ORNL-TM-4249

DESIGN OF A SYSTEM FOR THE NONDESTRUCTIVE ASSAY OF ^{233}U IN WASTE DRUMS

V. A. DeCarlo

OAK RIDGE NATIONAL LABORATORY
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ORNL-TM-4249

Contract No. W-7405-eng-26

CHEMICAL TECHNOLOGY DIVISION

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AUGUST 1973

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ABSTRACT

A system for the nondestructive assay of waste drums containing ^{233}U has been installed in Bldg. 3019. This system employs two 3 x 3 in. NaI scintillation detectors, and standard counting techniques and analyses of the 2.6-MeV gamma radiation emitted by ^{208}Tl. Thallium-208 is a product of the decay of ^{232}U, which is usually present as an impurity in ^{233}U. Results show that the system is capable of determining gram quantities of ^{233}U in a 55-gal rotating drum. Standard deviations of the results are also reported.

INTRODUCTION

It has been recognized for some time that nondestructive assay of the ^{233}U content of waste drums can be achieved using a scintillation counting technique.[1] This technique is applicable because ^{233}U usually contains parts-per-million quantities of ^{232}U and because ^{208}Tl, which emits a 2.6-MeV gamma ray, is a product of the ^{232}U decay chain. The decay chain is as follows:

$$^{232}U \rightarrow {}^{228}Th \rightarrow {}^{224}Ra \rightarrow {}^{220}Rn \rightarrow {}^{216}Po \rightarrow {}^{212}Pb$$

$$\rightarrow {}^{212}Bi \begin{cases} \nearrow {}^{212}Po \searrow \\ \searrow {}^{208}Tl \nearrow \end{cases} {}^{208}Pb$$

The 2.6-MeV gamma ray of ^{208}Tl can be measured using NaI scintillation detectors, and from this measurement calculations can be made to determine the ^{233}U content in the waste drum. The counting rate can be improved by a factor of 3 by including the single and double escape peaks at 2.1 and

1.6 MeV, respectively, in addition to the 2.6-MeV photo peak of ^{208}Tl. This is only applicable when there are no other gamma photo peaks in the 1.6- to 2.1-MeV range. It should be noted that ^{232}Th also yields ^{208}Tl. However, calculations show that 1 g of ^{233}U containing 10 ppm of ^{232}U, aged approximately 1 year after chemical processing, is equivalent to 6 kg of thorium. Therefore, the thorium content in most waste drum measurements will have little effect on the results.

The chemical processing of ^{233}U removes essentially all of the daughters of ^{232}U, but the ^{208}Tl activity gradually grows back into the ^{233}U as shown in Fig. 1.[2] The age of the waste material after separation is, therefore, important because the counting rate associated with the ^{233}U increases due to the increase in ^{208}Tl activity with time. Material aged less than 16 days should be avoided because of the different rates of ^{208}Tl buildup as shown in Fig. 1. Two different growth curves are observed because the ion exchange system removes the ^{228}Th but leaves variable quantities of ^{224}Ra, ^{220}Rn, and ^{212}Pb. The solvent extraction system, however, removes part of the ^{228}Th and more of the remaining daughters. Also, the fact that the ^{208}Tl background in Bldg. 3019 is equivalent to 0.5 g of ^{233}U aged 16 days after separation could affect the accuracy of the results for younger ^{233}U. The material can be analyzed, however, if enough detailed information on the waste is available.

Variables that can influence the accuracy of the results of the installed system are:

(1) Ratio of ^{232}U to ^{233}U.
(2) Age of waste.
(3) Counting geometries.
(4) Counting losses.
(5) Attenuation coefficient.
(6) Natural thorium content.

Some of these effects can be avoided by using an archive sample from the batch of material from which the waste is generated. This method assumes that the age of the archive sample is identical to that of the waste. When similar geometries are used for counting the archive sample

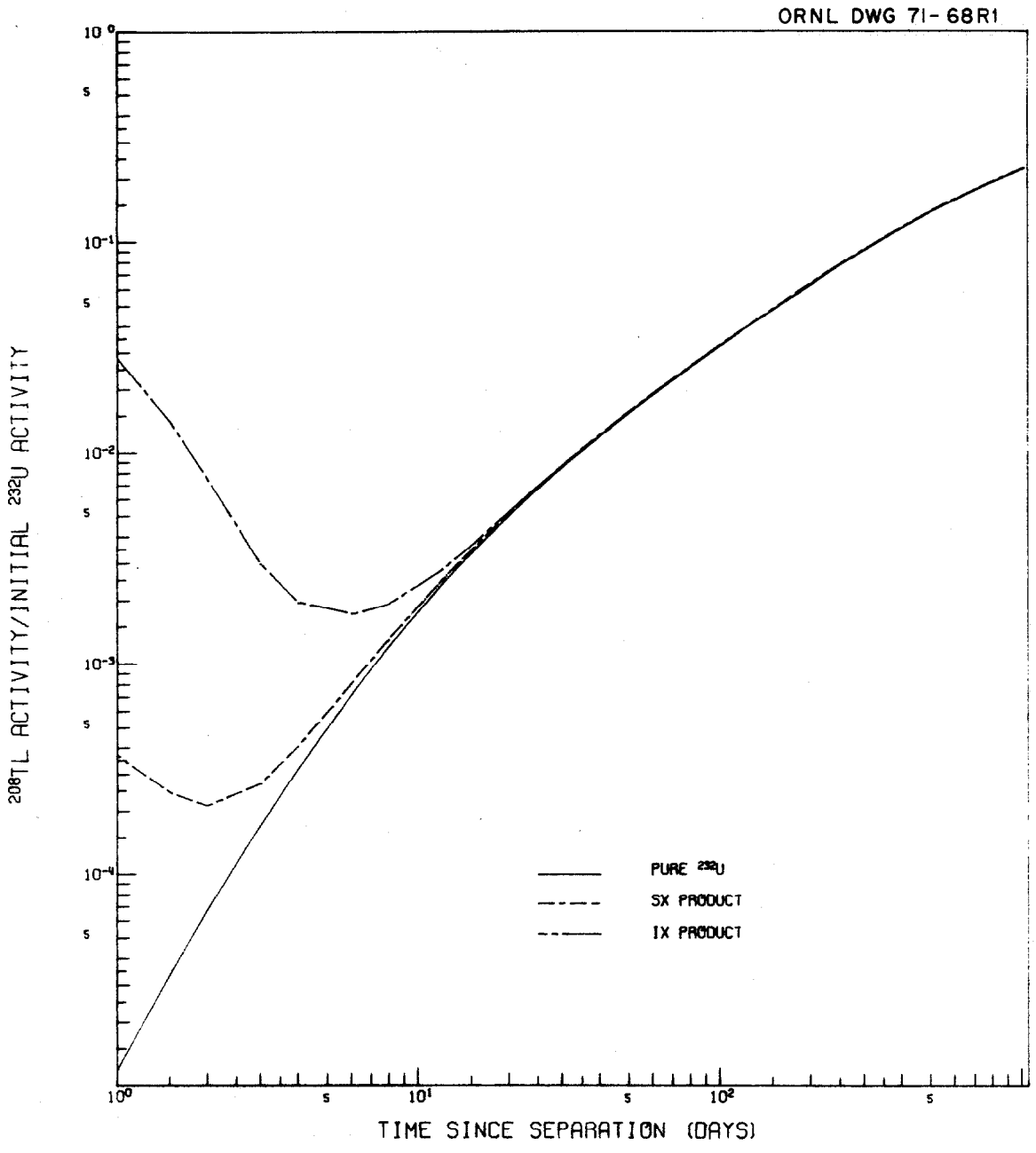

Fig. 1. The Effect of Postseparation Time and Type of Separation on the Radioactivity of ^{208}Tl per Unit Initial Radioactivity of ^{232}U.

and the waste drum, the results become directly comparable. The archive sample is placed and counted in the center of an empty drum. The results should be adjusted by taking into account the attenuation coefficient of the waste, which can be obtained by analyzing the archive sample in four positions — behind and in front of the waste drum and an empty drum. The calculations for determining the quantity of ^{233}U in the waste drum are given in Appendix I.

SYSTEM DESCRIPTION

The system consists of two 3 x 3 in. NaI scintillation detectors mounted in 2-in.-thick cylindrical lead shields positioned so that the center of each crystal is located 1 in. above and below the center of the upper and lower half of the drum respectively. The drum containing waste material is mounted on a table which rotates at 1 rpm. The housing for the detectors is mounted on tracks to allow for a variable distance between the waste drum and the detectors. Figure 2 is a photograph of the assay facility with a standard calibrating drum in place. The various holes used for calibrating can be seen through the top of the drum. The wire positioner for the standard samples is attached to the outside of the drum. The standard 55-gal drum is filled with foam glass. The spectrum is accumulated by a multichannel analyzer. The total count under the ^{208}Tl photo peaks is obtained automatically by an integrator with preset limits. Output is obtained on a typewriter. Figure 3 shows some of the instruments used with the system.

DISCUSSION OF DATA

Since the waste can be located anywhere within the waste drum, the drum is rotated around its vertical axis to average out the effect of distance between the material and the detectors. The distance between the waste drum and the detectors was determined by making a trade-off between the effects of waste location and detectable quantities of uranium because the effect of waste location in the drum diminishes with increased distance but the quantity of ^{233}U that can be detected is decreased. A distance of 52 in. between the center of the drum and the

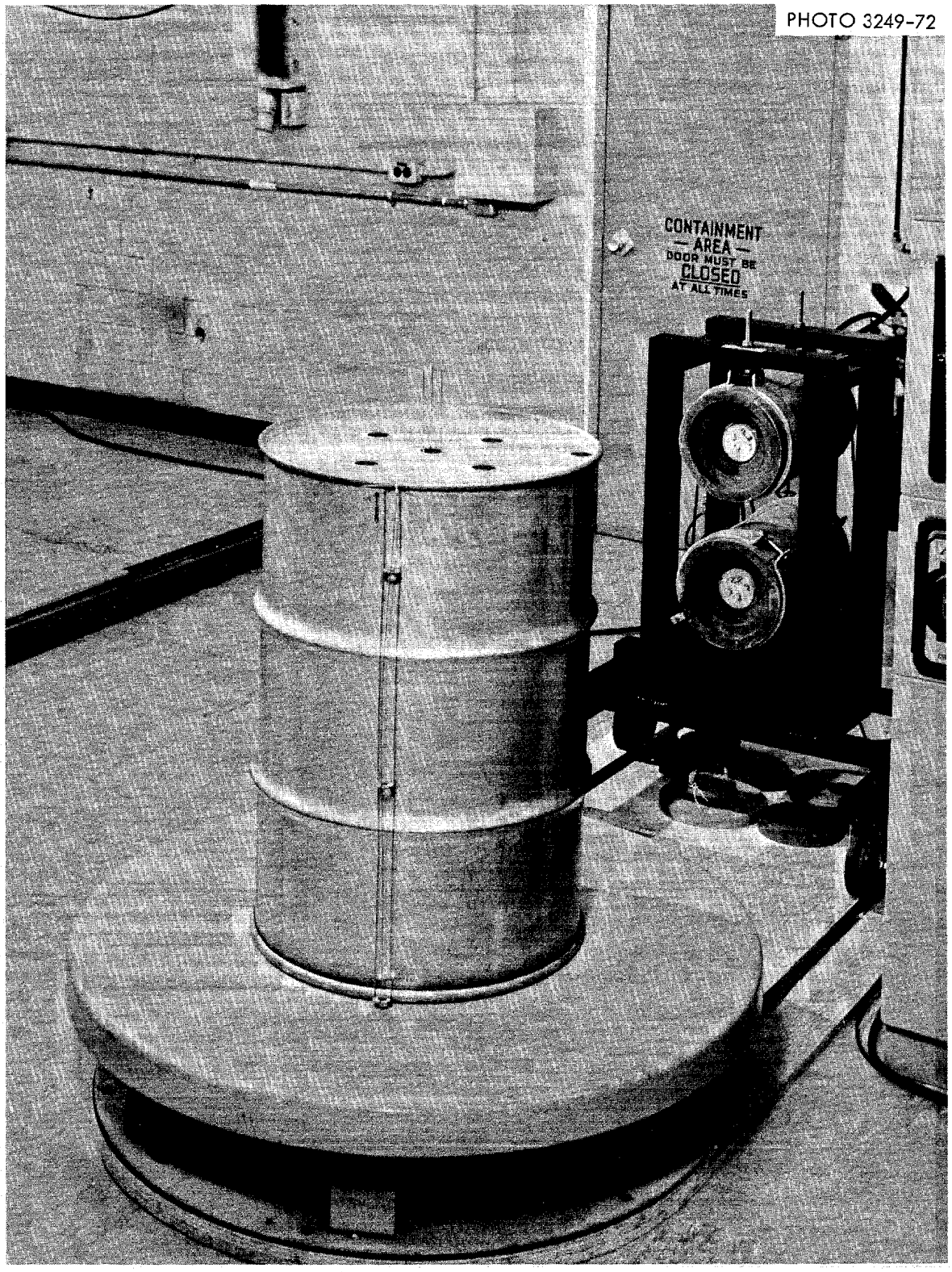

Fig. 2. ^{233}U Assay Facility for Waste Drums.

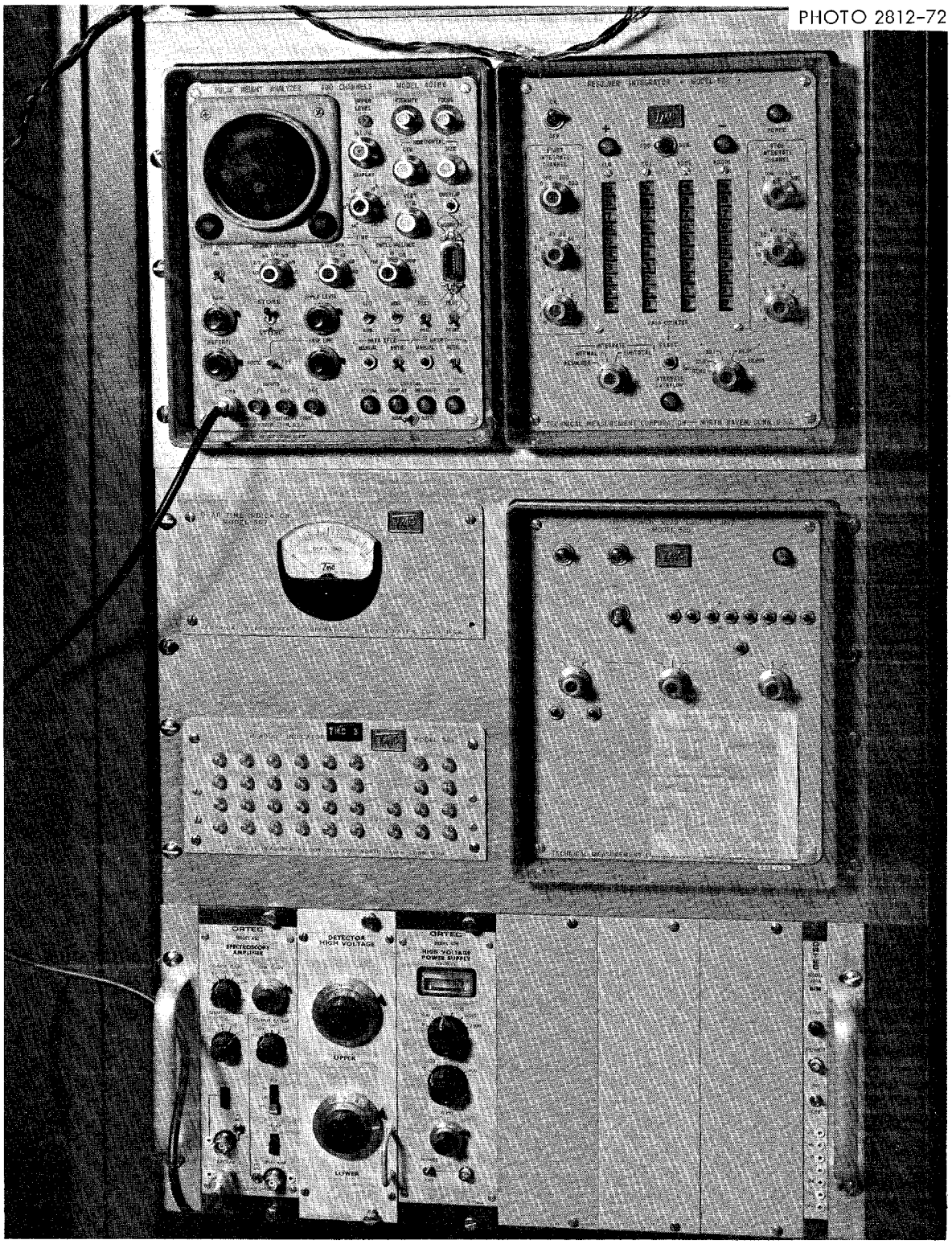

Fig. 3. Instrument Panel for ^{233}U Assay Facility.

detectors, with the crystals separated by 13-1/4 in., was determined to be adequate for this system.

To determine the standard deviation of the quantities of ^{233}U found in waste drums, the random errors in the following items need to be taken into consideration:

1. Location of waste within the drum.
2. Thallium-208 content of the drum.
3. Background.
4. Readings for obtaining the attenuation coefficient of the waste drum.
5. Weighing of archive sample.

To obtain the standard deviation of a waste drum, known quantities of ^{233}U were placed at various locations within the drum (see Tables 1-3) and counted twice in succession with the drum rotating and repeated for three ^{208}Tl radioactivity levels. Rotation of the drum was started at various positions to include the effect of starting the counting from any position. The calculations for determining the standard deviation are given in Appendix II.

The inaccuracy resulting from the location of the material can be decreased by locating the archive sample in the section of the standard drum corresponding to that where the greatest concentration of ^{233}U is believed to be present in the waste drum. The degree of improvement will depend on the precision of the operation.

As a check of this method, simulated waste drums were loaded with known amounts of ^{233}U, in the form of $^{233}UO_2$, in various locations. Results of these runs are given in Table 4.

To date, five waste containers have been analyzed. The results, along with their calculated standard deviations, are summarized in Table 5. The gamma-ray spectrum obtained from one of the drums is shown in Fig. 4. The ^{208}Tl portion of the gamma spectrum used for determining the ^{233}U content is indicated.

Table 1. Position of 1-g Samples for Statistical Analysis of Waste Location

Distance from Center of Drum (in.)	Height from Bottom of Drum (in.)	Starting Position	Radioactivity (counts/10 min)
8 1/16	7	15°	30,290 30,268
9 7/16	21 15/16	114°	30,196 30,256
5 7/16	7 3/4	311°	30,107 30,403
4 1/16	1/4	285°	28,856 28,866
10 5/8	25 1/2	162°	29,819 29,882
10 7/16	15 15/16	104°	30,669 30,388
4	24 3/16	46°	29,914 29,948
7 11/16	28 1/2	302°	30,468 30,576
6 1/16	3 5/16	227°	29,335 28,911
2 9/16	11 3/8	318°	31,120 31,102

Table 2. Position of 5-g Samples for Statistical Analysis of Waste Location

Distance from Center of Drum (in.)	Height from Bottom of Drum (in.)	Starting Position	Radioactivity (counts/10 min)
6 1/8	13 3/16	232°	118,639 118,063
4 1/16	32 11/16	23°	111,555 111,552
1 3/8	9 7/16	200°	114,548 114,527
9 15/16	7 9/16	70°	113,642 113,378
5 3/8	5 13/16	356°	111,639 111,735
9	28 5/16	138°	113,129 113,257
10 7/16	22 7/8	255°	118,821 118,981
8 5/8	27 1/2	314°	114,839 115,300
4 3/16	20 1/4	283°	120,224 119,864
10 7/8	2 3/4	154°	110,877 109,779

Table 3. Position of 12.5-g Samples for Statistical Analysis of Waste Location

Distance from Center of Drum (in.)	Height from Bottom of Drum (in.)	Starting Position	Radioactivity (counts/10 min)
10 15/16	29	30°	269,788 270,944
1 1/4	24 7/8	114°	277,217 277,116
6 5/16	11 9/16	262°	281,024 280,672
5 1/8	17 11/16	165°	280,479 280,623
7 3/16	31 15/16	320°	269,232 269,459
9 3/8	4 5/16	176°	266,075 265,163
10 1/2	10 5/8	40°	277,821 276,524
10 3/16	3 1/16	21°	263,963 263,735
7 15/16	9 5/8	345°	279,186 279,296
11 1/16	29 1/2	149°	266,493 266,385

Table 4. Summary of Results Obtained for Simulated Waste Drums

Drum No.	UO_2 Content (g)	UO_2 Analyzed (g)
RUTL	17.25	17.47
SYN-1	30.2	31.39
SYN-2	8.2	8.42

Table 5. Summary of Results Obtained for Actual Waste Drum Analyses

Drum No.	Quantity of ^{233}U (g), 95% Confidence Interval
OX-4	18.8 ± 1.29
OX-21	23.98 ± 1.54
OX-22	29.57 ± 1.78
OX-23	17.12 ± 1.11
OX-24/25	31.76 ± 2.03

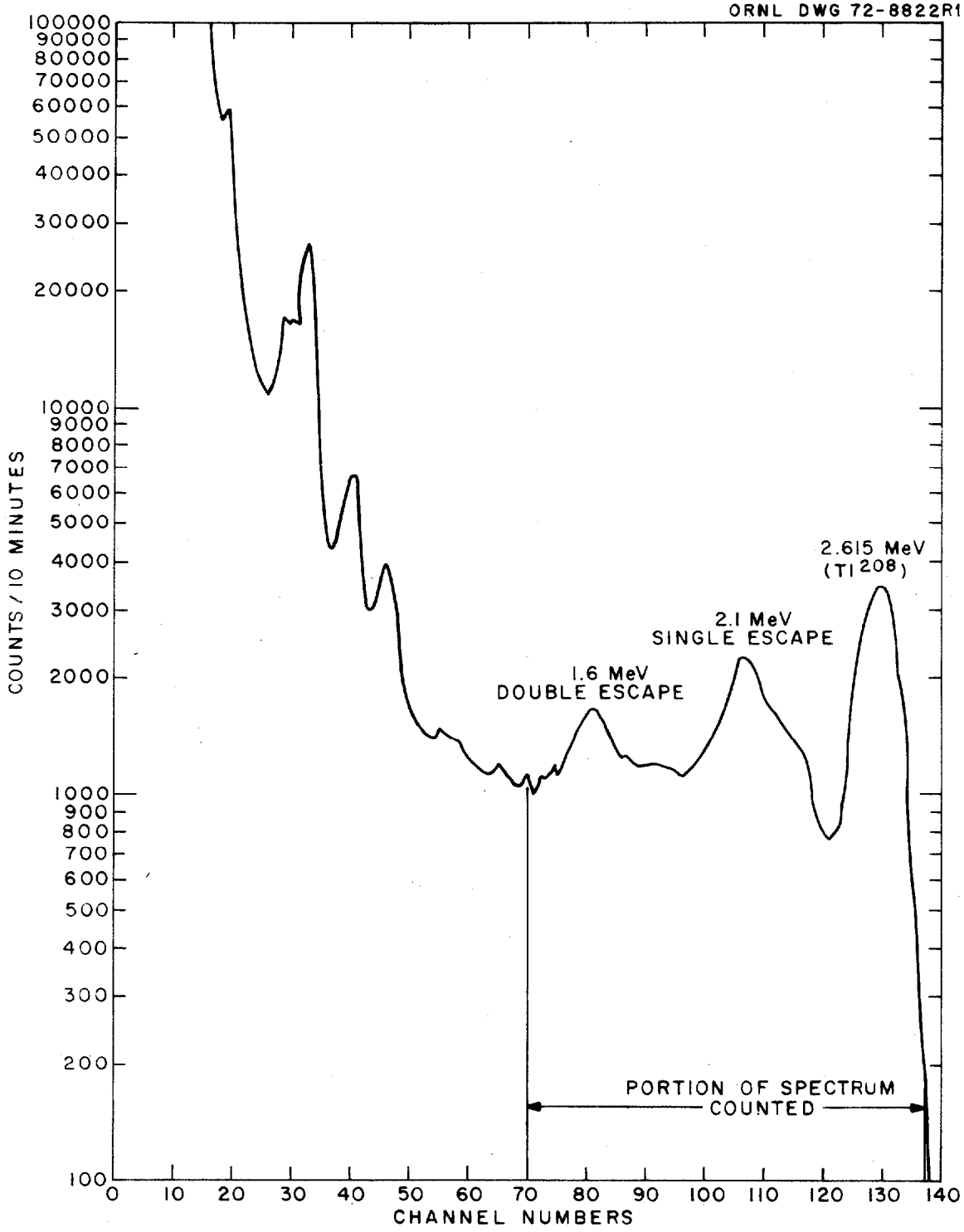

Fig. 4. Gamma Ray Spectrum of Waste Drum.

ACKNOWLEDGMENTS

The author is grateful for the helpful suggestions made by J. P. Nichols, D. Joy, and D. W. Magnuson, and for the assistance of W. L. Pattison, W. R. Whitson, and W. T. McDuffee in the installation of the facility in Bldg. 3019. I would like to express particular appreciation to J. S. Eldridge, Analytical Chemistry Division, and T. L. Hebble, Mathematics Division, for their analytical assistance.

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APPENDIX I

CALCULATIONS FOR DETERMINING ^{233}U IN WASTE DRUMS USING AN ARCHIVE SAMPLE

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CALCULATIONS FOR DETERMINING ^{233}U IN WASTE DRUMS USING AN ARCHIVE SAMPLE

W = gamma ray count of waste drum

S = gamma ray count of archive sample in empty drum

B = gamma ray count of background

G = grams of UO_2 in archive sample

ABW = gamma ray count of archive sample placed in back of waste drum

AFW = gamma ray count of archive sample placed in front of waste drum

ABS = gamma ray count of archive sample placed in back of an empty drum

AFS = gamma ray count of archive sample placed in front of an empty drum

Attenuation coefficient of waste:

$$\frac{S - \left(\frac{(AFW - ABW) - (AFS - ABS)}{2}\right)}{S} = AC$$

Grams of UO_2 in waste:

$$\left(\frac{W - B}{S - B}\right) \times G \times AC = g\ UO_2$$

Grams of ^{233}U in waste:

$$g\ UO_2 \times 0.875\ (\%\ \text{of U in } UO_2)$$

APPENDIX II

CALCULATIONS FOR DETERMINING STANDARD DEVIATION OF QUANTITY OF ^{233}U IN WASTE DRUM

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CALCULATIONS FOR DETERMINING STANDARD DEVIATION OF QUANTITY OF ^{233}U IN WASTE DRUM

The data given in Table 1 yield the expression

$$21.8 + 0.024\ W,$$

which is the variance due to the location of ^{233}U in the waste drum per minute of counting. From data obtained by counting various ^{208}Tl radioactivity levels at the center of an empty drum, the expression

$$30 + 1.62\ S,$$

which is the variance due to counting the archive sample per minute of count, is obtained. The variance due to weighing of archive sample was found to be 0.04.

SW = weight of archive sample in grams

WW = weight of ^{233}U in waste drum in grams

Therefore,

$$\text{Var (WW)} = \left(\frac{SW}{S}\right)^2 \text{Var (W)} + \left(\frac{W}{S}\right)^2 \text{Var (SW)} + \left(\frac{W(SW)}{S^2}\right)^2 \text{Var (S)},$$

where

$$\text{Var (W)} = (21.8 + 0.024\ W)^2$$

$$\text{Var (SW)} = 0.04$$

$$\text{Var (S)} = 30 + 1.62\ S$$

Standard deviations of WW = $\sqrt{\text{Var (WW)}}$. An approximate 95% confidence interval is given by:

$$WW \pm 2.26 \sqrt{\text{Var (WW)}}.$$

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