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The Statistical Sampling Plan for the TRU Waste Assay Facility

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OPERATED BY
UNION CARBIDE CORPORATION
FOR THE UNITED STATES
DEPARTMENT OF ENERGY

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Printed in the United States of America. Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road, Springfield, Virginia 22161
NTIS price codes—Printed Copy: A03 Microfiche A01

ORNL/CSD/TM-203
Distribution Category UC-23 and UC-70

THE STATISTICAL SAMPLING PLAN FOR THE TRU WASTE ASSAY FACILITY

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ORNL/CSD/TM--203

DE83 016962

Date Published - August 1983

Research sponsored by
Defense Waste Management Program,
Instrumentation Support for TRU Waste Assay
(FTP/A No. ONLWT10)

Union Carbide Corporation, Nuclear Division
operating the
Oak Ridge Gaseous Diffusion Plant • Paducah Gaseous Diffusion Plant
Oak Ridge Y-12 Plant • Oak Ridge National Laboratory
under Contract W-7405-eng-26
for the
Department of Energy

CONTENTS

LIST OF ILLUSTRATIONS

LIST OF TABLES

THE STATISTICAL SAMPLING PLAN FOR THE TRU WASTE ASSAY FACILITY

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ABSTRACT

Due to limited space, there is a need to dispose appropriately of the Oak Ridge National Laboratory transuranic waste which is presently stored below ground in 55-gal (208-ℓ) drums within weather resistant structures. Waste containing less than 100 nCi/g transuranics can be removed from the present storage and be buried, while waste containing greater than 100 nCi/g transuranics must continue to be retrievably stored. To make the necessary measurements needed to determine the drums that can be buried, a transuranic Neutron Interrogation Assay System (NIAS) has been developed at Los Alamos National Laboratory and can make the needed measurements much faster than previous techniques which involved γ-ray spectroscopy. The previous techniques are reliable but time consuming. Therefore, a validation study has been planned to determine the ability of the NIAS to make adequate measurements. The validation of the NIAS will be based on a paired comparison of a sample of measurements made by the previous techniques and the NIAS. The purpose of this report is to describe the proposed sampling plan and the statistical analyses needed to validate the NIAS.

1. INTRODUCTION: SCOPE AND PURPOSE

Oak Ridge National Laboratory (ORNL) transuranic (TRU) waste is presently stored below ground in weather-resistant structures under controlled surveillance (see Figure 1). The retrievably stored waste is contained in 55-gal (208-ℓ) drums. Due to the limited space available for storage (3654 drums) and the number of drums produced to date (≈2000), it would be advantageous and cost effective to reduce the volume of TRU waste retrievably stored. Waste containing less than 100 nCi/g transuranics (alpha-emitting radionuclides of atomic number greater than 92 and half-lives greater than 20 y) may be disposed of in shallow-land burial, while waste containing greater than 100 nCi/g transuranics must be retrievably stored (see Management of Transuranic Contaminated Material, 1982).

Classification of the ORNL waste into "TRU" and "non-TRU" is required for proper disposition. In addition, all material classified as TRU-contaminated must meet certain criteria for eventual permanent geological disposal at the Waste Isolation Pilot Plant (WIPP). The pertinent Waste Acceptance Criteria (WAC) include quantification of any spontaneous neutron emitters and fissile material present and identification of the radionuclides contained in the waste package. The task of classifying the ORNL waste is further complicated by the score of different TRU isotopes contained in the waste. Six independent TRU-waste-generating sources (see Section 4, Table 2) have contributed to the large array of isotopes. The TRU isotopes present in significant quantities include ^{233}U, ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{243}Am, and ^{246}Cm.

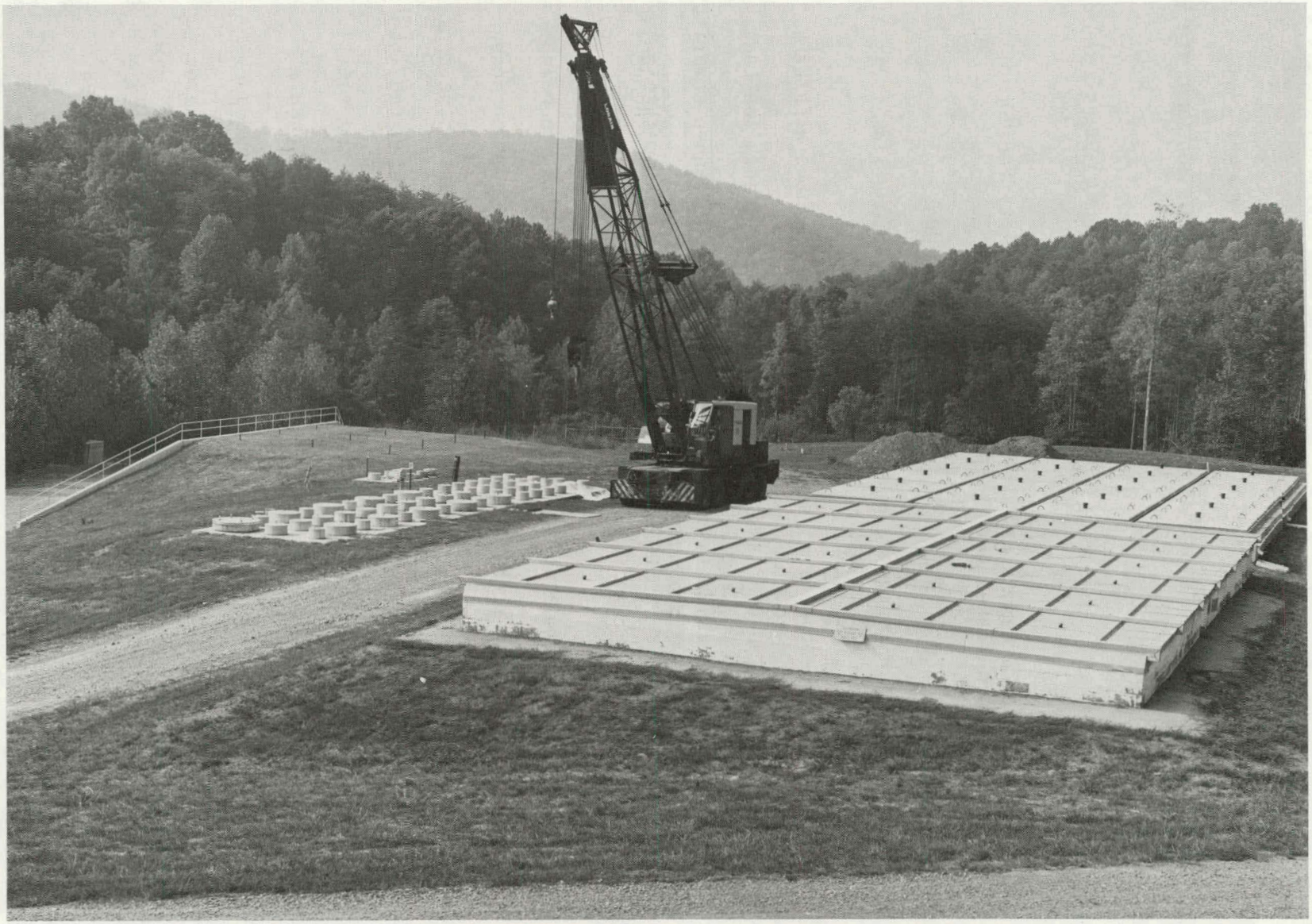

FIGURE 1. Transuranic Waste Retrievable Storage

ORNL has been selected as the demonstration site for a new transuranic assay system. The stated objectives of the cooperative program between the Los Alamos National Laboratory (LANL) and ORNL are as follows:

1. To field test, calibrate, and evaluate the LANL neutron interrogation assay system.
2. To provide a demonstration and training facility for DOE of this sophisticated technology.
3. To reduce the volume of TRU waste stored at ORNL.
4. To provide positive identification of the radionuclide content of the ORNL TRU waste.

In order to meet the objectives stated above, as well as the WIPP/WACs outlined earlier, a two-tier assay system has been employed. The LANL neutron interrogation assay system and a gamma-ray drum scanner comprise the nondestructive assay system (see Description of Equipment section). The results obtained from the two nondestructive assay (NDA) techniques must be verified independently to assure that reliable data are being obtained. A glove box facility has been constructed for this purpose. Selected TRU waste drums will be destructively assayed whereby the contents of each drum will be emptied into a glove box and segregated into four categories: cellulosics, plastics, glass and ceramics, and metals. Each type of material, excluding metals, will be weighed and then homogenized. A specified number of samples from each waste category will be collected and transferred out of the glove box. The samples will be assayed by a gamma-ray spectrometer with the results being stored on magnetic disks for later comparison to the NDA techniques.

This report describes the sampling plan employed in determining the kind and number of TRU waste drums selected for destructive assay as well as the number of samples to be collected from each waste category. The basis and details of the sampling plan are given in Sections 3, 4, and 5. Conclusions relevant to the proposed sampling plan are discussed in Section 6.

2. DESCRIPTION OF EQUIPMENT

2a. Neutron Interrogation Assay System (NIAS)

The transuranic neutron interrogation assay system (NIAS) developed at Los Alamos National Laboratory is a high-sensitivity (1-mg fissile assay sensitivity in a 208-ℓ drum) detection system capable of performing rapid, quantitative assays of low-fissile-content wastes and scraps contained in high- or low-density matrices (Kunz et. al., 1981).

The assay chamber (rectangular parallelepiped with a square cross section 67 cm on a side and 102 cm high) of the neutron system consists of a specially designed graphite (10.8 cm thick) and polyethylene (23 cm thick) structure (see Figure 2) built to accomodate packages as large as 55-gal (208-ℓ) drums. Interrogating neutrons are provided by a small, pulsed deuterium and tritium neutron generator (developed by Sandia National Laboratory and General Electric Corporation, Neutron Devices Division) located in the cavity (Rice, 1980). The pulses of 14-MeV neutrons (each pulse produces about 10^6 neutrons) are rapidly moderated and deflected back into the cavity by the surrounding graphite and polyethylene. The resultant interrogating neutrons induce fission reactions in fissile material within the sample. The ensuing prompt

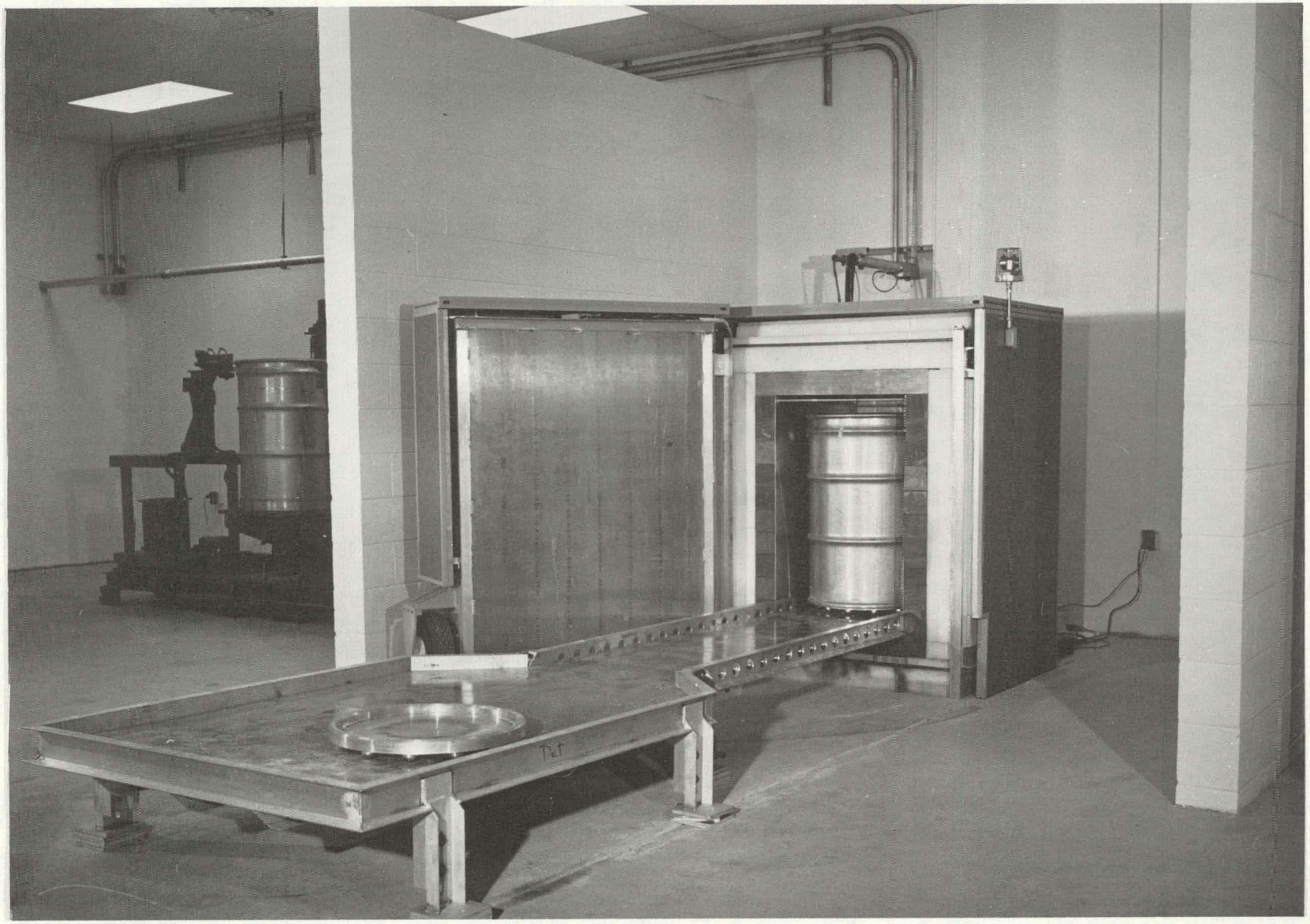

FIGURE 2. Neutron Interrogation Assay System

fission neutrons are detected by cadmium-shielded ^{3}He proportional counters embedded in the polyethylene (efficiency - 3.5%). To monitor the interrogating thermal neutron flux, a low-efficiency, bare ^{3}He proportional counter is positioned inside the assay chamber. The ratio of the prompt-fission neutron detector counts to the interrogating flux monitor counts is proportional to the amount of fissile material present.

The NIAS is also a passive neutron detection system (Kunz and Caldwell, 1982) where neutrons generated by spontaneous fissionable material are detected by both bare and cadmium-shielded ^{3}He proportional counters located within the chamber walls (total system detector efficiency - 14%). Total coincidence neutron detection and neutron multiplicity measurements can also be performed, if needed, for a more complete characterization of the sample package. The data acquisition subsystem is a LeCroy 3500 computer which collects, processes, and then stores the data on magnetic diskettes (see Figure 3).

2b. Gamma Drum Scanner

The gamma drum scanner used in the assay system is a modified Canberra Segmented Gamma Scanner Model 2220B (see Figure 4). It is able to assay gamma-emitting isotopes in 55-gal (208-ℓ) drums filled with low to medium density wastes such as rags, towels, rubber gloves and similar material. The drum scanner consists of a dual axis positioning system, a collimated solid state Ge(Li) detector, and a europium transmission source. The entire system is operated and controlled through the LeCroy 3500 computer.

FIGURE 3. Instrumentation Control Room

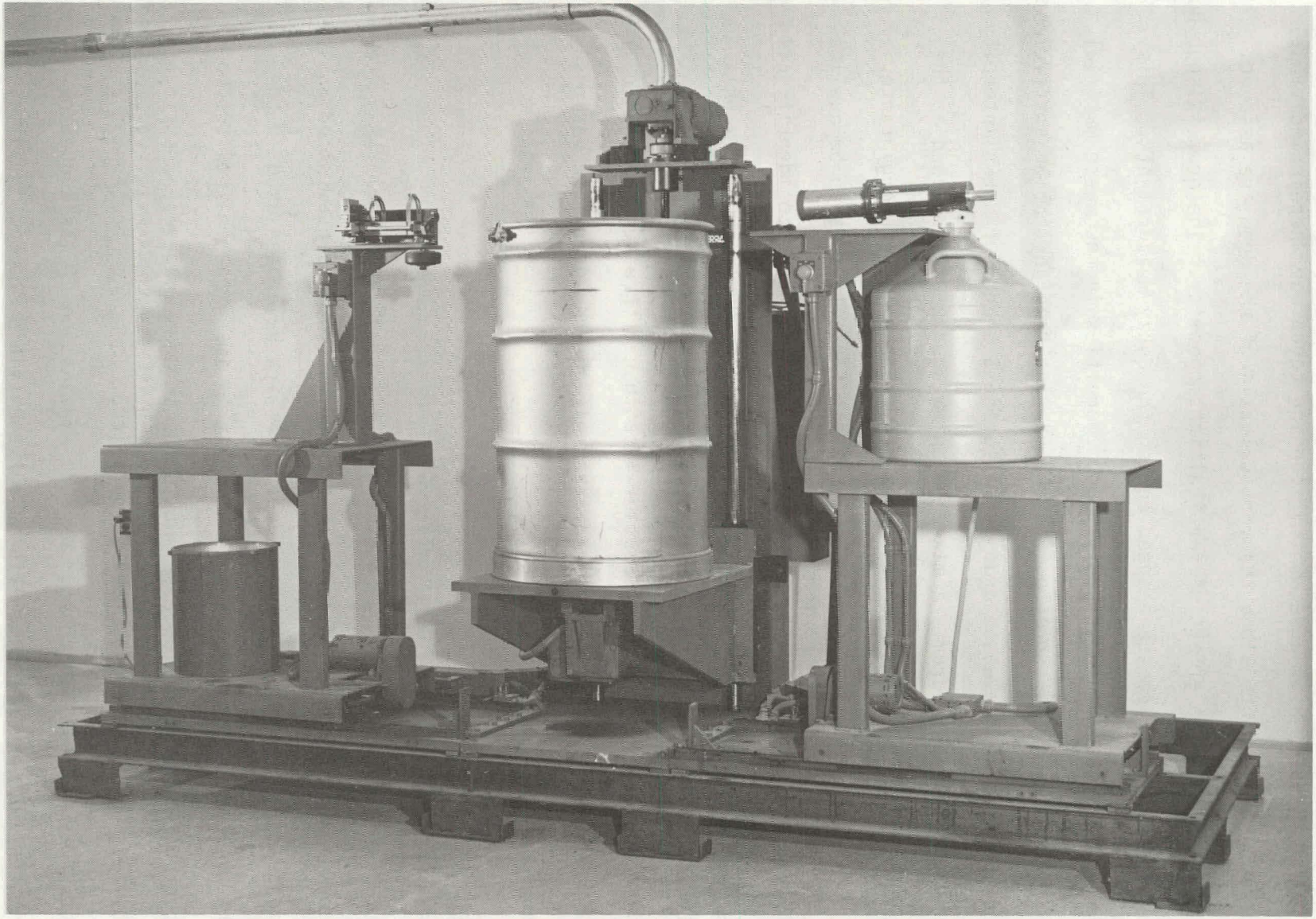

FIGURE 4. Gamma-ray Drum Scanner

The system uses a segmented scanning technique wherein individual horizontal segments along the vertical axis of the drum are assayed, thus reducing the effect of vertical inhomogeneity. The drum is spun on a turntable at a constant speed during the assay measurements. This tends to average out the radial inhomogeneities in the waste material. Each assay scan is corrected for the varying mass absorption coefficient encountered in the waste drum by measuring a source of known intensity through the waste drum. A mixed europium source will be used as the transmission source since its gamma rays span a large energy range.

The present assaying scheme provides for all drums to be qualitatively scanned by the segmented drum scanner in order to identify all gamma-emitting radioisotopes contained in each drum. However, only those drums which cannot be positively identified by the neutron interrogation system as possessing less than or greater than 100 nCi/g TRU concentrations will be quantitatively scanned using the various europium transmission sources.

3. SAMPLING PLAN

The population to be sampled consists of approximately 1800 drums containing materials (decontamination debris (DD), contaminated equipment (CE), dry solids (DS), and other) which have been used in the handling of radioactive substances. It is important to know the mean concentration of radioactive substances in each and every drum in order to determine the appropriate disposition. The concentration in a given drum may be determined by a complete examination of its contents. This method must be performed by hand but does yield accurate results. In

addition, this procedure is time consuming and costly. The new technique, discussed in the previous section, has been developed for obtaining the mean concentration of radioactive substances in each drum in a shorter period of time. However, the ability of the new technique to make adequate measurements needs to be validated. Therefore, the sampling plan proposed in this section has been suggested as a means of validating the measurements from the new technique. Every drum will be measured by the new technique (Method I) and a sample of drums will also be measured by hand inspection (Method II). For those drums which are measured by both methods, the measurements will be compared and the proportion of agreement between the matched observations will be determined and will form the basis for the validation.

3a. Selection of Drums

The population of drums comes from L different sources or sites. Let N_ℓ be the number of drums from the ℓ^{th} source for $\ell = 1, 2, \ldots, L$. (Note $\sum_{\ell=1}^{L} N_\ell = N \approx 1800.$) With the i^{th} drum from the ℓ^{th} source, we have associated a vector $(X_{\ell i}, Y_{\ell i})$ where $X_{\ell i}$ is the measurement obtained using Method I and $Y_{\ell i}$ is the measurement obtained using Method II for $i = 1, 2, \ldots, N_\ell$ and $\ell = 1, 2, \ldots, L$. If $X_{\ell i}$ and $Y_{\ell i}$ are "close", then they are said to agree, otherwise they are said to disagree. One definition of agreement might be that $X_{\ell i}$ and $Y_{\ell i}$ agree if $X_{\ell i}$ is within 25% of $Y_{\ell i}$. Let

$$\delta_{\ell i} = \delta_{\ell i}(X_{\ell i}, Y_{\ell i}) = \begin{cases} 1 \text{ if } X_{\ell i} \text{ and } Y_{\ell i} \text{ agree} \\ 0 \text{ otherwise} \end{cases}$$

Then the proportion of drums from the ℓ^{th} source, where the measurements agree, is given by

$$P_\ell = \frac{\sum_{i=1}^{N_\ell} \delta_{\ell i}}{N_\ell}$$

and the proportion of <u>all</u> the drums where the measurements agree, is given by

$$P = \sum_{i=1}^{L} \frac{N_\ell}{N} P_\ell .$$

P is an unknown population parameter. Of course, values of P close to one would suggest that both techniques are producing similar results.

To estimate P, a stratified random sample of size n will be taken. This is accomplished by randomly selecting n_ℓ of the drums from the ℓ^{th} source and determining $Y_{\ell i}$ for $i = 1, 2, \ldots, n_\ell$, $\ell = 1, 2, \ldots, L$. (Note that all drums will be measured by Method I but only n by Method II. Also note that $n = \sum_{i=1}^{L} n_\ell$.) We require that

$$n_\ell = \frac{N_\ell}{N} n.$$

Let $\hat{P}_\ell = \frac{\text{\# of agreements in sample measurements from the } \ell^{th} \text{ source}}{n_\ell}$.

Then an estimate for P is given by

$$\hat{P} = \sum_{\ell=1}^{L} \frac{N_\ell}{N} \hat{P}_\ell ,$$

and an estimate of its variance is given by

$$\hat{\text{Var}}\ (\hat{P}) = \sum_{\ell=1}^{L} \left(\frac{N_\ell}{N}\right)^2 \left(\frac{N_\ell - n_\ell}{N_\ell}\right) \frac{\hat{P}_\ell(1-\hat{P}_\ell)}{n_\ell - 1} .$$

The question remaining is how large should n be?

If the desire is to have

$$\Pr\ (|\hat{P}-P| \geq d) = \alpha ,$$

where α and d are given by the experimenter, then we should determine n using the following formula (see Cochran (1977) page 110)

$$n = \frac{\sum_{\ell=1}^{L} \frac{N_\ell}{N} P_\ell(1-P_\ell)}{\left(\frac{d}{Z_{\alpha/2}}\right)^2 + \frac{1}{N} \sum_{\ell=1}^{L} \frac{N_\ell}{N} P_\ell(1-P_\ell)}$$

where $\Pr(|Z| \geq Z_{\alpha/2}) = \alpha$ when Z is normally distributed with mean 0 and variance 1.

Since the P_ℓ's are unknown, we use $P_\ell = \frac{1}{2}$ for every ℓ. This will make n larger than necessary. Thus we have

$$n = \frac{\sum_{\ell=1}^{L} \left(\frac{N_\ell}{N}\right)\left(\frac{1}{2}\right)\left(\frac{1}{2}\right)}{\left(\frac{d}{Z_{\alpha/2}}\right)^2 + \frac{1}{N} \sum_{i=1}^{L} \left(\frac{N_\ell}{N}\right)\left(\frac{1}{2}\right)\left(\frac{1}{2}\right)}$$

$$= \frac{\frac{1}{4}}{\left(\frac{d}{Z_{\alpha/2}}\right)^2 + \frac{1}{4N}} .$$

For various values of d and α when N = 1800, we can determine the value of n. Table 1 gives a summary of n for different combinations of α and d.

After choosing n, n_ℓ is determined by $n_\ell = \frac{N_\ell}{N} n$. Since it can be assumed that drums will arrive for inspection in a random fashion, the stratified sample can be taken systematically with a random start.

TABLE 1: Sample Size (n) of the Number of Drums To Be Sampled as a Function of α and d for N = 1800.

d \ α	.01	.02	.03	.04	.05	.06	.07	.08	.09	.10
.04	656	577	523	482	451	423	399	379	361	343
.08	226	190	167	151	139	129	120	113	107	100
.12	108	90	79	71	65	60	56	52	49	46
.16	63	52	45	41	37	34	32	30	28	27
.20	41	34	29	26	24	22	21	19	18	17

4. STRATIFICATION OF SAMPLED DRUMS FROM DIFFERENT SOURCES AND WASTE TYPE BY ANTICIPATED ACTIVITY

The results from the previous section, with a choice of α = .05 and d = .08, indicated that approximately 150 drums should be sampled and the activity determined by both methods. Table 2 gives the total number of available drums and the number of drums to be sampled from each site-waste type configuration. Since 150 drums is approximately 8% of the total available population of drums, the number of drums to be sampled in each cell of Table 2 equals ≈8% of the number of available drums in that cell. However, if 8% of the number of available drums for a site-waste type configuration was less than 10, then the number of sampled drums was modified to equal the maximum of the total number of drums for

the site-waste type configuration or 10. This is the reason the total number of sampled drums (199) is ≈10% of the total number of available drums rather than ≈8%. The sampled drums from each cell of Table 2 will form a simple random sample, except for the drums containing dry solids (DS) exclusive of the PGDP drums. Because of the large number of DS drums to be sampled, it was decided to use a stratified random sample of DS drums at the sites of interest in order to be sure to include a wide range of drum activity values in the sample. From the data available at present to estimate the activity of each drum, the "cumulative square root of the frequency method" (Cochran (1977) pp. 127-131) was used to determine the strata limits. The number sampled in each strata was slightly skewed toward the activity value of 100 nCi/g assuming each drum weighed approximately 45 kg (100 lbs). Table 3 gives the activity strata limits for each site to be used in selecting the sampled drums.

TABLE 2: Total Number of Drums and Number of Drums To Be Sampled

Source / Type	ORNL 7920	ORNL 3019	ORNL Isotope Area	ORNL Other	PGDP*	Other Off-site	Total
DD	1 1	33 10	22 10	12 10	0 0	0 0	68 31
CE	1 1	2 2	6 6	38 10	0 0	0 0	47 19
DS	352 29	422 34	324 26	494 40	25 10	101 10	1718 149
Total	354 31	457 46	352 42	544 60	25 10	101 10	1833 199

*The 25 PGDP drums were considered separately.

TABLE 3: Strata Activity Limits for DS Drums By Site

(A) Building 7920

Strata Limits (curies/drum)	Number to be sampled
$1x10^{-5}$ - $3x10^{-4}$	7
$3x10^{-4}$ - $1x10^{-3}$	8
$1x10^{-3}$ - $3x10^{-3}$	7
$3x10^{-3}$ - $2x10^{0}$	7
	29

(B) Building 3019

Strata Limits (curies/drum)	Number to be sampled
$8x10^{-6}$ - $1x10^{-2}$	9
$1x10^{-2}$ - $3x10^{-2}$	9
$3x10^{-2}$ - $1x10^{-1}$	8
$1x10^{-1}$ - $6x10^{0}$	8
	34

(C) Isotopes Area

Strata Limits (curies/drum)	Number to be sampled
$2x10^{-6}$ - $1x10^{-3}$	7
$1x10^{-3}$ - $1x10^{-1}$	7
$1x10^{-1}$ - $1x10^{1}$	6
$1x10^{1}$ - $2x10^{3}$	6
	26

(D) Other ORNL Sites

Strata Limits (curies/drum)	Number to be sampled
$1x10^{-8}$ - $1x10^{-5}$	9
$1x10^{-5}$ - $1x10^{-2}$	12
$1x10^{-2}$ - $1x10^{-1}$	10
$1x10^{-1}$ - $1x10^{4}$	9
	40

(E) Other Off Site

Strata Limits (curies/drum)	Number to be sampled
$1x10^{-6}$ - $1x10^{-4}$	2
$1x10^{-4}$ - $1x10^{-3}$	3
$1x10^{-3}$ - $1x10^{-1}$	3
$1x10^{-1}$ - $1x10^{4}$	2
	10

5. SELECTION OF SAMPLES WITHIN A DRUM

In this section we will describe the method used to determine the number of samples taken within each sampled drum. We have assumed in this section that the contents of each sampled drum have been separated into C distinct components (metals, plastics, cellulosics, glass, and ceramics) from which subsamples may be taken. Set

π_c = proportion, by weight, of the material in the drum of component c (=1, 2,..., C);

μ_{cyi} = mean of the distribution of concentration in the i^{th} (=1, 2,..., I) isotope in the c^{th} component as determined by the γ-ray spectroscopy method (Method II);

σ^2_{cyi} = variance of the distribution of concentration in the i^{th} isotope in the c^{th} component as determined by the Method II analysis;

m_c = number of samples or observations taken from the c^{th} component;

y_{cik} = observed concentration in the c^{th} component of the k^{th} sample ($k=1,\ldots, m_c$) on the i^{th} isotope obtained by Method II;

$y_{ci.} = \frac{1}{m_c} \sum_{k=1}^{m_c} y_{cik}$ = estimate of μ_{cyi} from the m_c observations;

$y_{.i} = \sum_c \pi_c y_{ci.}$;

$y_{c.} = \sum_{i=1}^{I} y_{ci.}$;

σ^2_{cy} = Var $(\sum_{i=1}^{I} y_{cik})$, which is independent of k;

$\mathrm{Var}(y_{c.}) = \sigma^2_{cy}/m_c$ since $y_{c.}$ is a sum of averages involving m_c observations;

μ_{xi} = mean of the distribution of concentrations of the i^{th} isotope as determined by the new transuranic assay system (Method I);

X_i = estimate of μ_{xi} from Method I analysis; and

$X_\cdot = \sum_{i=1}^{I} X_i$ which may be estimated without knowledge of the individual X_i's by Method I.

The data from any sampled drum which comes from a particular source, can be summarized in the form given in Table 4. The subscripts of Section 3 associated with the source and drum have been suppressed in this table. The quantities of interest for comparison are

$$\mu_y = \sum_{c=1}^{C} \pi_c \sum_{i=1}^{I} \mu_{cyi},$$

estimated by

$$y = \sum_{c=1}^{C} \pi_c y_{c\cdot},$$

and

$$\mu_x = \sum_{i=1}^{I} \mu_{xi},$$

estimated by

$$X_\cdot = \sum_{i=1}^{I} X_i.$$

Additional comparisons for each isotope may be made, depending upon the availability of X_i values.

In order to determine m_c, assume the $y_{c\cdot}$ are independent and consider

$$\mathrm{Var}(y) = \sum_{c=1}^{C} \pi_c^2 \frac{\sigma_{cy}^2}{m_c}.$$

(Even if independence is not assumed, expressions for Var(y), which will still go to zero as $m_c \to \infty$, can be derived.)

TABLE 4. DATA FROM A SINGLE DRUM

Isotope	γ-Ray Spectroscopy						New Transuranic Assay System
	Component						
	Metals	Plastics	Cellulosics	Glass	Ceramics	Weighted Average	
1	y_{111}	y_{211}	y_{311}	y_{411}	y_{511}		
	•	•	•	•	•		
	•	•	•	•	•		
	•	•	•	•	•		
	y_{11m_1}	y_{21m_2}	y_{31m_3}	y_{41m_4}	y_{51m_5}		
Average	$y_{11\cdot}$	$y_{21\cdot}$	$y_{31\cdot}$	$y_{41\cdot}$	$y_{51\cdot}$	$y_{\cdot 1}$	X_1
2	y_{121}	y_{221}	y_{321}	y_{421}	y_{521}		
	•	•	•	•	•		
	•	•	•	•	•		
	•	•	•	•	•		
	y_{12m_1}	y_{22m_2}	y_{32m_3}	y_{42m_4}	y_{52m_5}		
Average	$y_{12\cdot}$	$y_{22\cdot}$	$y_{32\cdot}$	$y_{42\cdot}$	$y_{52\cdot}$	$y_{\cdot 2}$	X_2
•	•	•	•	•	•	•	•
•	•	•	•	•	•	•	•
•	•	•	•	•	•	•	•
I	y_{1I1}	y_{2I1}	y_{3I1}	y_{4I1}	y_{5I1}		
	•	•	•	•	•		
	•	•	•	•	•		
	•	•	•	•	•		
	y_{1Im_1}	y_{2Im_2}	y_{3Im_3}	y_{4Im_4}	y_{5Im_5}		
Average	$y_{1I\cdot}$	$y_{2I\cdot}$	$y_{3I\cdot}$	$y_{4I\cdot}$	$y_{5I\cdot}$	$y_{\cdot I}$	X_I
Total	$y_{1\cdot}$	$y_{2\cdot}$	$y_{3\cdot}$	$y_{4\cdot}$	$y_{5\cdot}$	y	$X_\cdot$
Component Proportion	π_1	π_2	π_3	π_4	π_5		

If we also assume that $m_c = \pi_c m$ where $m = \sum_{c=1}^{C} m_c$ then

$$\text{Var}(y) = \frac{1}{m} \sum_{c=1}^{C} \pi_c \sigma_{cy}^2 .$$

The $100(1-\alpha)\%$ confidence interval on μ_y now takes the form

$$y \pm Z_{\alpha/2} \left[\frac{1}{m} \sum_{c=1}^{C} \pi_c \sigma_{cy}^2\right]^{1/2} .$$

The determination of m can now be expressed in terms of the following:

Make m sufficiently large so that the width of the confidence interval is sufficiently small. The width of the confidence interval is

$$2Z_{\alpha/2}\left[\frac{1}{m} \sum_{c} \pi_c \sigma_{cy}^2\right]^{1/2} = 2\Delta .$$

Solving this expression for m we have

$$m = \frac{Z_{\alpha/2}^2}{\Delta^2} \left[\sum_{c} \pi_c \sigma_{cy}^2\right] .$$

The value of Δ would be determined from a knowledge of how much of a difference between μ_y and μ_x we want to be declared significant, the values of π_c should be easily determined, and the values of σ_{cy}^2 could be estimated from a pilot sample to give us a handle on a minimum value of m. The allocation of m would be done as $m_c = \pi_c m$. If we assume $\sigma_{1y}^2 = \ldots = \sigma_{cy}^2 = \sigma_y^2$ then m becomes

$$m^* = \frac{Z_{\alpha/2}^2 \ \sigma_y^2}{\Delta^2} .$$

Table 5 gives the value of m^* for different values of σ_y/Δ.

TABLE 5: Minimal Sample Sizes As a Function of α and σ_y/Δ When Variances Are Equal For Each Component

	α			
σ_y/Δ	0.01	0.05	0.10	0.20
.2	1	1	1	1
.4	2	1	1	1
.6	3	2	1	1
.8	5	3	2	2
1.0	7	4	3	2
1.2	10	6	4	3
1.4	13	8	6	4
1.6	17	10	7	5
1.8	22	13	9	6
2.0	27	16	11	7

6. CONCLUSIONS

The sampling plan described in the previous sections of this report can be used to verify the results from the nondestructive assay technique. The plan provides a method to determine the number of TRU waste drums for destructive assay as well as the number of samples to be collected from each waste category within a selected drum. The observations from the destructive and nondestructive assay can then be used to determine if the new detector system provides accurate estimates of the mean concentration of the TRU waste present in a drum. As additional observations become available it will be possible to evaluate (and modify, if necessary) the sample sizes given in Tables 1 and 5 to achieve the desired precision in the estimated concentration differences for the two assay methods.

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