GAMMA-RAY SCANNING SYSTEM FOR BARRELS
CONTAINING PLUTONIUM WASTE*

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SUMMARY

A system designed to perform quantitative assay of plutonium waste material in 30 to 55 gallon barrels is described. The system uses up to eight 2 in. x 2 in. NaI(Tl) detectors to make a detailed gamma ray scan of the barrel to be assayed. Barrels of 55 gallon size containing ^{239}Pu waste and 30 gallon barrels containing ^{238}Pu waste (from $^{238}\text{PuO}_2$ isotopic heat source production) have been scanned with this system. Experience has shown that a barrel assay can be performed in 10 to 15 minutes to an accuracy of $\pm 10\%$. The sensitivity of the system is one gram or better for ^{239}Pu or ^{238}Pu in low to medium density waste material.

INTRODUCTION

The problem of safeguarding fissionable nuclear material is becoming ever more important as a result of the rapidly expanding worldwide nuclear power industry. When the Baruch plan for international control of nuclear materials was advanced in 1946, the amount of material susceptible to diversion was negligible compared to existing military stockpiles. Today, however, the situation is drastically different; the U. S. Atomic Energy Commission estimates that by the late 1970's, nuclear power reactors around the world will be producing more than 100 Kg of plutonium per day. In other words, sufficient fissionable material will be produced over the world for the potential production of a substantial amount of the world's power requirements--or, alternatively, for the potential production of several nuclear weapons per day!

It is becoming increasingly apparent that physical nondestructive methods of detection and quantitative assay of fissionable materials can provide the best technical basis for an effective system of material accountability.⁽¹⁾ Most, if

not all, of the physical techniques required for such assay are in principle well known and have been employed in other nondestructive testing applications and geophysical exploration. However, most of these techniques have not been exploited to their full potential by the nuclear industry; in fact, the nuclear industry has relied almost entirely on destructive chemical analysis of small samples to determine quantities of materials for inventory and quality control. Such chemical analyses are relatively slow, expensive, and involve the inherent uncertainty that the samples analyzed may not be representative of the whole. In particular, chemical sampling procedures are almost totally inadequate for scrap and waste generated in the production of nuclear fuel. On the other hand, physical assay methods based upon the detection of penetrating nuclear radiations are relatively fast and, if properly done, well suited to the measurement of inhomogeneous mixtures.

This paper describes a system utilizing an array of eight NaI(Tl) detectors to scan containers up to 55 gallons in volume containing low to medium density nuclear fuel production waste. Although the concept of gamma-ray scanning waste drums is as old as the nuclear industry itself, the technique in general has not been advanced to employ new ideas in measurement techniques and instrumentation.

SYSTEM DESCRIPTION

Design criteria for the barrel scanning system included:

1. Rapid and accurate quantitative assay of heterogeneous waste containing from a few grams to hundreds of grams of plutonium.
2. Detection of the presence of dense materials that could cause a serious underestimate of the plutonium content.
3. An optional capability for the determination of the spatial distribution of

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plutonium within the barrel.

4. The system must be compact and easily transportable for field use.

The system developed to fulfill these criteria is shown in Fig. 1. Eight shielded 2 in. x 2 in. NaI(Tl) detector modules can be seen on the left of the figure. Each module has fixed vertical and variable horizontal lead collimators.

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Fig. 1. Over-all view of the eight channel scanning system.

Spacing between module centers is variable between 4 in. and 6 in. Opposite the detector stack is an array of eight collimated 100 μ Ci ^{22}Na sources used to make transmission measurements on a detector-by-detector basis. While measuring the gamma activity of the barrel, these sources are rotated 180° so as to be shielded from the detectors. The barrel to be scanned is placed on a rotating platform between the detector and ^{22}Na arrays.

Rotating the barrel while counting is a major factor in reducing the dependence of detector response on the radial distribution of plutonium and is frequently used when counting bulk samples.^(2,3,4) For example, the radial response of a detector located one barrel radius

from the periphery of the barrel will vary about $\pm 15\%$ for material densities less than about 0.1 g/cm³. If the detector is located closer to the barrel or if the material is more dense, the radial response variation will be even greater.

Experimental studies of spatial response were performed for barrel rotation combined with horizontal and vertical collimation. Figure 2 shows the two dimensional case, i. e. the Pu and detector are coplanar and horizontal collimation is used. The effect of the collimation is to selectively reduce the detector response to material located near the periphery of the barrel.

Fig. 2. Two-dimensional schematic for gamma ray assay in cylindrical containers. L_1 , L_2 , and D are varied to achieve flat response within the plane.

The collimation angle and displacement, D , can be adjusted to yield an almost flat radial response for barrels showing attenuations of as much as 100 for ^{239}Pu gamma rays across their diameters. For a given attenuation, there is a collimation angle that optimizes the spatial response of the detector; higher attenuation requires narrower collimation for greater suppression of peripheral response thereby compensating for the more highly attenuated central response. Previous schemes for waste container gamma ray scanning have attempted to flatten the detector response by moving the detector some distance from the container so that the material will appear approximately as a point source.^(3,4) This will reduce $1/R^2$ effects of material distribution within the container, but does nothing for response variations due to self absorption.

In order to flatten the three dimensional response from a 55 gal. barrel, the eight detectors are positioned 6 in. between centers and fixed vertical collimation (4 in. of lead beyond the crystal) along with variable horizontal collimation is used. The barrel is counted in two

positions displaced vertically from each other by 3 in. The horizontal collimation of each detector is individually adjusted on the basis of the measured attenuation at that detector position. (For material densities less than about 0.2 g/cm³, no horizontal collimation is required.) Using this double scan technique, the response to plutonium located at any position in the barrel is uniform to better than $\pm 10\%$ for material densities up to at least 0.8 g/cm³. For smaller 30 gal. drums, a single position scan is used with the vertical separation of the detectors set at 4 in. between centers.

One can also obtain the radial distribution of plutonium in the barrel, if desired, from the measurement of counting rate as a function of the angle of rotation. The multiscaling feature of a multichannel pulse-height analyzer offers a convenient means for collecting and displaying this type of data. Figure 3 shows the counting rate from a ²³⁹Pu sample located at three radial

Fig. 3. Multiscaler plots of response from a point Pu sample in a rotating barrel.

positions in a barrel as a function of the angle of rotation. The widths of the peaks in the time spectra are a function of the radial position of the sample and hence provide information on the radial distribution of plutonium within the barrel.

ELECTRONICS

A block diagram of the system electronics is shown in Fig. 4. The detectors are Harshaw "Integral Line" 2 in. x 2 in. NaI(Tl) crystals mounted on RCA 8053 photomultiplier tubes.

Fig. 4. Block diagram of barrel scanning electronics.

The gains of the photomultipliers were initially matched to within 10% at an operating voltage of 1 K. V. In order to reduce the low energy (60 keV) background from ²⁴¹Am present in Pu, each detector was wrapped with 0.030 in. of cadmium.

The detector signals are fed to preamplifiers packaged four per 7 in. x 4 in. x 1 3/4 in. module. Each preamplifier consists of a charge sensitive input stage followed by a gain-of-ten line driver as shown in Fig. 5. The input is protected from large voltage transients by the diodes in the gate of Q₁. The gate-to-source bias of Q₁ is set to about -0.5 V by means of the 1K pot in the base circuit of Q₂. Output pulses are linear to about 6 volts, and are RC-CR shaped with a rise time of about 0.5 μ s and a decay of 0.75 μ s.

Sufficient gain is incorporated in the preamplifier to obviate the need for further pulse amplification. However, it was found convenient to use an adjustable gain buffer following each preamplifier in order to easily match the pulse

Fig. 5. Preamplifier schematic diagram.

heights from each detector. A diagram of the buffer amplifier is shown in Fig. 6; eight amplifiers are contained in a single width NIM module. The gain of each amplifier is adjustable from 0 to X2 by front panel controls.

Fig. 6. Buffer amplifier schematic diagram.

Pulse-height analysis is performed in a dual-width NIM module containing eight independent single channel analyzers with common threshold and window width controls. A simplified diagram of a single section of the analyzer is shown in Fig. 7. The base-line restorer at the analyzer input has been found helpful in reducing the effects of low frequency noise; however, the restorer diodes must be closely matched between analyzer sections in order that each input will have the same DC offset voltage.

Integrated circuit differential comparators are used for pulse-height discriminators resulting in sharp and stable discrimination levels. The lower threshold is adjustable from 0 to 5V, and the window width is independently adjustable from 0 to 2.5 V. A temperature stabilized zener diode provides the window width reference voltage. Voltage followers are used to isolate the threshold and window width reference levels from loading effects. A peak detector is used to gate the analyzer output pulse in order to reduce time jitter, although this feature is not required for the present application. Output pulses from each analyzer are fed to individual scalers or to an eight input multiscaler unit. An "OR" signal is also available for scaling the total number of output pulses generated by all eight analyzers.

PLUTONIUM WASTE DRUM MEASUREMENTS

The system has been used to assay 55 gal. barrels containing ^{239}Pu mixed with various materials representative of process wastes. For these measurements, the group of gamma rays resulting from ^{239}Pu decay with energies between about 330 keV and 414 keV were used; a typical NaI(Tl) pulse height spectrum is shown in Fig. 8. The 511 keV annihilation radiation from the ^{22}Na sources was used for barrel transmission measurements.

As noted previously, the most uniform volume averaged response for 55 gallon barrels is obtained when 6 in. spacing between detector

Fig. 7. Simplified schematic of one single-channel analyzer section.

centers is used and the barrel is counted in two positions separated vertically by 3 in. In this arrangement, the top and bottom detectors contribute only slightly to the total count rate and are used only for end-effect corrections on full barrels. A single attenuation measurement is normally made with the barrel located midway between the two counting positions. Calibration curves such as shown in Fig. 9 are then used to determine the horizontal collimator settings and detector sensitivities on the basis of the attenuation measurements. The shape of the sensitivity curve shown in Fig. 9 was calculated by numerically integrating the response of a collimated cylindrical detector to a uniformly distributed self-attenuating source. Experimental measurements made on high and low density barrels containing known amounts of ^{239}Pu were then used to determine the absolute sensitivities at two points on the curve.

Fig. 8. NaI(Tl) pulse height spectrum of ^{239}Pu gamma rays.

In order to test the assay accuracy of the system, a series of "calibration" barrels (5) prepared by the Dow Chemical Co. Rocky Flats Plutonium Facility containing known amounts of

TABLE I

ASSAY OF STANDARD ^{239}Pu "WASTE" BARRELS

Matrix Material	^{239}Pu Content		M / K
	Known (g)	Meas. (g)	
Graphite	60	59	.98
"	195	209	1.07
"	145	151	1.04
Raschig Rings	40	41	1.03
"	185	168	.91
"	95	75	.79
"Washables"	10	9	.90
"	160	142	.89
"	90	77	.86
"Combustibles"	10	11	1.10
"	175	168	.96
"	165	155	.94
"Wet Combustibles"	29	26	.90
"	166	123	.74
Resin	25	28	1.12
"	110	106	.96
Plexiglas	75	62	.83
Total Pu	1735	1610	Mean=.942 $\rho=.107$

Fig. 9. Calibration curves for ^{239}Pu barrel assay.

^{239}Pu in various matrix materials were measured. The density of the material in these barrels was generally in the range 0.1 g/cm^3 to 0.5 g/cm^3 . Results of the measurements (6) are shown in Table I. The standard deviation between the known and measured ^{239}Pu content was 11%, with the largest deviation being 26%. The fact that the measured values tend to be somewhat lower than the known values is attributed to the fact that the barrels appeared to contain large voids so that the measured attenuations were in many cases too low.

The scanning system has also been used to assay about two hundred 30 gallon drums containing ^{238}Pu residues generated in connection with the SNAP and artificial heart-pump programs. For these smaller barrels, seven detector modules spaced 4 in. between centers were used and only one scan position was required. A typical ^{238}Pu NaI(Tl) spectrum is shown in Fig. 10. The 766 keV gamma-ray line was used for assay rather than the more intense 153 keV line due to the much larger attenuation corrections required for the lower energy gamma rays. Since the relative intensity of the 871 keV gamma ray produced by the $^{14}\text{N}(\alpha, p)^{17}\text{O}^*$ reaction varies greatly depending upon the nitrogen content of the Pu compound, a rather narrow

single-channel window was used in order to reduce interference from this line. An additional measurement was made with the analyzer window set above the 871 keV line in order to correct for Compton background contributed by higher energy lines arising from various (α, n) reactions. Transmission measurements were made using the 1274.5 keV gamma ray from ^{22}Na . Since the waste drums also contained varying amounts of ^{22}Na produced by the $^{19}\text{F}(\alpha, n)^{22}\text{Na}$ reaction, two measurements were made at 1274.5 keV, one with and the other without the transmission sources.

At the start of the ^{238}Pu assay program, about 200 small residue containers ranging in size from 4 in. diameter x 2 in. tall to 8 in. diameter x 14 in. tall were carefully assayed with a high resolution Ge(Li) scanning system. The 200 small containers were then loaded into thirteen

Fig. 10. NaI(Tl) pulse height spectrum of ^{238}Pu gamma rays.

30 gallon drums and re-assayed as outlined above. The agreement between the piecemeal and total drum assays is shown in Table II. Based on these results, all remaining residue containers were placed in 30 gallon barrels and the barrels were then assayed to obtain measured accountability values for the residues.

Experience gained with the eight channel NaI system has shown that a barrel assay can be performed in 10 to 15 minutes, depending on the degree of automation in data and sample handling available. The accuracy is about $\pm 10\%$ at the one sigma level and the sensitivity of the system is one gram or better for either ^{239}Pu or ^{238}Pu in bulk waste containers.

A somewhat more sophisticated system employing a single Ge(Li) detector and the transmission-corrected segmented scan principle has also been developed for assaying plutonium and uranium scrap and waste. (7)

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TABLE II
COMPARISON OF ^{238}Pu DRUM ASSAY AND PIECEMEAL CAN ASSAY

Drum No.	Number of Containers in Barrel	NaI Barrel Assay (g Pu)	Ge(Li) Piecemeal Assay (g Pu)	Ratio: Barrel/ Piecemeal
1	30	33.3	32.7	1.02
2	32	0.5	0.3	*
3	18	45.3	39.1	1.16
4	22	1.0	0.6	*
5	2	34.9	39.4	0.89
6	4	37.9	39.6	0.96
7	11	41.1	39.1	1.05
8	13	38.3	32.8	1.17
9	10	32.9	34.5	0.94
10	8	30.0	34.2	0.88
11	12	30.5	26.6	1.15
12	9	34.1	39.6	0.86
13	9	15.8	17.3	0.91

Mean = $\overline{1.00}$

(σ) deviation = 0.12

* The two low level drums are not included in the calculation of σ since the system was not set up to measure these low level wastes accurately. These drums were just measured to check for any gross errors.

ONE OF 2 IDENTICAL CIRCUITS PER BOARD
2 BOARDS PER UNIT

ONE OF 8 IDENTICAL CIRCUITS

GAIN = 0 TO 2