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MEASUREMENT OF TRANSURANIC SOLID WASTES AT THE 10-nCi/g ACTIVITY LEVEL

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ABSTRACT

The measurement of transuranic contaminated solid wastes at 10-nCi/g activity levels is presented, including a variety of different approaches to the problem. The development of two nondestructive assay systems, the MEGAS and the Pancake Counter, is described in detail. These are NaI-based detection systems to be used specifically for low-density solid transuranic wastes. They are of value in effective implementation of new federal regulations pertaining to transuranic waste management in that they supplement the usual administrative decision making in waste handling.

I. INTRODUCTION

Solid wastes contaminated with transuranic elements are to be handled differently according to their activity levels, as prescribed in AECM Chapter 0511.¹ The criterion has been set at 10 nCi of transuranics per gram of waste, representing the maximum concentration that alpha wastes may contain and still be placed into nonretrievable storage (i.e., burial). These alpha wastes are defined as, "...those contaminated with certain alpha-emitting radionuclides of long half-life and high specific radiotoxicity to greater than 10 nCi/g...."¹ The radionuclides included are plutonium and transplutonium nuclides (except ²³⁸Pu and ²⁴¹Pu) and ²³³U and its daughter products. Table I lists the isotopes of major interest in this regulation and their specific activities.

Single, pure isotopes of plutonium are never found in solid wastes. Isotopic mixtures of fairly constant composition result

from the various waste sources. For example, weapons-grade plutonium typically contains the isotopes ²³⁹Pu, ²⁴⁰Pu, and ²⁴¹Pu. The ²⁴¹Pu is typically 1% or less by weight. This small amount of ²⁴¹Pu does not of itself produce any appreciable alpha activity, but does produce an alpha-emitting daughter--²⁴¹Am.

Before the 10-nCi/g criterion could be effectively implemented, an assay scheme was needed that would allow measurements at and below this activity level with some acceptable degree of confidence. Owing to very low natural radiation emissions at the 10-nCi level, it was necessary to take an approach somewhat different from the more conventional assay systems. Historically, the two accepted techniques for the quantitative analysis of plutonium-bearing solid wastes have been either spontaneous fission neutron counting or high-energy gamma-ray detection. For example, if the plutonium

TABLE I

SPECIFIC ACTIVITIES OF RADIOISOTOPES FOUND IN ALPHA WASTES

Isotope	Alpha Half-Life (yr)	Alpha Specific Activity ($\alpha/s-g$)	Spontaneous Fission Activity (f/s-g)	Major γ -Ray Activities		L X-ray Activity (x/s-g)
				γ Energy (keV)	Activity ($\gamma/s-g$)	
^{233}U	1.6×10^5	3.5×10^8	-	42	6.7×10^5	1.4×10^7
				55	1.4×10^5	
				97	2.1×10^4	
				146	6.4×10^4	
^{238}Pu	8.6×10^1	6.4×10^{11}	1.1×10^3	44	2.5×10^8	7.4×10^{10}
				100	4.7×10^7	
				153	6.5×10^6	
^{239}Pu	2.4×10^4	2.3×10^9	1.0×10^{-2}	52	3.2×10^5	9.2×10^7
				129	1.3×10^5	
				375	3.5×10^4	
				414	3.5×10^4	
^{240}Pu	6.6×10^3	8.4×10^9	4.1×10^2	45	3.8×10^6	8.8×10^8
				104	8.4×10^5	
				160	8.7×10^4	
^{242}Pu	3.8×10^5	1.4×10^8	7.7×10^2	-	-	1.4×10^7
^{241}Am	4.6×10^2	1.2×10^{11}	-	60	4.2×10^{10}	4.6×10^{10}
				99	2.6×10^7	
				103	2.3×10^7	

content of 4 kg of solid wastes is to be measured, the analysis system must be capable of measuring just 560 μg of plutonium (with a 6% ^{240}Pu content) because that amount of plutonium is equivalent to 10 nCi/g when averaged over the entire waste package. Such an amount of plutonium will yield only approximately 0.014 spontaneous fissions/s and 18 high-energy (414-keV) gamma rays/s.

II. ANALYSIS OF POTENTIAL DETECTION SCHEMES

Neither fission neutron counting nor high-energy gamma-ray measurement is capable of detectabilities of 10 nCi/g for large packages of wastes ranging in volume to 0.06 m^3 (2 ft^3) -- a reasonable volume for 4 kg of low-density (paper, rags, etc.) waste. The systems described below are designed specifically for the analysis of low-density transuranic wastes well below 10-nCi/g concentrations.

Table I lists the alpha emitters of major interest and shows what radiations are available for the passive assay of the sample. Although the specific alpha activities are high, the extremely short range of plutonium alpha particles in matter, (e.g., less than 40 μm in water) precludes alpha detection for anything but surface contaminations.

Table I, column 4, lists the spontaneous fission activities which, although low, have been used quite successfully^{2,3} in the nondestructive analysis (NDA) of plutonium fuel, scrap, and wastes. Since the spontaneous fission activities result primarily from the 'even' isotopes of plutonium (^{238}Pu , ^{240}Pu , ^{242}Pu) which are generally present in minor quantities relative to the ^{239}Pu content, an accurate isotopic analysis of the plutonium is necessary before a reliable total plutonium assay is

possible. However, neutron counting techniques have the highly desirable characteristic of being quite insensitive to waste matrix and self-absorption (e.g., plutonium 'lumps') effects, especially when compared to gamma-ray methods. Because the chemical form of the plutonium (Pu, PuO₂, PuF₄, etc.) is not usually well characterized in wastes, hence giving rise to unpredictable and large 'singles' neutron backgrounds from (α ,n) reactions in the sample, coincidence neutron counting is generally the method of choice for plutonium wastes. For such wastes contained in 115-l drums, this counting technique^{2,3} allows assays with errors less than $\pm 30\%$ and detectability limits (3σ [standard deviations] above background at sea level, 1000-s count time) of ~ 60 mg Pu ($6\% \text{ }^{240}\text{Pu}$). Singles counting with the same instrument will allow an improvement in the detectability limit by $\sim 4\times$, but a knowledge of the chemical form of the plutonium is necessary.³ In a typical waste package containing 4 kg of waste in a volume of 0.06 m³, the detection limit of 60 mg would be equivalent to 1070 nCi/g. In addition, ²⁴¹Am content would go unnoticed since this method is insensitive to gamma rays. Obviously, neutron counting would not be capable of assaying such plutonium wastes at the 10 nCi/g level so another technique was required.

Columns 5 and 6 of Table I list the major gamma-ray activities that could be used for the analysis of transuranic wastes.⁴ Such gamma rays provide good specific yields coupled with fair-to-good penetrabilities depending on the sample matrix and material distribution (e.g., plutonium 'lumps'). High-energy gamma-ray detection ($E_\gamma \geq 100$ keV) has, like the coincidence neutron counting mentioned above, already become a proven analytical tool for plutonium-bearing wastes in bulk form.⁵⁻⁸ The technique is reasonably accurate ($\sim \pm 20\%$) if matrix attenuation and self absorption^{6,9}

effects are properly accounted for. Detectability limits (3σ , 1000-s counting time) can be as low (good) as ~ 10 nCi/g, depending on the sample geometry (e.g., 1-l samples of ash). However, for the 0.06-m³ waste package previously mentioned, a detectability limit of ~ 3 mg plutonium,³ or ~ 54 nCi/g for a 4 kg waste sample, is all that is attainable using a good quality Ge(Li) detector. Although the monitoring of high-energy gamma rays provides detectability limits that are generally not adequate for 10 nCi/g analysis, it is a promising approach to the problem.

Column 7 of Table I lists the L x-ray yields¹⁰ of these radioisotopes. The L x rays ($E_x \sim 17$ keV, average) are emitted as a result of internal conversion in the daughter nucleus following the initial alpha decay by the parent. The x rays are thus characteristic of the daughter element and not the parent (e.g., uranium x rays result from plutonium alpha decay). The L x-ray yields are impressive, being $10^2 - 10^3$ greater than higher-energy gamma-ray yields (²⁴¹Am excepted). Owing to their low energy, however, the L x rays have only poor-to-fair penetrability in material (e.g., only one L x ray in five will successfully traverse 10 mm of water), but their penetrability is adequate for assaying low-density waste packages of limited size.

III. SELECTION OF ASSAY PRINCIPLE MEGAS

The assay principle that has been adopted for measuring bulk packages of low-density wastes is actually a hybrid of a gamma-ray detection scheme coupled to one detecting x rays. This Multienergy Gamma Assay System (MEGAS) permits monitoring of the x-ray region for the sub-10-nCi/g samples, subsequently crossing over to progressively higher-energy gamma rays as the activity level increases. The detector is

a 127-mm-diam by 50-mm-thick NaI(Tl) crystal with a 0.25-mm beryllium window. To improve backgrounds and energy resolution, the NaI(Tl) crystal is coupled to the phototube through a 50-mm-thick pure NaI light pipe. The beryllium window is thin enough to allow x-ray transmission into the crystal, whereas the detector itself is thick enough to absorb high-energy gamma rays (e.g., the 400-keV complex from plutonium).

Figure 1 shows what a typical photon spectrum from unshielded plutonium (plus ^{241}Am) would look like, collected with both a NaI(Tl) detector and a Ge(Li) detector.¹¹ The five prominent photon energy regions that can be used for analysis purposes are listed in the caption. For routine application to plutonium wastes we have chosen to use just three regions: the 17-keV x-ray region (Pu + ^{241}Am), 60-keV region (^{241}Am), and 400-keV 'complex' (^{239}Pu).

Such a hybrid system combines the high sensitivity (and relatively poor accuracy) of x-ray detection at the 10-nCi/g-or-lower level with the lower sensitivity, but improved accuracy, of gamma-ray detection at higher activity levels. The errors referred to here are due to absorption by the matrix material and self-shielding by the transuranics themselves. The self-shielding effects are severe for all photon energies considered here, particularly for the low-energy L x rays.⁹ Part of the self-shielding problem can be alleviated by using

proper standards (e.g., dried liquid plutonium standards for simulating process line-generated wastes from ion-exchange columns). Also, for typical room-generated wastes which are reasonably free of contamination, no self-absorption effects exist.

Pancake Counter

This small-package counter uses only the two lowest-energy, highest-intensity photon regions shown in Fig. 1. With low-density wastes, a 2-l package will contain less than ~400 g of material. This corresponds to ~56 μg plutonium (6% ^{240}Pu) at the 10-nCi/g level. Such an amount of plutonium, in 2l of waste matrix, is difficult (if not totally impossible) to measure using the low-intensity high-energy gamma rays. Therefore, for small packages of waste the previous MEGAS analysis scheme defaults to the high-intensity L x-ray region (17 keV) and the 60-keV region due to ^{241}Am alone. The detector is a 127-mm-diam by 1.6-mm-thick NaI(Tl) crystal with a 0.25-mm beryllium window. The detector, of the FIDLER¹² (Field Instrument for the Detection of Low-Energy Radiation) design, provides 100% intrinsic detection efficiency up to ~100 keV with lower backgrounds than those afforded with thicker crystals. A 25-mm-thick quartz light pipe provides improved energy resolution and lower backgrounds.

IV. SYSTEM DESIGN AND PHYSICAL PARAMETERS Box Counter (0.06 m³)--MEGAS

The large-package counter shown in Fig. 2 is designed to assay waste containers ranging in size up to the ~0.06 m³ (2 ft³) standard cardboard carton used at the Los Alamos Scientific Laboratory (LASL) for transuranic wastes. Such containers generally have a mass of 4 to 8 kg when loaded with low-density wastes. The MEGAS detector discussed above views the rotating and translating package through a 25-mm-wide by 100-mm-high iron collimator 25-mm thick. The detector is housed in an iron cylindrical shield 0.3-m long and 76-mm thick. A

Fig. 1. Plutonium spectra - Ge(Li) vs NaI detectors. The five main assay regions using the NaI detector are at 17 keV, 60 keV, 100 keV, 200 keV, and 400 keV.

Fig. 2. Box Counter (MEGAS).

lead plug shields the detector assembly from the rear. The face of the NaI detector is 0.3-m from the turntable centerline. The signals from the detector are processed and stored by a mini computer (with ADC interface) or a multichannel analyzer (shown). A complete pulse-height spectrum is collected for each sample and room background subtracted. The photon yields in each region of interest (17 keV, 60 keV, 400 keV, etc.) are then totaled and compared to a calibration curve yielding the net activity of plutonium and americium in the sample. A strain-gage-based load cell is located in the turntable with a visual and electronic digital readout (the latter going to the computer). Hence, a final activity/gram value is attached to the sample allowing an easy "go/no go" decision to be made on the disposition of the waste package.

Package Counter (2-2)--Pancake Counter

The small package counter is shown in Fig. 3. The uncollimated detector, FIDLER, views the flat sample from above and is located in a 13-mm thick lead cylindrical shield. The detector views the sample through a 0.12-mm plastic film stretched over a 0.3-m-diam hole cut into the lid of the stainless steel counting chamber connecting the two sorting dryboxes (see Fig. 3). Such a scheme provides for easy decontamination of the counting chamber as well as free access to the detector assembly outside the drybox. The distance from the detector face to the bottom of the sample is 0.3 m. Sample sizes can range up to 0.46 m in diam and up to 0.2 m high, but typical sample dimensions are much smaller than these extremes--hence improving considerably the assay accuracy. The signals from the detector are processed by an electronic package located next to the primary sorting box (see Fig. 4). This electronic package consists of an amplifier, two single-channel analyzers (17 keV and 60 keV), two scalers, and a timer. The Reset and Start controls for the scalers are located inside the sorting drybox so that the operator never needs to remove his hands from

Fig. 3. Small Package (Pancake) Counter - Detector (shielded), and secondary sorting box.

Fig. 4. Small Package (Pancake) Counter - Primary sorting box and electronics package.

the drybox once sorting has begun. The operator first places a preweighed sample beneath the detector, then presses the Reset and Start buttons on the inside wall of the sorting box. After the counting is completed (normally 10 s), the operator mentally compares the results to a previous "go/no go" calibration and quickly decides the disposition of the package--retrievable or nonretrievable storage.

V. CALIBRATION, DETECTABILITY, AND ACCURACY

A. Calibration

Calibration of the MEGAS and Pancake Systems is accomplished using clean representative waste samples spiked with thin (unshielded) plutonium and americium sources of known isotopic content that reasonably match the isotopic content of the plutonium wastes. Note that generally a sizeable change in the isotopic content of the plutonium in the wastes has a much

smaller absolute effect on the specific x-ray and gamma-ray yields of the plutonium. For example, the total L x-ray yield from plutonium of 94% ^{239}Pu and 6% ^{240}Pu is only 16% less than the L x-ray yield from plutonium of 90% ^{239}Pu and 10% ^{240}Pu . In this case the ^{240}Pu content has increased 67% in absolute content. Correspondingly, the yield of the high-energy 400-keV complex from ^{239}Pu decreases only 4% with a ^{240}Pu isotopic increase from 6% to 10%.

In the calibration procedure, the spiked waste standards are assayed with the Pu-Am sources randomly located in the sample. An average of the maximum and minimum responses (background subtracted) yields not only a mean calibration for the various photon energy regions, but also provides a measure of the response deviation due to different spatial locations in the sample. Using such a technique to calibrate the Box Counter for 0.06-m^3 boxes of low-density wastes (total mass ~ 6 kg), a variation of $\pm 50\%$ of the mean response was noted for the 17 keV photon region. Using the 400-keV complex, a variation of $\pm 30\%$ was observed. Again, this variation was due only to different responses from different spatial locations in the 0.06-m^3 vol. Similarly, for the Package Counter a $\pm 50\%$ and $\pm 35\%$ variation in response was noted for the respective 17-keV and 60-keV regions.

The photon spectrum measured from typical plutonium wastes generated at LASL (typical isotopic composition is $\sim 94\%$ ^{239}Pu and $\sim 6\%$ ^{240}Pu , with some ^{241}Am ingrown from ^{241}Pu) is shown in Fig. 1. While the 60-keV and 400-keV regions are essentially due to ^{241}Am and ^{239}Pu , respectively, the 17-keV region is a composite of L x rays from both ^{241}Am and Pu decay. In order to provide a plutonium analysis at 10-nCi/g levels, hence necessitating the use of the 17-keV region, the ^{241}Am contribution to this region must first be removed. This is accomplished by measuring the photon spectrum from clean wastes spiked only with

²⁴¹Am. Such a measurement provides a strip-out factor or ratio of the ²⁴¹Am intensity in the 17-keV region to that found in the 60-keV region. This factor was found to vary between ~0.6 and ~0.9, depending on the waste matrix (rubber, paper, etc.) and sample volume (e.g., 0.06 m³, 2%, etc.). The strip-out factor is then applied to the measured intensity (background removed) of the 60-keV region giving the appropriate ²⁴¹Am contribution to be removed from the 17-keV region. The remaining intensity in the 17-keV region is then considered due only to plutonium (of assumed known isotopic composition). The absolute plutonium activity is now calculated by comparison with a calibration curve that has the matrix attenuation correction included. (The new computer-based system, now being completed, will automatically correct for average matrix attenuation by including an $e^{\mu x}$ term in the calibration, where μ is the mass attenuation coefficient and x is a function of the measured mass, hence the density of the sample. A similar correction term will be included in the ²⁴¹Am strip-out calculation performed on the 17-keV yields.) The ²⁴¹Am content is determined by comparison of the 60-keV intensity to a calibration curve constructed with ²⁴¹Am sources. Similarly, the 400-keV intensity, when compared to the appropriate calibration curve, yields the ²³⁹Pu content in the waste sample. Note, however, that due to low yields (giving rise to large statistical errors), the 400-keV region cannot generally be used for 10-nCi/g samples.

B. Detectability

Detectability is herein defined as that minimum quantity of material, or minimum activity specific to a given radioisotope, that can confidently be measured at some stated level above background. Detectability must also always be associated with a given count time. As is generally accepted in trace element analysis, we chose to de-

fine our detectability as that amount of material that gives a measured response above background of three standard deviations (3σ) of that background. Such a criterion allows, statistically speaking, approximately one chance in one hundred of a false reading above that detectability limit. In other words, if strictly uncontaminated (clean) room trash is being measured, better than 99% confidence that all of the measurements would fall below the detectability limit could be expected.

For both the MEGAS and Pancake Counter, the detectability limit will depend on the isotope of interest, counting time, sample matrix, and total sample mass. However, to give an idea of the capabilities of these detection systems, consider Tables II and III for the MEGAS and the Pancake Counter, respectively. The tables show that both systems are capable of sub-10-nCi/g detectability limits in very short count times (e.g., with the Pancake Counter, 10 s is the count time routinely used for waste sorting and characterization studies). For 0.06-m³ waste volumes (using MEGAS), crossover to the 400-keV region for plutonium analysis would occur above ~36 nCi/g as shown in Table II.

Obvious ways to improve detectability are to increase sensitivity by increasing counting efficiency and improving energy resolution, which effectively reduce background contributions. Since we are working at the 3σ level above background, counting longer (say, 10 times longer) will improve (lower) the detectability by a factor of $\sqrt{10} \approx 3.2$. A reduction in background by 10 times will also improve the detectability by ~3.2.

C. Accuracy

Accuracy statements for the MEGAS and the Pancake Counter inherently contain precision statements, and while an instrument's precision may be excellent, its total accuracy may be very poor. In the case of the MEGAS and the Pancake Counter,

TABLE II

PLUTONIUM AND ^{241}Am DETECTABILITY LIMITS FOR MEGAS

Photon Energy Region (keV)	Isotope	DETECTABILITY LIMIT ^a			
		100-s Count		1000-s Count	
		nCi/g	μg	nCi/g	μg
17 (L x rays) ^b	Pu ^c	0.13	7.0	0.04	2.2
60	^{241}Am	8.5×10^{-3}	9.8×10^{-3}	2.7×10^{-3}	3.3×10^{-3}
490	Pu ^c	1.1×10^2	6.3×10^3	36.0	2.0×10^3

^aDetectability limits at the 3σ level for 4000 g of low-density wastes in a 0.06-m^3 corrugated paper box.

^b ^{241}Am contribution removed before analysis.

^cPlutonium with 94% ^{239}Pu and 6% ^{240}Pu .

the largest possible absolute error is due to self-absorption of the low-energy L x rays in the transuranic material itself (i.e., lumps). Matrix absorption is not a severe problem for low-density waste and can be canceled out to a large degree by using proper matrix standards or techniques such as transmission scans.⁸

The problem of lumps is a difficult one to correct and is generally ignored by many

experimenters. However, as previously pointed out,^{6,9} such lumps can cause errors of 50%, or even factors of two and higher, depending on the photon energy measured and the particulate size. The magnitude of this effect decreases with increasing photon energy; however, even for the "penetrating" 414-keV gamma ray, only 50% of these photons escape if 1 g of plutonium metal is contained in a single sphere. As mentioned earlier, much of this self-absorption problem can be solved by the use of proper standards that correctly simulate the particulate size (as well as matrix) of transuranium material in the unknown (e.g., simulating process line-generated wastes from ion-exchange columns with standards made from dried plutonium solutions). Additionally, in assaying typical LASL room-generated wastes which have been found¹³ essentially free of contamination, no self-shielding problem exists. However, in using the lowest-energy photon regions, particularly the 17-keV region, attempts at quantitative measurements can contain large errors that force the analysis to yield low activity values for the sample. Note that

TABLE III

PLUTONIUM AND ^{241}Am DETECTABILITY LIMITS FOR THE PANCAKE COUNTER

Photon Energy Region (keV)	Isotope	Detectability Limit ^a	
		10-s Count nCi/g	μg
17 (L x rays) ^b	Pu ^c	1.1	6
60	^{241}Am	0.07	8.5×10^{-3}

^aDetectability limit at the 3σ level for 390 g of low-density wastes in a plastic bag.

^b ^{241}Am contribution before analysis.

^cPlutonium with 94% ^{239}Pu and 6% ^{240}Pu .

it is the forte of the systems discussed here to prove the absence of activity above 10-nCi/g levels, not necessarily to provide a highly quantitative assay at these levels. However, as activity levels increase to the point where the high-energy gamma rays can be used for analysis--e.g., above ~36 nCi/g for MEGAS--a much more quantitative measurement results. This is not to say that accurate results cannot be obtained using the 17-keV region. On the contrary, such results can be obtained under routine plant conditions, but only after certain correct assumptions are made regarding the wastes. These assumptions include wastes of only low-density character (no glass or metal) and no lumps of transuranic materials.

An extensive series of standards measurements has been performed with the MEGAS to determine assay precision. Four standards (4 nCi/g, 14 nCi/g, 100 nCi/g, and 450 nCi/g) were repeatedly counted with the MEGAS over a period of several weeks. The mass of the 0.06-m³ sample varied from 3.7 to 7.0 kg and the count times were either 200 or 500 s. The results, shown in Table IV, indicate that the measurement precision at or below 10 nCi/g is ~10% while at approximately 100 nCi/g it is somewhat worse.

This is due to counting 17-keV x rays at 10-nCi/g levels, hence having excellent (low) statistical errors, and 400-keV gamma rays at 100-nCi/g levels with larger statistical uncertainties. In addition, fluctuations in backgrounds (generally the result of other plutonium samples being moved near the MEGAS during counting) tend to have a much greater absolute effect on the less intense gamma rays than on the high-yield L x rays. Similar precision measurements on the Pancake Counter yielded standard deviations of less than 10%. Here, the sample was removed from the counting chamber after each measurement, then replaced for the next measurement.

Measurement accuracies for MEGAS and the Pancake Counter are inherently limited by the ~±50% errors (or ~±30% for gamma-ray regions) mentioned earlier, due to different spatial locations of the transuranic material. The assumption of no lumps of material, or at least the use of standards that reasonably represent such wastes, must hold true. With these assumptions clearly in mind, ten 0.06-m³ containers of plutonium wastes from the Plutonium Processing Facility at LASL were assayed with the MEGAS after having been first assayed in the 210-2 drum neutron coincidence counter³

TABLE IV
ASSAY PRECISION - MEGAS

Box I.D.	Sample ^a Mass (kg)	Number of Measurements	Assay Time (s)	Assay Results (nCi/g)	Standard Deviation	
					nCi/g	(%)
1	7.0	12	200	3.9	0.4	10
2	5.6	13	500	14	1.5	11
3	5.6	40	500	100	19	19
4	3.7	9	200	450	51	11

^aSamples were contained in 0.06-m³ cardboard cartons.

located at the facility's nondestructive assay station. The boxes were then unpacked, sorted, and reanalyzed in the Pancake Counter as discrete packets (10 to 20 per box). The individual activities of these packets were then summed to give a total activity per box. It was intended that this 'round robin' would provide improved accuracies and confidence in solid transuranic waste assay with both the MEGAS and the Pancake Counter.

Table V lists the assay results for the 10 boxes. The counting times used were: 200 s for the neutron counter, 500 s for the MEGAS, and 10 s for the Pancake Counter. The detectability limit for the neutron counter at Los Alamos is 500 mg plutonium (6% ^{240}Pu) for a 200-s counting time. Since the neutron coincidence counter detects only spontaneous-fission neutrons, no americium or total activity results can

be given for the drum counter. When the neutron counter was able to detect plutonium in the box (No. 10), the results agreed within indicated errors with the MEGAS results. The neutron counter results for the other nine boxes are also consistent with the MEGAS results. In general, the MEGAS results also agree quite well with those obtained with the Pancake Counter. Such comparisons as these demonstrate that not only is solid transuranic waste assay possible at 10-nCi/g levels, but that adequate accuracies can be obtained for both flat geometries and bulk packages.

VI. APPLICATIONS

The Pancake Counter was installed in a sorting glove box line at the LASL Plutonium Processing Facility in September, 1973. MEGAS was installed in the Facility's non-destructive counting room in June, 1974.

TABLE V
ASSAY RESULTS--ROUND ROBIN

Box I.D.	Plutonium (mg)		Activity (nCi/g) ^a	
	Neutron ^b Counter	MEGAS ^c	MEGAS ^c	Pancake Counter ^d
10	700±200	400±120	2400±720	- ^e
1	<500	130± 40	1400±420	- ^e
31	<500	0.14	<2.0	<1.0
34	<500	0.04	<0.4	<1.0
10A	<500	1.1	7.6 ± 5.0	1.3±0.6
2	<500	0.9	13.0 ± 7.0	12.0±6.0
1A	<500	30.0	210.0 ± 70	220±110
4	<500	0.3	3.0 ± 1.5	6.3±3.2
RTI	<500	0.2	3.5 ± 1.8	6.1±3.1
RTC	<500	0.4	5.1 ± 2.6	5.9±3.0

^aTotal activity per gram of wastes; plutonium and americium.

^bCount time = 200 s.

^cCount time = 500 s.

^dCount time = 10 s.

^eBox contained much metal, hence negating pancake counter results.

Since their initial installation, both instruments have performed reliably with almost no down time for instrument repair and/or recalibration. The results of their many months of routine service are covered elsewhere.¹³ Summarily, the instruments aided immensely in the waste sorting and characterization studies being conducted at LASL in support of ERDA waste management programs. The MEGAS is now used routinely to assay all low-density room-generated wastes that are produced by the plutonium facility. The monthly throughput with MEGAS is ~155 packages (0.06 m³ each), containing ~770 kg of waste material. The counting time for the MEGAS (now 200 s) is such that 10 times that throughput can be easily accommodated. The utility of the MEGAS is that all containers of room trash can be reliably assayed. Disposition of the waste--retrievable storage or burial--can now be determined by the assay and need no longer solely depend on administrative decision. Economic analyses¹³ have shown that sorting room trash with the MEGAS may yield up to 70% savings over administratively assigning all room trash to retrievable storage.

Besides ²³⁹Pu wastes, MEGAS has also been calibrated for ²³⁸Pu, ²³⁵U, and several fission products (e.g., ¹³⁷Cs). While the analysis cannot presently be accomplished for all of these isotopes (plus ²³⁹Pu and ²⁴¹Am) at once, it can be done separately with good accuracy and detectability. Since the person in charge of the waste generation generally knows the isotopes contained in his wastes, it is not necessary to attempt a purely 'black box' assay.

Wastes containing ²³⁸Pu at LASL are indicated by the presence of 17-keV x rays and the absence of any 60-keV ²⁴¹Am activity. Such wastes also exhibit a 43-keV gamma-ray characteristic of ²³⁸Pu decay. For low-level (10-nCi/g) wastes, the L-x rays at 17 keV are used for quantitative

analysis. The calibration can be based on the ²³⁹Pu data on a nCi basis knowing that the L x rays are ~2.4 times more prolific from ²³⁸Pu than from ²³⁹Pu.¹⁰ Hence, detectability for ²³⁸Pu is 2.4 times lower (better) than that obtained for ²³⁹Pu using the 17-keV region and on a nCi basis.

Boxes containing ²³⁵U have been measured with the MEGAS. Using the 186-keV gamma ray from ²³⁵U decay, detectabilities (3σ, 500 s) of ~12 mg ²³⁵U in 0.06-m³ of wastes are possible. For 4 kg of waste, this is equivalent to ~4 pCi/g. The high-energy gamma ray of ²³⁵U, coupled to its high specific branching ratio (~52% per alpha decay), is responsible for the improved detectability over ²³⁹Pu analysis on an activity basis. The usual ±30% error applies.

Similarly, for ¹³⁷Cs and other high-energy gamma emitters (e.g., ⁶⁰Co, ²²Na, etc.), MEGAS is capable of quantitative assays. In these cases, the pulse-height-analysis system in MEGAS is adjusted to measure the gamma-ray intensity specific to the isotope of interest (662 keV for ¹³⁷Cs, etc.). For such radioisotopes, MEGAS is capable of ~3-pCi/g detectability for 4 kg of waste. This detectability was measured at the 3σ level in 500 s. Typical accuracy is ±30%.

If, for example, the ¹³⁷Cs is combined with plutonium, a quantitative assay for plutonium is still possible. The analysis is first performed for ¹³⁷Cs. Then the appropriate ¹³⁷Cs-induced background is removed from the plutonium-ameridium regions (17, 60, and 400 keV). This induced background also includes the ~32-keV x rays from ¹³⁷Ba, the ¹³⁷Cs daughter. Once this background is removed, the remaining intensity is considered due to plutonium and ameridium. Under such circumstances the detectability for plutonium (3σ level, 500 s) degrades to 1 nCi/g (using the L x-ray region) when 80 μCi of ¹³⁷Cs is present. In other words, MEGAS can tolerate 80 μCi of

^{137}Cs and still be capable of 1-nCi/g detectabilities. Larger assay errors result, however.

VII. CONCLUSIONS

Both the MEGAS and the Pancake Counter have demonstrated detectabilities well below the necessary 10-nCi/g level. The systems provide reasonably quantitative results, particularly in light of the types of samples they are intended to measure and their stated limitations. Using the L x-ray region, the counters provide an excellent "go/no go" tool for effective waste management control. Using the higher energy gamma-ray regions, improved accuracies and confidences are possible, similar to other gamma-ray-based detection systems. The instruments are not intended as a panacea for all types of transuranic wastes generated by the nuclear industry, particularly the high-level, or fission-product-contaminated, wastes and those wastes of high density (metal, etc.).

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