate large deposits of Pu which were not removed in the clean-out following shutdown, and then to obtain a relative mapping of the contamination levels. Based on this information, precise assays of strategically selected points can be conducted and the results extrapolated over larger areas.

The preliminary survey is performed with a hand-held 5.1-cm diameter x 1.3-cm thick NaI(Tl) probe. Short counts (0.1 min) were taken of the area within a grid system marked on the external surface of the box. A 7.6-cm x 7.6-cm grid was used in these experiments. A 122-cm x 183-cm section of the box floor can be covered with a 35% spatial efficiency in approximately 1/2 hour. A contamination contour map is generated on which the "hot spots" are easily located and from which average contamination levels of various areas can be determined. The NaI(Tl) detector was contained in a 1-cm-thick Pb shield with a 20-cm^2 aperture. The electronics were adjusted so that the SCA window would encompass the 60-keV ^{241}Am line. Comparison of the ^{241}Am and ^{239}Pu concentrations derived from more precise measurements at various locations on the box floor have shown that the $^{241}Am/^{239}Pu$ ratio remains constant $\pm$ 7%. Thus an americium survey also provides an estimate of plutonium spatial distribution.

Detector energy calibrations were performed with a set of standards constructed from PuO_2 having similar isotopic composition to the material processed in the #350 glove boxes (^{238}Pu - 0.02%; ^{239}Pu - 91%; ^{240}Pu - 8.3%; ^{241}Pu - 0.5%; ^{242}Pu - 0.05%; ^{241}Am - 0.4%). The source material was deposited on high-purity aluminum disks in spots with a nominal area of 5 cm^2. The sources ranged in Pu content from 11 mg to 870 ng. The sources were alpha-counted to determine the total alpha count rate, and the Pu isotopic ratios

were determined by gamma-ray spectrometry. A 20-mil polyvinyl chloride (PVC) covering was used on each source to protect against surface abrasion. The relative uncertainties in ^{239}Pu and ^{241}Am content were less than 1% for all sources.

The detector linearity of the 5.1-cm x 1.3-cm NaI(Tl) probe was verified in the range 10^5 dpm (^{241}Am) to 3×10^8 dpm (^{241}Am). The americium detection limit was determined in the absence of external background by assaying the standards in a realistic configuration with typical gamma-absorption conditions (0.93-cm Al plate). Levels of 7×10^5 dpm [$\sim$ 5 ng (^{241}Am)/cm^2] could be reproducibly determined at 2σ above room background.

The second stage of the analysis involves an assay of geometrically well-defined areas to determine the ^{239}Pu and ^{241}Am content. The areas are chosen to be representative of the range of count rates observed in the glove box during the preliminary survey. The assay includes a determination of the gamma activity from the 60-keV transition of ^{241}Am and the 413-keV peak complex of ^{239}Pu (Fig. 3). The net count rate of each region is obtained by subtracting a compton scattered background contribution from the gross count rate. The compton background is estimated from the activity in a higher energy window adjacent to the photo peak. Background contributions due to adjacent contamination deposits must also be removed. The Pu and Am concentrations are then determined by comparative assay of a set of standards in a physical arrangement similar to the unknown assay. Corrections are made for absorption conditions or geometric arrangements in which the glove-box assay deviates from the calibration configuration. The key parameters are the source geometry, the source-to-detector distance, and the photon-absorber composition and thickness.[9,10] The count rate is inversely proportional to the square of the source-to-detector distance. This is critical at small distances. In the majority of cases, we are considering flat surfaces (such as glove-box floors). Corrections for curved surfaces (such as air ducts) may be performed by applying a calculated correction factor[11] or by constructing a mock-up using a similarly shaped pipe.

Used in these assays was a 5.1-cm x 5.1-cm NaI(Tl) detector. It was contained in a 1.25-cm-thick Pb shield and was collimated to a 1.9-cm diameter

using a double-slit system. The detector was recessed 4.5 cm from the external face of the collimating aperture. This arrangement was mounted on a mobile support designed to reproducibly position the crystal relative to an area of interest. The linearity of the detector was verified in the range 1.3 x 10^3 dpm to 7 x 10^7 dpm for ^{239}Pu and 3 x 10^8 dpm to 10^5 dpm for ^{241}Am. The detector limits were determined by assay of known sources through 0.9-cm Al in the absence of external background. Levels of 4 x 10^7 dpm (40 μg/cm^2 for ^{239}Pu and 3 x 10^4 dpm (4 ng/cm^2) for ^{241}Am could be reliably detected. (The TRU limits for ^{241}Am and ^{239}Pu are 8.3 ng/cm^2 and 441 ng/cm^2, respectively.)

Low-Level Contamination

The assay technique for discriminating between TRU and non-TRU samples is analogous to that previously discussed for high-level contamination. The principal differences between the two methods result from focusing attention on more intense, lower energy radiation emitted by the Pu isotopes. Among the difficulties encountered in measuring low-energy photons is the low penetrability of matter by L X-rays. This increases the uncertainty in assays of materials with nonuniform absorption properties. In addition, detectors used in assaying material at these contamination levels are highly sensitive to external background sources.

As decontamination efforts approach the TRU limit and large Pu deposits are eliminated, the remaining contamination will be reasonably uniformly distributed and will be beneath the sensitivity limits for detection of the 375-450 keV gamma-rays through the Al floor. At these disintegration rates (22200 dpm/g of waste), high-probability transitions, such as L X-rays and the 60-keV ^{241}Am decay, are monitored (Fig. 3). The L X-rays, which have energies between 13-22 keV, are emitted following the alpha decay of the isotopes of Pu and ^{241}Am. They result from the internal conversion of the gamma ray from the first 2^+ state (∿ 50 keV). The intensities of the L X-rays for the various isotopes of Pu and ^{241}Am differ in the number of X-rays/alpha-decay (Table 1). Consequently, in order to unambiguously relate the measured count rates to dpm, we must know the sample isotopic composition. The isotopic ratios can be obtained from an analysis of samples from the box floor with the use of a high-resolution gamma spectrometer. However, this procedure

would greatly increase the experimental complexity, and the low Pu concentrations would limit the accuracy of the measurement. Simplifying approximations can be made which will result in upper limits useful for these assays.

The principal constitutent of reactor grade fuels, ^{239}Pu, emits the fewest L X-rays/alpha-decay. Consequently, if we assume that all L X-rays are emitted by ^{239}Pu and proceed to reduce the contamination accordingly, we will be safely below the TRU limit. If the contamination has the composition ^{238}Pu = 0.05%, ^{239}Pu = 87%, ^{240}Pu = 11.5%, ^{241}Am = 0.5%, the L X-ray intensity would be 29 X-rays per minute per nanogram [(X/min)/ng], as compared to 6.4 (X/min)/ng for pure ^{239}Pu. The TRU limit would be satisfied by a factor of x4.5. A more exacting determination of the count rate-dpm relation may be obtained by subtracting the ^{241}Am contribution to the L X-rays. Since the 60-keV line is unambiguously related to americium, we can calculate the CPM [(L X-rays from Am)/CPM (60 keV)] by assaying a sample of known ^{241}Am content. This assay should be performed in the identical arrangements as the unknown assays in order to account for photon absorption, geometry, and detector efficiency. Eliminating ^{241}Am from the above sample results in a Pu L X-ray intensity of 15 (X/min)/ng.

In order to assay these low-energy photon transitions with maximum efficiency, thin crystal NaI(Tl) detectors are used in an assay configuration approaching 2π geometry. Either a 12.7-cm x 1-mm or a 5.1-cm x 2-mm detector is used, depending upon contamination level and box geometric restrictions. The crystal thickness is chosen to optimize the detection efficiency in the L X-ray region and to minimize the background from higher energy photons. Because the transmission of the L X-rays is less than 1% through 0.9-cm-thick Al plate, it is necessary to have the detector in intimate contact with the contaminated surface. This is done by inserting the detector into the contaminated glove box inside a PVC sleeve. The transmission of 20-keV photons through 0.5-mm PVC is 70%. The detector is housed in a shield constructed from layers of cadmium, copper, and aluminum. The multi-element structure enables the detector to be used in high 60-keV background areas and also avoids low energy background due to X-ray fluorescence of the shield. The linearity of the detectors was tested with reactor-grade Pu sources having total dpm levels in the range 1.7×10^5 to 10^8. The detection limit of the

12.7-cm x 1-mm detector was determined to be below 25 pg (^{241}Am)/cm^2 in the absence of room background. The detection limit of the 5.1-cm x 1-mm detector was approximately 0.5 ng (^{241}Am)/cm^2 in backgrounds typical of those encountered inside a #350 glove box.

III. OPERATIONAL EXPERIENCE

An evaluation of the gamma-assay procedure was conducted during a typical glove-box decontamination sequence. The objectives of this experiment were to determine the assay-system response to realistic radiation environments and to observe the relative effectiveness of successive decontamination steps. The parameters of interest in the evaluation were the Pu and Am detection limits of the various counters, the measurement sensitivity to differing levels of contamination, and the factors which contributed to the measurement uncertainty. The experiment was conducted by performing a series of measurements preceeding the various steps in the decontamination procedure. This technique permitted modification of the cleaning routine or the assay plan based on the results from a preceeding cleaning effort.

Glove-box PF-5, the machine shop, was selected as the site for the assay evaluation (Fig. 1). This box contained a hydraulic press, a metal lathe, a drill press, a milling machine, and a mechanical shear, as well as numerous hand tools. Preliminary clean-out reports indicated that large Pu deposits had been removed. No attempt had been made to remove any of the equipment. It was decided to concentrate measurement effort on a limited area of the glove box. In this way a maximum number of measurements could be performed on an area exclusively devoted to one type of manufacturing process. The section containing the mechanical shear was chosen as it contained the maximum accessible open area and the minimum vertical structure (Fig. 5). The shear was oriented in the north-south direction, slightly to the west of center, in a 7 x 10^3 cm^2 section of floor. A brief survey showed contamination levels in the mg/cm^2 range.

A detailed set of procedures for the decontamination and volume reduction of each glove box in #350 was written prior to the start of the decommissioning project.[2] In general, the procedures may be separated into a series of well-

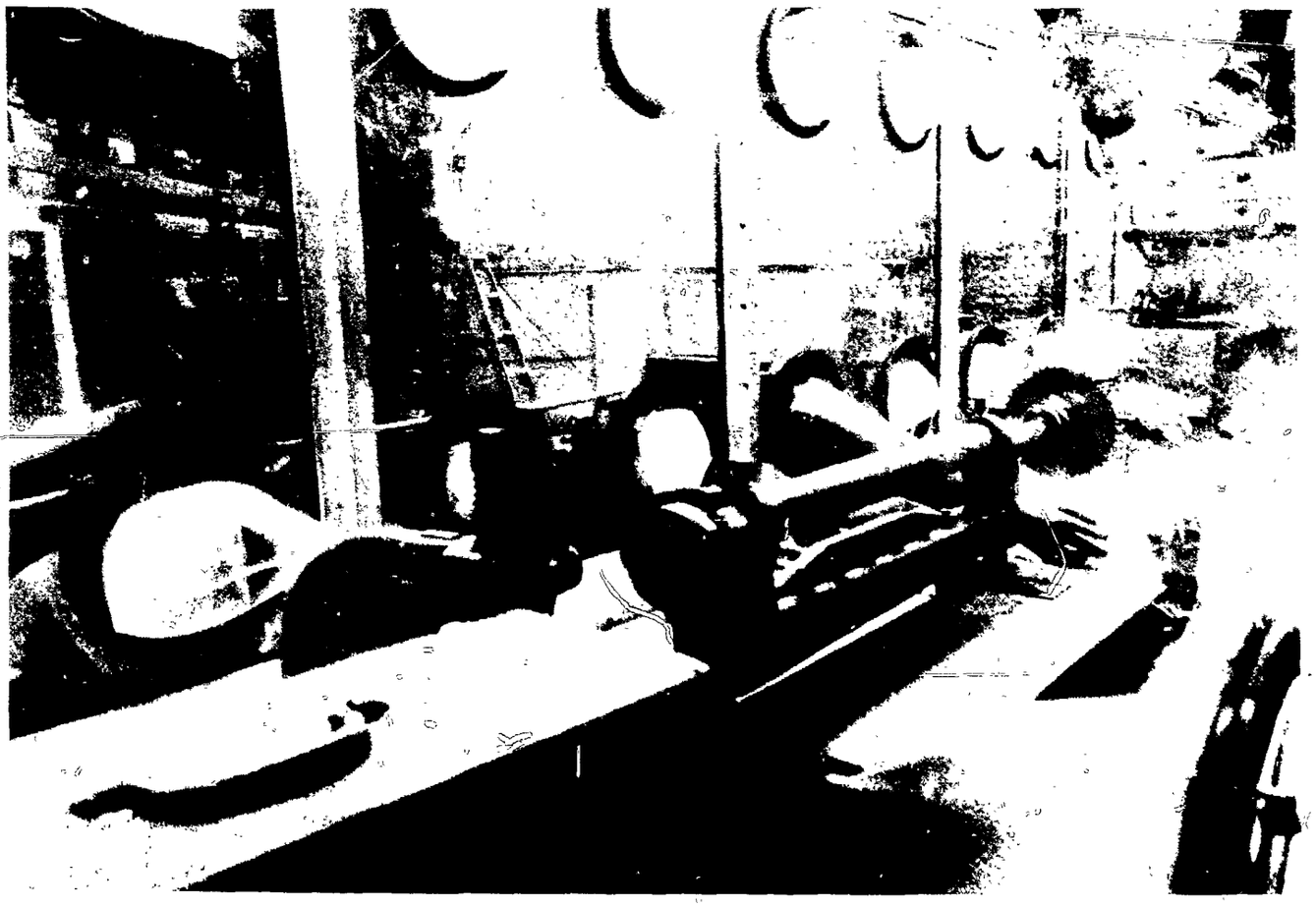

Fig. 5. Internal arrangement of PF-5 in the vicinity of the mechanical shear prior to clean-out. The figure shows the area to be assayed viewed from the northwest corner of the box. ANL Neg. No. 150-79-27 #12.

defined steps. Prior to any cleaning, the hand tools and small scrap would be removed and the major equipment disassembled. A dry cleaning (sweeping and vacuuming) is then performed, and as much of the major equipment as possible is removed. This is followed by a series of washes with a general-purpose cleaner containing detergents, buffering salts, and water softeners (pH - 11.2). The final step prior to volume reduction is to paint the surface to fix any remaining contamination. This process provides a number of convenient break points for performing measurements (Fig. 6). A series of high-level contamination assays (using both the 5.1-cm x 1.3-cm and the 5.1-cm x 5.1-cm detector systems) were performed prior to clean-out, after removal of the major equipment and following each wash. A low-level contamination assay was performed prior to the painting of the internal surface.

The high-level contamination assay consisted of an external scan of the glove-box floor for 60-keV radiation, followed by more precise measurements of ^{239}Pu (375-450 keV) and ^{241}Am (60 keV) concentrations. In this experiment, six sets of external measurements were performed sequentially during the decontamination process. Assays A and B were performed prior to and after the equipment was removed. Assays C and D were conducted after washes with a caustic solution, while E and F followed washes with an HF solution (pH < 1). The scan technique generated a contamination map which was used to determine the average ^{241}Am concentrations on various segments of the box floor. The assay of geometrically well-defined areas provided the $^{241}Am/^{239}Pu$ ratio and related the scan values to the ^{241}Am contamination levels. The resulting contamination values at each step are shown in Table 2. The subdivision of the box floor into four regions was an attempt to relate contamination to physical conditions within the box. Section 1 (394 cm^2) refers to the area beneath the shear. Section 2 (394 cm^2) consists of the portion of the floor immediately adjacent to the shear on either side. Sections 3 (535 cm^2) and 4 (858 cm^2) refer to the segments of the box closest to the western and eastern glove ports (Fig. 7).

The uncertainties in the ^{241}Am levels in the various sections include the variance of the scan data within each section, and the statistical and calibration errors of the americium assay of the geometrically well-defined area. The $^{241}Am/^{239}Pu$ ratio was determined in steps A, B, and C. The average

GLOVE BOX

DECONTAMINATION AND ASSAY

FLOWCHART

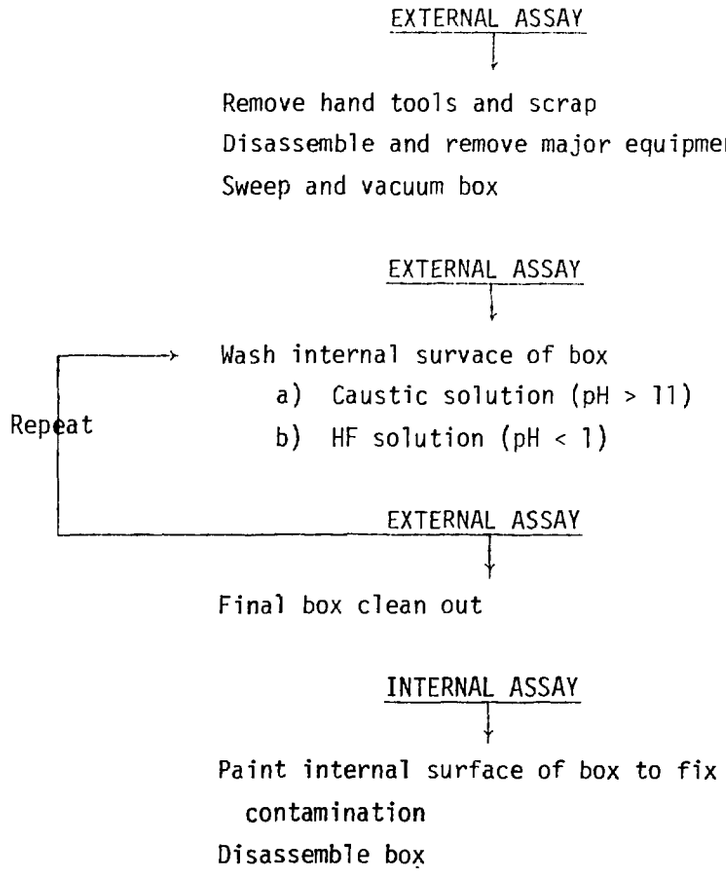

Fig. 6. Glove-box decontamination and assay procedure flowchart.

TABLE 2. SNM contamination in glove-box PF-5.

Section	^{241}Am(a) 1	2	3	4	TOTAL		P1	P2
Decon	(394 cm^2)	(394 cm^2)	(535 cm^2)	(858 cm^2)	^{241}Am	^{239}Pu(b)	20 cm^2	20 cm^2
A	1.3 ± 0.6(c)	4.2 ± 0.4	2.2 ± 0.4	1.6 ± 0.1	2.2 ± 0.3	337 ± 73	4.3	1.6
B	0.24 ± 0.05	1.8 ± 0.4	2.0 ± 0.2	1.1 ± 0.1	1.3 ± 0.2	217 ± 62	2.1	1.1
C	0.15 ± 0.05	1.1 ± 0.4	0.7 ± 0.08	0.9 ± 0.1	0.75 ± 0.15	125 ± 35	0.4	0.9
D	0.1 ± 0.05	0.5 ± 0.2	0.5 ± 0.05	0.8 ± 0.1	0.55 ± 0.12	92 ± 25	0.12	0.7
E	0.08 ± 0.05	0.1 ± 0.05	0.17 ± 0.05	0.3 ± 0.05	0.19 ± 0.05	32 ± 10	0.08	0.3
F(d)	0.08 ± 0.05	0.1 ± 0.05	0.1 ± 0.02	0.2 ± 0.05	0.14 ± 0.04	23 ± 8	0.08	0.2

(a) ^{241}Am contamination in $\mu g/cm^2$.

(b) ^{239}Pu determined from $(^{241}Am/^{239}Pu) = 0.006 \pm 0.001$.

(c) Uncertainties include variance of scan values and uncertainty in ^{241}Am assay.

(d) TRU limits: ^{241}Am - 0.008 $\mu g/cm^2$ ^{239}Pu - 0.441 $\mu g/cm^2$.

Fig. 7. Internal arrangement of PF-5 in the vicinity of the mechanical shear prior to clean-out. The figure shows the work area in front of the shear (Section 4). ANL Neg. No. 150-79-27 #7.

of these values, 0.006 ± 0.001 was assumed to apply throughout the experiment. The ^{239}Pu ($\mu g/cm^2$) values in Table 2 were derived from the corresponding ^{241}Am data and used the average ($^{241}Am/^{239}Pu$). The ^{239}Pu count rates were reduced to within 2σ of the background levels by the second caustic wash. This corresponds to a detection limit of approximately 125 $\mu g/cm^2$ for ^{239}Pu. The ^{241}Am detection limit was set at approximately 80 ng/cm^2 as a consequence of the uncertainties in the 60-keV scan at this level. Both of these detection limits are above the values necessary to determine when the TRU criteria have been met. (The TRU limit for ^{241}Am is 8.3 ng/cm^2; for ^{239}Pu it is 441 ng/cm^2.)

The effectiveness of successive decontamination steps in various areas of the box floor is shown in Fig. 8. The major contaminant removal occurred in the early washes. This was most noticeable in Sections 1 and 2, where the contamination would be loose due to shielding from the equipment. P1 is a small area (20 cm^2) in Section 2 which was readily accessible to cleaning effort. The americium level was reduced 90% by the first wash and was brought down to the detection limit by the second. The surface of Section 4 (which included P2) was noticeably darker than the rest of the floor due to material imbedded in the aluminum. This section did not respond well to initial washes with the caustic solution; however, an HF solution, which etched the aluminum, had better results.

Estimates of the contamination levels present at the end of the decontamination process were performed with a 5.1-cm x 2-mm NaI(Tl) detector in a clean 0.5-mm-thick PVC sleeve. Prior to this assay, the floor was etched with an HF solution, and the entire box was again washed with a caustic solution. The 60-keV transition from ^{241}Am and the L X-rays following Pu and Am decay were measured. The L X-rays were analyzed under the assumption that the $^{241}Am/^{239}Pu$ ratio was 0.006 ± 0.001 and that ^{241}Am accounted for 50% of the L X-rays, while ^{239}Pu accounted for 20%. (See above discussion.) The data showed ^{239}Pu ranging from 22.5 $\mu g/cm^2$ to 0.67 $\mu g/cm^2$ and ^{241}Am ranging from 90 ng/cm^2 to 3 ng/cm^2. Estimated uncertainties in these assays are of the order of ± 40% based on the error in the $^{241}Am/^{239}Pu$ and Pu isotopic ratios, on the variances in repetitive assays, and on errors in calibration and geometry. Although no attempt was made to relate the data to the average

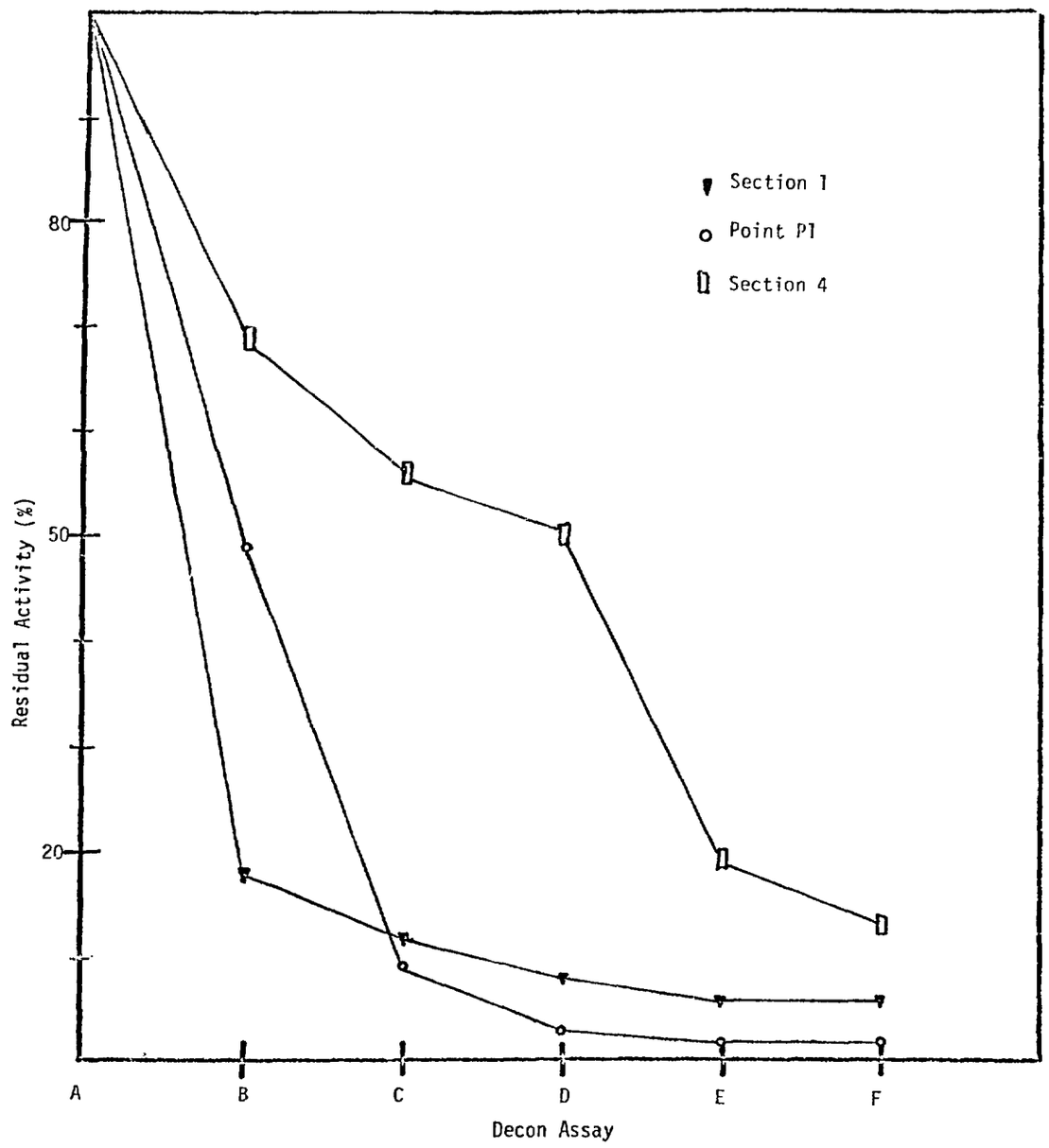

Fig. 8. PF-5 external ^{241}Am assay.
ANL Neg. No. 150-79-35.

contamination levels for the entire box, assays of typical areas gave ^{239}Pu concentrations of 1.4 $\mu g/cm^2$ and ^{241}Am concentrations of 6 ng/cm^2.

IV. CONCLUSIONS

In summary the data suggest that a gamma-spectrometric technique using an NaI(Tl)-based detection system should be adequate to determine the ^{241}Am and Pu contamination during the various stages of decontaminating the #350 glove boxes. External assays are useful during the early stages of the process, but must be supplemented by internal low-energy photon analysis as the 10 nCi/g limit is approached. In addition, while the TRU limit may be reached on surfaces accessible to cleaning, contamination hidden in shielded, nonaccessible regions (such as bolt holes, crevices, and air ducts) must be considered in the final disposal decision.

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