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**A REVIEW OF METHODS FOR THE DETECTION
OF 10 nCi/g OF TRANSURANIC ISOTOPES
IN SOLID WASTE**

W. C. King

January 7, 1977

Prepared for U.S. Energy Research & Development
Administration under contract No. W-7405-Eng-48

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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, VA 22161

Price: Printed Copy \$: Microfiche \$3.00

Page Range	Domestic Price	Page Range	Domestic Price
001-025	\$ 3.50	326-350	10.00
026-050	4.00	351-375	10.50
051-075	4.50	376-400	10.75
076-100	5.00	401-425	11.00
101-125	5.50	426-450	11.75
126-150	6.00	451-475	12.00
151-175	6.75	476-500	12.50
176-200	7.50	501-525	12.75
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276-300	9.25	601-up	*
301-325	9.75		

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A REVIEW OF METHODS FOR THE DETECTION OF 10 nCi/g OF TRANSURANIC ISOTOPES IN SOLID WASTE

Abstract

The U.S. Atomic Energy Commission has defined transuranic-contaminated waste as material that is contaminated with greater than 10 nCi/g of long-lived and highly toxic radionuclides. In this report we review the analyses and techniques that show potential for detection at this low level. The physics involved with the methods described is relatively straightforward, but difficulties arise in the actual application of the techniques. Therefore, we concentrate our discussion on the application of the analyses, including the necessary calculational considerations and the potential problems that may be encountered.

We conclude that no simple method presently exists for measuring transuranic isotopes in solid waste material at the low level of 10 nCi/g. In the absence of an interfering background (< 1 mR/hr), gamma-ray spectroscopy is the best method available. Monitoring helium production in sealed waste material shows good potential for detection at the 10-nCi/g level. The only other viable method involves a complex procedure of counting spontaneous-fission neutrons, neutron irradiation, and counting delayed neutrons. However, if the minimum detectable level permitted in waste material were raised to 1 μ Ci/g, we would be able to use several more measurement methods and detect almost all the transuranic isotopes present in solid waste.

Introduction

In 1973, the Atomic Energy Commission (AEC)* issued Manual Chapter 0511 on Radioactive Waste Management. Transuranic-contaminated solid waste is defined as "material contaminated with certain alpha-emitting radionuclides of long half-life and high specific radiotoxicity to greater than 10 nanocuries/gram" (see Part 1 of Appendix 0511, Ref. 1). The radionuclides in this category were further defined as ^{233}U (with its daughter products), plutonium, and

* In early 1975, the AEC was reorganized. The Energy Research and Development Administration (ERDA) was created and took over all the functions of the old AEC with exception of the Regulatory Function. All Manual Chapter requirements were recognized by ERDA and are currently in effect.

transplutonium nuclides excepting ^{238}Pu and ^{241}Pu . The packaging and storing conditions of such waste required that the packages be readily retrievable in an intact, contamination-free condition for 20 yr.

The AEC recognized that, for typical ^{239}Pu -contaminated waste, 10 nCi/g probably could not be measured by direct external means and that indirect measurements or estimates and administrative control would have to be applied to satisfy the requirements of Manual Chapter 0511. In addition, the AEC allowed the activity density to be averaged over the contents of individual shipping containers (such as 55-gal drums) including materials added for shielding or sorption of liquids.

Several ERDA contractors^{2,3} have developed instrumentation in attempts to measure the 10-nCi/g level for ^{239}Pu . Generally, gamma-ray spectrometers are used to measure either the 375-keV gamma from ^{239}Pu or the 60-keV gamma from ^{241}Am , a daughter of the ^{241}Pu isotope which is always found with ^{239}Pu . In the absence of a gamma background, gamma spectroscopy works reasonably well with light waste material; however, it is not usable in a gamma background of a few milliroentgens per hour.

Large amounts of transuranic (TRU) wastes will be generated in the course of the power reactor fuel recycle. These wastes will include such items as contaminated scrap metal, glassware, and discarded process equipment, as well as laboratory wastes such as resins, filters, clothing, and paper products. Much of these wastes will be associated with residual activation and fission products that contribute a significant gamma background level. As a result, detection of 10 nCi/g of the TRU with the more conventional gamma-ray analyses is impossible. Thus the question arises: Are there other methods that can be used to measure 10 nCi/g of TRU in waste in the presence of a gamma background up to 1 r/hr? In this report, we review a number of possible methods, evaluating the merits and drawbacks of each.

The largest single source of TRU-contaminated waste will be generated by the chemical processing of recycled fuel and the conversion of plutonium nitrate to plutonium oxide. Over 99% of the TRU inventory will be recovered for reuse or will end up with the high-level liquid wastes. Table 1 lists the spent-fuel inventory of TRU activity from LWPR (light water power reactor) fuel burn-up of 35,000 MWd/MTU (tonne of uranium) at a specific power level of 40 kW/kg uranium.⁴ The fuel elements are assumed to be cooled for 160 d before processing and TRU isotopes with activity less than 1 Ci/MTU are not shown.

Table 1. Inventory of TRU activity from spent LWPR fuel.

Transuranic Isotope	Half-life (yr)	Activity (Ci/MTU)	Relative Activity
^{238}Pu	8.64×10^1	3.48×10^3	1.7×10^{-2}
^{239}Pu	2.44×10^4	3.36×10^2	1.6×10^{-3}
^{240}Pu	6.58×10^3	6.30×10^2	3.1×10^{-3}
^{241}Pu	1.32×10^1	1.59×10^5	7.7×10^{-1}
^{242}Pu	3.79×10^3	3.00×10^0	1.5×10^{-5}
^{241}Am	4.58×10^2	2.69×10^2	1.3×10^{-3}
^{242}Am	1.52×10^2	5.00×10^0	2.4×10^{-5}
^{242}Cm	4.46×10^{-1}	3.94×10^4	1.9×10^{-1}
^{243}Cm	3.20×10^1	2.40×10^1	1.1×10^{-4}
^{244}Cm	1.76×10^1	3.32×10^3	1.6×10^{-2}
Total Activity		2.06×10^5	

Other sources of TRU-contaminated wastes include fuel fabrication plants, nuclear research facilities, nuclear medical facilities, and source production plants. Table 2 lists all the alpha-emitting TRU isotopes to which the 10 nCi/g criterion applies. Some physics data are also given in Table 2. Isotopes with half-lives less than 100 d are not considered to be a waste management problem except for the fact that many of the short-lived TRUs with $Z > 95$ do decay into long-lived TRU daughters. Such ingrowth must be taken into account.

Description of Analytical Methods

Nondestructive analytical methods for TRU isotopes in solid waste fall into two categories, passive and active analyses. Passive methods use the natural radiation emissions to identify and quantify the isotopes that are present. In active analyses, external radiation is used to induce a measured response or change in the TRU emissions. The physics involved with all the methods is fairly straightforward. The difficulties arise in their application.

Table 2. Alpha-emitting TRU isotopes with $T_{1/2} > 100$ d.

Isotope	$T_{1/2}$ (yr)	$T(SF)_{1/2}$ (yr)	Thermal Neutron Cross Sections (barn)	Specific Activity ($\mu\text{Ci/g}$)	(SF) Activity (fission/ sec-g)	λ (SF)	Parent	Daughter
$^{231}_{92}\text{U}$	1.62×10^5		$\sigma_c = 49$ $\sigma_f = 524$	1.05×10^2	0		^{231}Pa ^{231}Np	^{229}Th
$^{237}_{93}\text{Np}$	2.14×10^6	10^{18}	$\sigma_c = 170$ $\sigma_f = 0.019$	1.42×10^2	5.6×10^{-5}		^{237}U ^{241}Am ^{242}Pu	^{233}Pa
$^{236}_{94}\text{Pu}$	2.85×10^0	3.5×10^9	$\sigma_c = 170$	1.88×10^{-3}	1.6×10^4		^{240}Cm ^{236}Np	^{232}U
$^{238}_{94}\text{Pu}$	8.64×10^1	4.9×10^{10}	$\sigma_c = 500$ $\sigma_f = 16.8$	5.75×10^{-2}	1.13×10^3	2.26	^{238}Np ^{238}Am ^{242}Cm	^{234}U
$^{239}_{94}\text{Pu}$	2.44×10^4	5.5×10^{15}	$\sigma_c = 274$ $\sigma_f = 741$	1.63×10^1	1.00×10^{-2}	2.2	^{239}Np ^{243}Cm ^{239}Am	^{235}U
$^{240}_{94}\text{Pu}$	6.58×10^3	1.34×10^{11}	$\sigma_c = 286$ $\sigma_f = 0.08$	4.4×10^0	4.1×10^{-2}	2.17	^{244}Cm ^{240}Np ^{240}Am	^{236}U
$^{242}_{94}\text{Pu}$	3.79×10^5	7.1×10^{10}	$\sigma_c = 19$ $\sigma_f = 0.2$	2.57×10^2	7.7×10^2	2.16	^{246}Cm ^{242}Am	^{238}U
$^{244}_{94}\text{Pu}$	7.6×10^7	2.5×10^{10}	$\sigma_c = 1.8$	5.2×10^{-4}	2.7×10^3		^{248}Cm	^{240}U F.P.
$^{241}_{95}\text{Am}$	4.58×10^2	2×10^{14}	$\sigma_c = 700$ ^{242}Am $\sigma_c = 100$ ^{242m}Am $\sigma_f = 3.0$	3.08×10^{-3}	2.7×10^{-1}	2.3	^{241}Pu ^{245}Bk ^{241}Cm	^{237}Np
$^{242m}_{95}\text{Am}$	1.52×10^{-2}		$\sigma_c = 2000$ $\sigma_f = 6000$	1.03×10^{-1}			a	^{242}Am ^{233}Np
$^{243}_{95}\text{Am}$	7.95×10^3		$\sigma_c = 74$ $\sigma_f = 0.07$	5.40×10^0			^{243}Pu ^{247}Bk	^{239}Np
$^{242}_{96}\text{Cm}$	4.45×10^{-1}	7.2×10^6	$\sigma_c = 20$ $\sigma_f = 5$	3.01×10^{-1}	7.58×10^6		^{242}Am ^{246}Cf	^{238}Pu F.P.
$^{243}_{96}\text{Cm}$	3.2×10^1		$\sigma_c = 250$ $\sigma_f = 662$	2.2×10^{-2}			^{243}Bk	^{239}Pu

Table 2. (Continued).

Isotope	$T_{1/2}$ (yr)	$T(SF)_{1/2}$ (yr)	Thermal Neutron Cross Sections (barn)	Specific Activity (g/Ci)	(SF) Activity (fission/ sec-g)	ν (SF)	Parent	Daughter
$^{244}_{96}\text{Cm}$	$1.76 \cdot 10^1$	$1.31 \cdot 10^7$	$\sigma_c = 15$	$1.2 \cdot 10^{-2}$	$4.14 \cdot 10^6$		^{248}Cf ^{244}Am ^{244}Bk	^{240}Pu F.P.
$^{245}_{96}\text{Cm}$	$9.3 \cdot 10^1$		$\sigma_c = 200$ $\sigma_f = 1900$	$6.4 \cdot 10^0$			^{249}Cf ^{245}Am ^{245}Bk	^{241}Pu
$^{246}_{96}\text{Cm}$	$5.5 \cdot 10^1$	$1.7 \cdot 10^7$	$\sigma_c = 15$	$3.8 \cdot 10^0$	$3.2 \cdot 10^6$		^{250}Cf ^{246}Am ^{246}Bk	^{242}Pu F.P.
$^{247}_{96}\text{Cm}$	$1.6 \cdot 10^2$		$\sigma_c = 180$	$1.1 \cdot 10^4$			^{247}Cm	^{243}Pu
$^{248}_{96}\text{Cm}$	$4.7 \cdot 10^5$	$4.6 \cdot 10^6$	$\sigma_c = 6$	$3.27 \cdot 10^2$	$1.16 \cdot 10^7$		^{252}Cf ^{248}Bk	^{244}Pu F.P.
$^{250}_{96}\text{Cm}$		$1.7 \cdot 10^4$			$3.1 \cdot 10^9$		^{245}Cf	F.P.
$^{247}_{97}\text{Bk}$	$1.4 \cdot 10^3$			$9.66 \cdot 10^{-1}$			^{247}Cf	^{243}Am
$^{248}_{98}\text{Cf}$	$9.59 \cdot 10^{-1}$	$1.5 \cdot 10^4$		$6.64 \cdot 10^{-4}$	$3.6 \cdot 10^9$		^{252}Fm ^{248}Bk	^{244}Cm F.P.
$^{249}_{98}\text{Cf}$	$3.50 \cdot 10^2$	$1.5 \cdot 10^9$	$\sigma_c = 270$ $\sigma_f = 1735$	$2.51 \cdot 10^{-1}$	$2.5 \cdot 10^4$		^{249}Bk ^{249}Es	^{245}Cm F.P.
$^{250}_{98}\text{Cf}$	$1.32 \cdot 10^1$	$1.7 \cdot 10^4$	$\sigma_c = 1500$ $\sigma_f = 350$	$9.24 \cdot 10^{-3}$	$3.1 \cdot 10^9$		^{254}Fm ^{250}Bk ^{250}Es	^{246}Cm F.P.
$^{251}_{98}\text{Cf}$	$8 \cdot 10^2$		$\sigma_c = 3000$ $\sigma_f = 3000$	$6 \cdot 10^{-1}$			^{255}Fm ^{251}Es	^{247}Cm
$^{252}_{98}\text{Cf}$	$2.65 \cdot 10^0$	$8.5 \cdot 10^1$	$\sigma_c = 30$	$1.86 \cdot 10^{-3}$	$6.2 \cdot 10^{11}$	3.8	^{256}Fm	^{248}Cm F.P.
$^{252}_{99}\text{Es}$	$3.84 \cdot 10^{-1}$			$2.7 \cdot 10^{-4}$			^{256}Md	^{248}Bk
$^{254}_{99}\text{Es}$	$7.56 \cdot 10^{-1}$	$7 \cdot 10^5$	$\sigma_c = 40$	$5.4 \cdot 10^{-4}$	$7.4 \cdot 10^7$		— ^a	^{250}Bk F.P.

^aHas no parent isotope but is produced by a (n, γ) reaction.

Sophisticated instrumentation is required to measure the events and skilled personnel are needed to operate and maintain the equipment as well as to analyze the resulting data. Therefore, we concentrate our discussion on the application of these various methods to actual analysis of the TRU isotopes in waste material. The advantages and problems involved with each technique are described in a separate section.

PASSIVE ANALYSES

Neutron Emission from Spontaneous Fission

Two groups of neutrons are considered, those that are emitted at the time of fission (prompt or fission neutrons) and those that are emitted at some time after fission (delayed neutrons). On an average, more than two prompt neutrons are emitted per fission. These prompt neutrons cover a broad energy spectrum, ranging from less than 0.05 MeV to greater than 15 MeV. The average prompt-neutron energy is 1 to 2 MeV. Delayed neutrons, on the other hand, are emitted from several of the fission product isotopes and are emitted individually as single particles. Average energies of these delayed neutrons are generally less than 0.5 MeV. Because more than two prompt neutrons are emitted simultaneously during fission, coincidence counting may be used to discriminate against the neutron background that is caused by the delayed neutrons and the (α ,n) reactions.

Prompt Gamma-Ray Emission from Spontaneous Fission

About three times more prompt gammas than prompt neutrons are emitted during fission. These gamma rays are emitted simultaneously and have energies in the range of 0.3 to 10.5 MeV. Counting the prompt gammas in coincidence with the prompt neutrons is a possible technique for measuring the quantity of TRUs present in the solid waste.

Neutron Emission from (α ,n) Reactions

Many light elements yield neutrons when bombarded with energetic alpha particles. The emitted neutron energy depends on the energy of the bombarding alpha particle, the binding energy of the neutron, and the energy excitation of the residual nucleus. A rather broad continuous energy spectrum results with a maximum energy from 1 MeV for lithium to 10 MeV for beryllium. The neutron yield depends on the energies of the bombarding alpha-particles and on the composition of the target nucleus. For example, the alpha particles

of ^{238}Pu bombarding the oxygen atoms in $^{238}\text{PuO}_2$ will yield about 1.4×10^4 n/sec-g whereas $^{239}\text{PuO}_2$ will yield only 45 n/sec-g. Thus, a measurement of the neutron yield from the (α, n) reaction can perhaps be used to measure TRU isotopes in waste.

Production of Helium from α Decay

Alpha particles are helium nuclei. Measuring the production of helium from alpha decay in the TRU-laden waste may provide a method of determining the amount of TRUs in a container.

Production of Krypton and Xenon by Spontaneous Fission

Many isotopes of krypton and xenon are TRU fission products and most are radioactive. Thus, measuring the production of these gases through the spontaneous fission of the TRUs may be used as a method for detecting the TRU isotopes in a waste container.

Gamma and X-Ray Emission

Almost all of the TRU isotopes emit gamma rays and fluorescent x rays during decay. These characteristic emissions can also be used to identify and quantify the TRUs in solid waste.

ACTIVE ANALYSES

Neutron Emission from Induced Fission

For most of the TRU isotopes with odd mass numbers, the spontaneous-fission rate is so low that the fission neutrons, particularly in quantities as small as 10 nCi/g, cannot be detected above the (α, n) background. Introducing neutrons or high-energy photons from an external source increases the fission rate. If a sufficient neutron or photon flux can be applied to the internal volume of the waste, it is possible to detect the induced fission neutrons. Counting the fission neutrons individually and in coincidence pairs yields the quantity of TRU isotopes present.

Also, with increased fissioning, the yield of delayed neutrons is increased. Thus, the quantity of TRU isotopes present perhaps can be determined by counting the delayed neutrons following a specified irradiation time.

If neutrons are used as the irradiation source, the neutron energy should be in the thermal or epithermal range for greatest sensitivity. On the other hand, if photons are used as an irradiation source, the energy of the photons must be greater than 8 MeV to induce fission in the TRU isotopes.

Prompt Gamma-Ray Emission from Induced Fission

The techniques used to count the prompt gammas from induced fission are the same as those used to measure spontaneous-fission gamma rays.

Production of Gaseous Krypton and Xenon by Induced Fission

Increasing the fission rate by irradiation with neutrons or with high-energy photons results in an increased production of the fission-product gases. Because the fission yields differ for the isotopes of krypton or xenon for different TRU isotopes, identification of the fissioning TRU nuclei may be possible by determining the ratios of the fission-gas isotopes.

Neutron Activation

It may be possible to transmute the TRU isotopes that are difficult to detect into isotopes that are easily detectable. Transmutation is accomplished by irradiating the atoms with thermal neutrons.

Evaluation of Methods

In our evaluation of each nondestructive analytical method, we include a description of the required instrumentation and the minimum signal that might be detectable above the noise or background level. Also, a mathematical discussion for determining the amount of TRU that can be detected at the minimum signal level is provided. Because there are so many TRU isotopes that may be present in the waste (see Table 2), only one or two are discussed to illustrate each approach. We also consider problems that may be encountered in the actual application of the methods.

Gamma-ray analysis is the only method that has successfully detected such small activities and has been limited to plutonium analysis in the absence of a gamma background.² All the others show varying degrees of potential but have yet to be practically applied to the detection of TRUs at the 10-nCi/g level.

DETECTION BY NEUTRON ANALYSIS

Neutron measurement is possible either with gas-filled proportional counters or with liquid or plastic scintillators, depending on the desired sensitivity. Other methods such as film emulsions or thermoluminescent dosimeters either involve tedious procedures or do not provide adequate sensitivity and thus are not suitable. Although the liquid or plastic

scintillation detectors are very sensitive and provide pulse heights that are proportional to the neutron energy, they are also sensitive to the gamma rays which generate pulses of similar height. Therefore, it is necessary to employ pulse-shape discriminators to minimize the effects of the gamma activity.⁵

The gas used in the proportional counters is primarily BF_3 , ^3He , or ^4He . The BF_3 gas is used for neutrons at thermal energies. The ^3He gas is used to count either thermal or fast neutrons while ^4He gas is best for counting fast neutrons. Gas pressure in the counter ranges from slightly less than one to several atmospheres. Counters filled to pressures greater than one atmosphere are more sensitive to the neutrons but are also more expensive to fabricate and maintain.

The BF_3 gas is enriched in ^{10}B to provide a high (n,α) cross section (3837 barn) for thermal neutrons with the reaction, $^{10}\text{B}(n,\alpha)^7\text{Li}$. In about 90% of the reactions, the ^7Li nucleus is excited, leaving 2.31 MeV as the kinetic energy of the recoiling ^7Li atom and α particles. The excited ^7Li nucleus returns to its ground state by emission of an 0.477-MeV gamma. In the other 10% of the reactions, the recoil energy for the ^7Li atom and α particle is 2.79 MeV.

The ^3He gas has a large (n,p) cross section (5327 barn) for thermal neutrons. The reaction of interest is $^3\text{He}(n,p)^3\text{H}$; 0.764 MeV is shared as kinetic energy in the recoil of ^3H and the proton.

Interaction of fast neutrons in ^4He and ^3He gas occurs by elastic collision. A large proportion of the collisions result in forward or 180° scatter of the incident neutrons. The recoiling helium atoms have an energy spectrum that peaks at an energy representing 180° scatter. When the counter is operated in the proportional range, it is possible to discriminate against the slower neutrons by recording only those counts in the high-energy peak. However, because of the low-collision cross section (~ 1 barn) for elastic collision, this counting method is impractical for TRU detection at the 10-nCi/g level.

We conclude then, that the most practical method for detection of neutrons in waste monitoring is with BF_3 or ^3He counters imbedded in a moderating material to slow the fast neutrons to thermal energies. A cadmium shield can be used to absorb the neutrons thermalized in the waste matrix before they enter the moderator to minimize their interference with the fast neutrons.

A large well counter that will accept a drum of waste is a possible waste monitor. Our conceptual design is shown in Fig. 1. Polyethylene or water completely surrounding the waste container positioned for monitoring are suitable as moderating materials. Here, 36 matched BF_3 counters, 2 in. in diameter and 36 in. in length, are filled to a gas pressure of 700 Torr. The counters are placed in two concentric rings around the waste container with 18 counters per ring. The counter long axis runs parallel to the long axis of the waste container. A cadmium sleeve is placed between the waste container and the moderating medium.

Fig. 1. Conceptual design of the neutron irradiator and well counter.

With this design, electronic options include coincidence counting and pulse-height discrimination to help minimize the effects of gamma activity and neutrons from the (α ,n) reactions. Overall counting efficiencies for such a well counter are estimated at 15 to 20% for total-neutron counting and at 1 to 3% for counting coincidence pairs.⁶

The counting techniques are straightforward and have been demonstrated with counting systems presently in use at two national laboratories.⁵⁻⁷

With this counting well, we then ask: What is the minimum measurable quantity of TRUs in a standard 55-gal drum of solid waste?

Spontaneous-Fission Neutrons Counted in Coincidence Pairs

If we count neutrons produced by spontaneous fission in coincidence pairs, we should be able to determine the quantity of TRU isotopes present in the waste. Therefore, let P = the number of atoms of a specific TRU, λ_{SF} = the spontaneous fission constant [$0.693/T(SF)_{1/2}$], and ν = the average number of neutrons emitted per fission. The spontaneous fission rate = $\lambda_{SF}P$ and the number of neutrons emitted per unit time = $\nu\lambda_{SF}P$.

With these values, we can develop a general equation for the neutron count rate observed for neutrons emitted by Cg Ci/g of a specific TRU isotope undergoing spontaneous fission. Thus, if N_e = the number of neutrons emitted (sec^{-1}), N_C = the number of neutrons counted (sec^{-1}), ϵ = the overall counter efficiency, $N_C = N_e \epsilon$ (when counting in coincidence pairs, ϵ takes into account that two neutrons make one count), M = the mass of waste material (kg), $T(SF)_{1/2}$ = the spontaneous fission half-life (yr), and $T(R)_{1/2}$ = the radioactive half-life (yr), then

$$N_C = \frac{3.7 \times 10^{13} \nu \epsilon \text{ Cg } M T(R)_{1/2}}{T(SF)_{1/2}} \quad (1)$$

Other processes also generate neutrons, especially the (α ,n) reaction in light elements. This (α ,n) reaction involves the transport of alpha particles through the material being tested and therefore, the emission rate of these neutrons is highly dependent on the matrix material. In any given test, we do not know the contribution of the (n, α) neutrons to the total count. Therefore, we must discriminate against these unwanted neutrons by registering only those counts that occur in coincidence pairs. Because the average neutron lifetime in a proportional detector is long (15 to 100 μsec), large

coincidence gates must be used for an efficient counting system. R. Sher of Brookhaven National Laboratory has reported on techniques of coincidence counting in their Neutron Well Coincidence Counters.⁶ Such techniques are readily applicable to waste material monitoring.

We can now apply Eq. (1) to the TRU isotopes to determine the minimum amount measurable. Several assumptions are made.

- Light laboratory wastes such as Kimwipes, pylox gloves, plastic shoe covers, and used plastic test tubes are packaged in a 55-gal drum at a packing density of 50 kg/m^3 . (In practice, the packing density will be several times greater; however, 50 kg/m^3 is used for all calculations in this report as a conservative measure.) Packing density is defined as the mass of the waste material divided by the volume of the waste container. Here, mass of the waste material $M = 10.4 \text{ kg}$ and volume of the 55-gal drum $V = 0.208 \text{ m}^3$.
- The coincidence counting efficiency is 2% for the above-described well counter.
- With a small ^{252}Cf source in the technique described by Sher,⁶ the matrix material in the waste reduces the true neutron emission by 10%.
- Count time is set for 600 sec (10 min).
- The background count rate is less than 1 count/sec (known from previous calibration counts made with $^{239}\text{PuO}_2$).
- The significant minimum neutron count rate is 10 count/sec. (In reality, however, it will probably be closer to 1 count/sec.)

Determining N_C , we find

$$N_C = 6000 \text{ count}/600 \text{ sec} (1 - 0.10) = 11 \text{ count/sec.}$$

Obtaining values of $T(R)_{1/2}$, $T(SF)_{1/2}$, and ν from Table 2 for ^{239}Pu and solving Eq. (1) for C_g , we have

$$11 = 3.7 \times 10^{13} (2.2)(0.02)C_g(10.4)(2.44 \times 10^4/5.5 \times 10^{15}),$$

$$C_g = 1.5 \times 10^{-1} \text{ Ci/g.}$$

It is obvious that this method is not appropriate for measuring ^{239}Pu at the low levels we desire.

Let us try ^{242}Cm assuming $\nu = 2.2$ and $\epsilon = 0.02$:

$$11 = 3.7 \times 10^{13} (2.2)(0.02)C_g(10.4)(4.45 \times 10^{-1}/7.2 \times 10^6),$$

$$C_g = 10^{-5} \text{ Ci/g} = 10 \text{ } \mu\text{Ci/g.}$$

If we set $C_g = 10^{-8}$ Ci/g (10 nCi/g), $N_C = 1$ count/sec (the minimum detection level), $v = 2.5$ (the maximum value excluding ^{252}Cf), and $\epsilon = 0.04$ (the maximum expected efficiency), then from Eq. (1),

$$\frac{T(R)_{1/2}}{T(SF)_{1/2}} = \frac{3 \times 10^{-5}}{M} \quad (2)$$

Equation (2) is used to identify those TRU isotopes for which this method will detect 10 nCi/g in M kilograms of waste material. If we count SF neutrons in coincidence pairs from 10.4 kg of waste, we will detect the following TRU isotopes at the 10-nCi/g level: ^{242}Pu , ^{244}Pu , ^{244}Cm , ^{246}Cm , ^{248}Cm , ^{250}Cm , ^{248}Cf , ^{250}Cf , ^{252}Cf , and possibly ^{254}Es if $M > 30$ kg. If the detection limit is raised to 1 $\mu\text{Ci/g}$, ^{240}Pu and ^{242}Cm can also be quantitatively detected. However, there are several problems that may be encountered with the neutron coincidence well counter.

- Background counts caused by (n, α) reactions and high-gamma intensities cannot be determined but will have to be estimated from calibration data.
- The effects of the waste matrix on moderating the spontaneous-fission neutrons before they exit the container will have to be determined for each container that is monitored. This is feasible by counting a small ^{252}Cf source at the bottom of the well, taking a second count with the waste container and the source, and then making a third count with only the container. Thus, each container requires a minimum of three counts.
- The TRU isotopes are not individually identified by the method. To quantify the isotopes in the waste, we must know beforehand, what isotopes are present and in what proportions. However, this generally can be determined with radiochemical analyses of the materials handled in the plant.

Prompt Neutrons from Induced Fission

The same counting arrangement is used to detect the induced-fission neutrons as is used to count the spontaneous-fission neutrons. Thermal and epithermal neutrons are introduced into the waste materials at a flux density of approximately 10^6 n/cm²-sec to induce fission in the TRU isotopes. Prompt neutrons from the induced fission are counted in coincidence pairs to reduce the background that results from the sources, the (α ,n) reactions, the

delayed neutrons, and the gamma-ray background. However, computerized neutron-diffusion calculations have revealed that the source neutrons will contribute a background count so high that it will mask the counts from prompt neutrons. Therefore, we must consider this method to be impractical and unapplicable.

Induced fission by high-energy photons does reduce source interference. Again however, this method is impractical because extremely large and expensive equipment is required to provide the necessary photon energy and flux density.

Delayed Neutrons from Induced Fission

The same equipment and method used to count the prompt neutrons is used to count the delayed neutrons. A count-rate recorder to provide a time record of the count rate as the delayed neutrons decay is the only additional piece of equipment used. Fissioning is induced in the TRU isotopes by external neutron sources for a sufficiently long time (approximately 10 min) to establish an equilibrium of the longest-lived, delayed neutron producer. The neutron sources are then removed and a count rate vs time history is made of the delayed neutrons. Six prominent groups of delayed neutrons are recognized by their half-life. Table 3 presents the properties of these groups of delayed neutrons.

Table 3. Properties of delayed neutrons.⁸

Group	Half-life ^a (sec)	Absolute Yield from Thermal Fission (n/100 fissions)	
		²³³ U	²³⁹ Pu
1	55.3	0.060 ± 0.005	0.022 ± 0.009
2	23.0	0.206 ± 0.013	0.191 ± 0.034
3	5.6	0.173 ± 0.040	0.135 ± 0.045
4	2.1	0.192 ± 0.024	0.209 ± 0.033
5	0.62	0.036 ± 0.024	0.055 ± 0.027
6	0.26	0.023 ± 0.013	0.028 ± 0.015

^aHalf-life given is for ²³⁹Pu fission.

If equilibrium has been established, the instant induced fissioning is stopped, the number of delayed neutrons emitted per second for the i th group is

$$N_{ei} = \sigma_f \phi Y_i P.$$

At any time t (sec) after irradiation, the neutron emission rate of group i is

$$N_{ei} = \sigma_f \phi Y_i P e^{-\lambda_i t},$$

where Y_i = the yield of group i delayed neutron per fission, λ_i = the delayed neutron decay constant for group i = $0.693/T_{i1/2}$, and σ_f , ϕ , P are as defined previously.

The total number of neutrons emitted per second at time t after irradiation is given by:

$$N_e = \sum_{i=1}^6 N_{ei} = \sum_{i=1}^6 \sigma_f \phi Y_i P e^{-\lambda_i t}, \quad (3)$$

$$N_c(t) = N_e \epsilon = \sigma_f \phi P \epsilon (Y_1 e^{-\lambda_1 t} + Y_2 e^{-\lambda_2 t} + \dots + Y_6 e^{-\lambda_6 t}),$$

where P (atoms) is again given as $(1.68 \times 10^{21}) C_g \text{ MT(R)}_{1/2}$. Therefore,

$$N_c(t) = (1.68 \times 10^{21}) \sigma_f \phi \epsilon C_g \text{ MT(R)}_{1/2} [Y_1 e^{-\lambda_1 t} + Y_2 e^{-\lambda_2 t} + \dots + Y_6 e^{-\lambda_6 t}]. \quad (4)$$

Here, we assume that $\phi = 10^6$ n/cm²-sec, $\epsilon = 0.20$, $M = 10.4$ kg, $t = 3.0$ min = 180 sec, and $N_c(180) = 1$ count/sec above the (α ,n) and gamma background.

Next we ask: What is the minimum detectable amount of ^{239}Pu and ^{233}U after 3 min? If

$$1 = (1.68 \times 10^{21})(7.41 \times 10^{-22})10^6(0.20)C_g(10.4) \\ \times (2.4390 \times 10^4)(3.1 \times 10^{-5}),$$

then

$$C_g = 4.9 \times 10^{-7} \text{ Ci/g for } ^{239}\text{Pu}.$$

For ^{233}U , we have

$$1 = (1.68 \times 10^{21})(5.24 \times 10^{-22})10^6(0.20)C_g(10.4) \\ \times (1.62 \times 10^5)(6.7 \times 10^{-5}),$$

and

$$C_g = 4.3 \times 10^{-8} \text{ Ci/g.}$$

If we project back to $t = 0$ on the decay curve and set $N_c(0) = 10$ count/sec, then for ^{239}Pu , we find

$$10 = 1.68 \times 10^{21} (7.41 \times 10^{-22}) 10^6 (0.20) C_g (10.4) \\ \times (2.4390 \times 10^4) (6.1 \times 10^{-3}),$$

and

$$C_g = 2.4 \times 10^{-8} \text{ Ci/g.}$$

If a large enough delayed-neutron count is obtained, permitting the decay to be followed accurately, this method can help identify the predominant TRU isotopes in the waste material. This is possible because each fissioning isotope has a characteristic time for delayed-neutron emission. The one problem that might confuse the decay data is caused by the neutrons, both delayed and prompt, that are produced by the spontaneously fissioning isotopes present in the waste.

(α, n) Reaction Neutrons

Again, the same instrumentation is used to count the spontaneous, induced, and delayed neutrons produced by the (α, n) reaction. Although sufficient neutrons are produced to be counted, there is no way to relate the count observed to the quantity of TRU isotopes present in the waste. Thus we do not consider the use of this reaction to be a viable method.

DETECTION BY FISSION-GAS GENERATION

The noble gases krypton and xenon are generated in the fissioning process and diffuse throughout the voids in the waste material. If the waste container is sealed, the concentration of these gases will increase to an equilibrium value as fissioning continues. Because many of the isotopes of krypton and xenon emit gamma rays, we can measure and identify them with standard gamma-ray spectroscopy.

The volume of the void in the waste container is unknown and therefore, some technique must be developed to circumvent this difficulty. There are two readily feasible methods and either or both can be used. First, after the container has been sealed for a specified time, a vacuum pump removes gas from the bottom of the container as fresh air is introduced at the top.

The noble gases are sucked out of the waste container, through a sample line, and into a monitoring chamber where the concentrations of the gases are continuously monitored. The monitoring chamber is shielded from the influence of background gamma activity. As pumping continues, the concentration of noble gases decreases exponentially. A semilog plot of this decrease in concentration vs time is made and the volume of voids can be determined from the slope of the curve.

The second method involves collecting the noble gases in a molecular sieve until essentially all noble gases accumulated are removed from the waste container. The molecular sieve is attached to the sample line, downstream of the monitoring chamber. After sampling is complete, the molecular sieve is removed and taken to the laboratory where a sensitive gamma spectrometer is used to determine the quantity of a specific isotope of krypton or xenon in the sample.

Krypton and Xenon Generated by Spontaneous Fission

The method best suited to measure the krypton or xenon created by spontaneous fission is to monitor the concentration in the exhaust gas as the waste container is ventilated. We define the following terms: C = the concentration in the exhaust stream of a specific isotope of krypton or xenon at any sample time t (atom/cm³), λ_{SF} = the spontaneous-fission rate constant (min⁻¹), λ_r = the decay constant of the specific isotope monitored (min⁻¹), Y = the fission yield in percent of the specific isotope monitored, P = the number of atoms of TRU isotopes in the waste container, V = the volume of voids (cm³), and R = the flow rate of the exhaust stream (cm³/min).

The time rate of change in concentration is given by

$$\frac{dC}{dt} = \left(\frac{\lambda_{SF} P Y}{V} \right) - \lambda_r C - \delta C,$$

where $\delta = R/V$. The boundary conditions are that, at $t = 0$,

$$C_0 = \frac{\lambda_{SF} P Y}{V \lambda_r} (1 - e^{-\lambda_r t}),$$

where t is the time the waste container has been sealed before sampling. The solution of the differential equation is therefore

$$C = \frac{\lambda_{SF}^P Y}{(\lambda_r + \delta) V} \left[1 - e^{-(\lambda_r + \delta)t} \right] + C_0 e^{-(\lambda_r + \delta)t},$$

or

$$C = \frac{\lambda_{SF}^P Y}{(\lambda_r + \delta) V} \left[1 - e^{-(\lambda_r + \delta)t} \right] + \frac{\lambda_{SF}^P Y}{V \lambda_r} (1 - e^{-\lambda_r t}) e^{-(\lambda_r + \delta)t}. \quad (5)$$

If we change the equation to indicate the radioactive disintegration rate (dis/min/cm³), we have

$$\begin{aligned} \lambda_r C = & \left[\frac{\lambda_r \lambda_{SF}^P Y}{(\lambda_r + \delta) V} \right] \left[1 - e^{-(\lambda_r + \delta)t} \right] \\ & + \left(\frac{\lambda_{SF}^P Y}{V} \right) (1 - e^{-\lambda_r t}) e^{-(\lambda_r + \delta)t}. \end{aligned} \quad (6)$$

As an example of applying this method, let us say we want to know the quantity of ²⁵²Cf in a 55-gal waste container (assuming ²⁵²Cf is the only TRU present in the waste). The fission-gas isotope monitored is ⁸⁸Kr which has several high-energy gammas associated with its decay. A selected gamma ray is monitored with a 3 × 3 in. NaI detector using a single-channel analyzer to discriminate against all other gamma energies. Pulses passing through the energy discriminator feed to a count-rate meter and recorder. Through previous calibrations, the observed count rate can be directly related to the activity concentration in the monitoring chamber. Thus, we know that $\lambda_r = 0.693/2.80 \text{ hr} = 4.13 \times 10^{-3} \text{ min}^{-1}$, $\lambda_{SF} = 0.693/85 \text{ yr} = 1.55 \times 10^{-8} \text{ min}^{-1}$, $Y = 1.3$ (assumed; actual value must be determined), $\lambda_r t \geq 10$ (such that $1 - e^{-\lambda_r t} \approx 1$), $R = 10^3 \text{ cm}^3/\text{min}$, and $M = 10.4 \text{ kg}$.

In Fig. 2, we provide the semilog plot of the concentration vs time that was obtained. For this example, sampling was conducted for 120 min to display the full curve. In practice, a 30-min sampling period is sufficient. As a first approximation, the initial slope of the curve represents $\lambda_r + \delta$ in Eq. (6).

Fig. 2. Time plot of ^{88}Kr concentration during sampling. The broken line represents the initial slope of the curve.

Having found that

$$\lambda_r + \delta = 0.103 \text{ min}^{-1},$$

and knowing that

$$\lambda_r = 4.13 \times 10^{-3} \text{ min}^{-1},$$

we know therefore that

$$\delta = 0.099 \text{ min}^{-1}.$$

By definition, $\delta = R/V$. The sampling rate $R = 10^3 \text{ cm}^3/\text{min}$ and therefore the volume of voids in the waste container $V = 1.01 \times 10^4 \text{ cm}^3$.

Returning to Eq. (6) and taking the value of $\lambda_r C$ when $t = 0$, we can solve the equation for P . Thus at $t = 0$,

$$\lambda_r C = \frac{\lambda_{SF} P Y}{V}.$$

Therefore,

$$P = \frac{(9.3 \text{ dis/min/cm}^3)(10^4 \text{ cm}^3)}{(1.55 \times 10^{-8} \text{ min}^{-1})(1.3 \times 10^{-2})},$$

$$= 4.62 \times 10^{14} \text{ atoms}.$$

This represents $1.04 \times 10^{-4} \text{ Ci}$ of ^{252}Cf . Since 10.4 kg of waste material is contained in the drum, the average specific activity is 10 nCi/g.

In reality, the interpretation of these data is more complex because other radioactive isotopes of krypton and xenon are also present to some degree. Compton scattering of higher-energy gammas from these other isotopes is counted in the energy channel selected for ^{88}Kr . This, in turn, results in exaggerated calculations of the TRU content in the waste container. However, this difficulty can be overcome by proper calibration of the detector and by careful selection of the fission-gas isotope and gamma energy for the TRU isotope being measured. It is also possible to monitor several gamma energies simultaneously to obtain a ratio of fission-gas isotopes present. In some cases, this helps to identify the TRU undergoing spontaneous fission.

To obtain the desired sensitivity in an actual situation, the monitoring chamber should have a volume of one liter or more. The main disadvantage of this method is the low sensitivity because detection is possible only when

$$\frac{T(R)_{1/2}}{T(SF)_{1/2}} > \frac{4.5 \times 10^{-8} V}{M Y},$$

where V is the volume in cubic centimeters of the void in the waste container, M is the mass in kilograms of the waste, and Y is the fission yield of the noble gas isotope selected for measurement. Because V is unknown and varies from container to container, the minimum detectability cannot be determined. However, the value of V must be less than the volume of the container and thus, we can establish a lower limit of detectability. The only TRU isotopes that can be detected at the 10-nCi/g level by this method are ^{248}Cm , ^{250}Cm , and ^{252}Cf . If we increase the limit to 1 $\mu\text{Ci/g}$, ^{244}Pu , ^{246}Cm , and ^{250}Cf may also be measured by this method.

Krypton and Xenon Generated by Induced Fission

The method used here is essentially the same as that used to detect the krypton and xenon generated by spontaneous fission. A sealed drum of waste is irradiated by neutrons to induce fission in the TRU isotopes. After irradiation, the gases are removed from the drum and carried through a sample line to a monitoring chamber. A molecular sieve located downstream of the monitoring chamber collects the noble gases. The sieve increases the sensitivity of detection for two reasons. First, the sieve effectively collects all the gases in a small volume and second, counting takes place under controlled laboratory conditions.

Once again, we let C equal the concentration (atcm/cm³) of the noble-gas isotope of interest at any time t after neutron irradiation for a time t. Immediately after irradiation, the drum is sampled as described. The time rate of change of C during sampling is then $dC/dt = -\lambda_r C - \delta C$.

The boundary conditions for solution of the equation are that, at t = 0,

$$C_0 = \frac{\sigma_f \phi P Y}{V(\lambda_r + \sigma_c \phi)} \left[1 - e^{-(\lambda_r + \sigma_c \phi)t} \right],$$

where σ_f = the fission cross section of TRU (barn), σ_c = the capture cross section of the noble-gas isotope (barn), ϕ = the neutron flux density (n/cm²-sec), and t = the irradiation time (sec). Solving for C, we find

$$C = C_0 e^{-(\lambda_r + \delta)t},$$

or

$$C = \frac{\sigma_f \phi P Y}{V(\lambda_r + \sigma_c \phi)} \left[1 - e^{-(\lambda_r + \sigma_c \phi)t} \right] e^{-(\lambda_r + \delta)t}. \quad (7)$$

To determine the number of atoms of the noble gas collected on the filter after a sampling time t , we let $dq/dt = (CR) - (\lambda_r q)$, where q = number of atoms of the noble-gas isotope of interest on the filter and $C = C_0 e^{-(\lambda_r + \delta)t}$. The boundary conditions are that, at $t = 0$, $q = 0$ and $q = (C_0 R / \delta)(1 - e^{-\delta t}) e^{-\lambda_r t}$. Alternatively, in terms of the radioactive disintegration rate where $\delta = R/V$,

$$\lambda_r q = \frac{(\lambda_r \sigma_f \phi P Y)}{(\lambda_r + \sigma_c \phi)} \left[1 - e^{-(\lambda_r + \sigma_c \phi)t} \right] (1 - e^{-Rt/V}) e^{-\lambda_r t}. \quad (8)$$

In the following examples, we use and assume the following conditions. The TRU in the waste is ^{239}Pu (for simplicity, only one TRU isotope is assumed to be present). The neutron flux density is 10^6 n/cm²-sec for an irradiation time of 30 min. The 55-gal drum is sampled at a rate of 10 liter/min for 120 min. The molecular sieve is counted 10 min after collection and 180 dis/min of ^{88}Kr are observed in the sieve. The total mass of material in the drum is 10.4 kg. Therefore, we can solve Eq. (8) since we know that: $\lambda_r = 4.13 \times 10^{-3}$ min⁻¹, $\sigma_f = 7.41 \times 10^{-22}$ cm², $\sigma_c \approx 0$, $\phi = 6 \times 10^7$ n/cm²-min, $Y = 1.39 \times 10^{-2}$, $t = 30$ min, $R = 10^4$ cm³/m, $M = 10.4$ kg, $t = 120$ min, and $\lambda_r q = 180 e^{4.13 \times 10^{-3}(10)} = 188$ dis/min. Solving Eq. (8) for P , we find

$$188 \text{ dis/min} = (7.41 \times 10^{-22})(6 \times 10^7) P(1.39 \times 10^{-2}) \\ \times \left[1 - e^{(-4.13 \times 10^{-3})30} \right] e^{(-4.13 \times 10^{-3})120}.$$

Since

$$1 - e^{-R/V t} = 1, \quad P = 4.28 \times 10^{18} \text{ atoms},$$

we know that 4.28×10^{18} atoms of ^{239}Pu equals 1.04×10^{-4} Ci. The waste weighs 10.4 kg and therefore the specific activity in the container must be 10^{-8} Ci/g or 10 nCi/g.

It is obvious that because a number of variables can be changed in the sampling procedure, an optimum selection of these variables is necessary for

any given application. If we assume that the minimum amount of ^{88}Kr which can be detected on the molecular sieve is 100 dis/min, that the waste mass is 10.4 kg in a 55-gal container, that the fission yield is at least 1.3% for ^{88}Kr , and that the sample rate, irradiation time, and sample time are as given in the example, then this method can measure ^{233}U , ^{239}Pu , and ^{245}Cm at the 10-nCi/g level. If the detection limit is raised to 1 $\mu\text{Ci/g}$, then ^{237}Np , ^{242}Pu , $^{242\text{m}}\text{Am}$, ^{249}Cf and ^{251}Cf can also be measured.

In an actual application, interpretation of the data is complex. Several TRU isotopes are generally present, each contributing to the fission-gas inventory. Several of the fission-gas isotopes contribute to the radioactivity of the molecular sieve. Uncertainty is also associated with the product of σ , ϕ , and Y . However, this last problem can be overcome by calibration runs on all TRU isotopes that may be encountered in the waste.

DETECTION BY HELIUM PRODUCTION

The very high sensitivity achieved in the today's helium-leak detectors has opened the possibility of using helium detection to measure the quantity of TRU isotopes in waste. Because alpha particles are helium nuclei, the production rate of helium is simply the disintegration rate of the TRU isotopes. Such a detection method would be very simple and straightforward and would provide a measurement of all alpha-emitting TRU isotopes.

If we assume that TRU-contaminated waste in a 55-gal drum weighs 10.4 kg, then 10 nCi/g would produce helium at the rate of 2.3×10^8 atom/min or 3.3×10^{12} atom/wk. Helium detectors currently available have sensitivities in the range of 10^{-11} atm-cm³/sec which converts to 2.7×10^8 atom He/sec entering the detector.

However, the high level of naturally occurring helium in the atmosphere does constitute a major difficulty with this method. One atmosphere of air contains approximately 5.24×10^{-4} mol-fraction% of He^9 or about 1.4×10^4 atom/cm³. The natural helium content in air obscures (by several orders of magnitude) the helium that would be produced by alpha decay in a reasonable time. Therefore, for any practical application of the method, air must be excluded from the waste material being tested. One means by which to overcome the helium background is to purge the waste container with a pure gas such as nitrogen before sealing the container for the buildup period.

After a week or more has elapsed since the drum was purged and sealed, the container is sampled with the helium detector and the concentration of helium is measured. The void volume of the container is unknown and thus, several measurements must be made as known volumes of pure nitrogen are purged through the container. A semilog plot of the decrease in the helium concentration yields the void volume.

If we let C equal the concentration of helium in the container at any time t after purging with nitrogen at a rate R cm³/min, then the time rate of change of C in the container is

$$\frac{dC}{dt} = \frac{\lambda R}{V_a} - \delta C,$$

where V_a is the apparent void volume of the container and $\delta = R/V_a$. The boundary conditions in the solution of this equation are that, at $t = 0$ (the time purging begins),

$$C_0 = \frac{\lambda R \uparrow}{V_a},$$

where $\uparrow$ is the time in minutes for which the container has been sealed for helium buildup. Therefore,

$$C = \frac{\lambda R}{V_a \delta} (1 - e^{-\delta t}) + \frac{\lambda R \uparrow}{V_a} e^{-\delta t}. \quad (9)$$

Suppose, for example, we assume an initial purge of the container (55-gal drum containing 10.4 kg of TRU waste) with 4 m³ of pure nitrogen over a 30-min period to eliminate all but 5×10^{-9} fraction of the air. The drum is then sealed for 10 d, after which the helium concentration in the drum is measured. The container is purged with pure nitrogen at a rate of 1.0 liter/min and the concentration is measured every 5 min for the first 20 min. The following data is obtained for each time t (see Fig. 3):

Fig. 3. Time plot of helium concentration during sampling.

$$\begin{aligned}
 t = 0, & \quad C = 1.66 \times 10^9 \text{ atom/cm}^3; \\
 t = 5 \text{ min}, & \quad C = 1.29 \times 10^9 \text{ atom/cm}^3; \\
 t = 10 \text{ min}, & \quad C = 1.01 \times 10^9 \text{ atom/cm}^3; \\
 t = 15 \text{ min}, & \quad C = 7.83 \times 10^8 \text{ atom/cm}^3; \\
 t = 20 \text{ min}, & \quad C = 6.11 \times 10^8 \text{ atom/cm}^3.
 \end{aligned}$$

From the curve in Fig. 3 we obtain $\delta = 0.050 \text{ min}^{-1}$. Since $\delta = R/V_a$ and $R = 10^3 \text{ cm}^3/\text{min}$, then $V_a = 2 \times 10^4 \text{ cm}^3$. We return to Eq. (9) and solve for λP when C equals the value at $t = 0$:

$$\begin{aligned}
 \lambda P &= (2 \times 10^4 \text{ cm}^3) \\
 &\times \frac{(1.66 \times 10^9 \text{ atom/cm}^3)}{1.44 \times 10^4 \text{ min}},
 \end{aligned}$$

or

$$\lambda P = 2.3 \times 10^9 \text{ dis/min} \approx 10^{-3} \text{ Ci.}$$

The specific activity in the waste is therefore 10^{-7} Ci/g .

The minimum-detection level varies with the length of time the drum is sealed, the void volume, and the sampling rate of the helium detector. If the detector sampling rate is set at $10 \text{ cm}^3/\text{sec}$ and the concentration is measured after 10 d, the minimum-detectable amount of TRU is determined to be $(8.3 \times 10^{-12})(V_a/M) \text{ Ci/g}$. This is ten times the instrument minimum sensitivity for a determination of V_a by the concentration clean-up method. However, it is still questionable whether this method will be able to detect TRU at the 10-nCi/g level because V_a will be large (probably $> 10^4 \text{ cm}^3$) and M generally will not be greater than 50 kg for handling reasons. Significant problems associated with this method are as follows:

- It is difficult to clean out the air from the waste container to eliminate the natural occurring helium.
- The presence of other alpha emitters in the waste such as uranium and radium daughters also will generate helium.

- The presence of tritium in the waste will generate ^3He through beta-minus decay.
- It is also difficult to seal a container for any length of time to retain the helium.

DETECTION BY GAMMA-RAY ANALYSIS

Decay Gamma Rays

All of the TRU isotopes emit photons in their radioactive decay, either as gamma rays or as fluorescent x rays. Many of these emissions are of low energy and low yield, compounding the difficulty of measurement. The presence of a gamma background of a few tenths of a milliRoentgen per hour degrades the sensitivity so that most TRU isotopes cannot be detected at the 10-nCi/g level. However, in the absence of a gamma background, this is one of the more sensitive methods of measuring TRU in waste. The method has been successfully applied at several laboratories for the detection of ^{239}Pu in waste material.^{2,3}

Let us assume that the TRU is uniformly distributed through the waste packaged in a 55-gal fiberboard drum. The photon flux density ϕ at a distance from the edge of the drum is given by

$$\phi = \frac{(B S_V) R_0^2}{4(a + Z)} F(\theta_1, b_2) + F(\theta_2, b_2) \quad (10)$$

Here, B = the buildup factor, S_V = the source strength of a volume source (photon/sec-cm³), R_0 = the radius of the cylinder (cm), a = the distance from edge of cylinder to point of measurement (cm), Z = the effective self-attenuation distance (cm), and $F(\theta, b) = \int_0^\theta e^{-b \sec \theta} d\theta$. Also, $B_2 = \sum_{i=1}^n \mu_i t_i + \mu_s Z$, μ_1 = the microscopic cross section of shielding (cm⁻¹), μ_s = the microscopic cross section of the source matrix material (cm⁻¹), t = the thickness of shielding (cm), θ_1 = angle 1 as shown in Fig. 4, and θ_2 = angle 2 as shown in Fig. 4. Values of ϕ , R_0 , t , and a are obtained by direct measurement. Values of μ_1 , μ_s , and Z are obtained from published data or by empirical inference. For example, μ_s is obtained from a point source of monoenergetic photons of known energy (usually photons from a ^{137}Cs source) and extrapolating to the energy of interest. The source is placed on one side of the container and the flux density of the 662-keV photons is measured on the opposite side. If we use a gamma spectrometer to measure ϕ at a discreet energy and if the contribution of scattered photons of higher energies is subtracted

Fig. 4. Cylindrical source of photons.

from the observed photopeak, B can be taken as 1. Once we know ν_s , Z can be obtained along with θ_1 and θ_2 .

If a fiberboard barrel is used to package the waste for measurement, the shield thickness is simply the thickness of the fiberboard drum plus the air between the container and the point of measurement. The shielding effect of the drum and air is negligible and can be ignored. However, if a 55-gal steel drum is used, the metal will significantly attenuate the low-energy photons, reducing the sensitivity of the method.

Solving Eq. (10) for S_V , we obtain

$$S_V = \frac{4 \phi (a + Z)}{R_0^2 [F(\theta_1, b_2) + F(\theta_2, b_2)]} \quad (11)$$

Hence the specific activity SA can be obtained as

$$SA = \frac{S_V V_c}{M f (3.7 \times 10^7)} ,$$

where V_c is the volume of the 55-gal drum (m^3), M is the mass of waste material (kg), and f is the photon yield of energy E_i per disintegration.

In reality, the packaged waste is not a homogeneous mix of TRU isotopes. To compensate for this, the photon-flux density emitted from the container is measured at a number of positions around the drum and the resulting values of S_V obtained by each measurement are averaged. This is a tedious procedure. Most current systems have been automated and make one long count (> 1000 sec)

while the waste drum is rotated and the detectors scans the length of the drum from top to bottom. The detector is shielded from scattered radiation as much as possible to increase the sensitivity.

Usually a NaI detector (3 × 3 in.) is used for its high sensitivity. However, in special applications, other detectors may be more appropriate. For example, if we know that all gamma-energy measurements will be below 100 keV, then a thin NaI detector such as a Fiddler probe (0.125 × 5 in. diameter) can be used to reduce the instrument's response to high-energy photons. Or, if we have a large mixture of gamma emitters in the waste, a high-resolution Ge(Li) detector may be desirable. Thus, the type of detector selected would be the one best suited for the application at hand.

A multichannel, pulse-height analyzer is also often used to accumulate data over a wide energy spectrum (e.g., 20 to 1000 keV for a 3 × 3 in. NaI detector, 10 to 120 keV for a 0.125 × 5 in. diameter probe). This permits maximum utilization of the data and several TRU isotopes can be identified and quantified with one measurement.

For example, let us assume that a 55-gal fiberboard drum of waste of net weight 10.4 kg contains ^{237}Np . With a ^{137}Cs calibration source, we measure $\dot{\phi}_c$ as 780 photon/sec-cm² through air (ϕ obtained by measurement of the 662-keV photon peak). Without moving the discriminator, the source, or the detector, we insert the waste drum between the source and the detector and obtain 635 photon/sec-cm². We then remove the source, adjust the window to 86 keV for ^{237}Np , and begin the automated scan count of the drum. From this scan, we obtain an average ϕ of 480 photon/cm²-sec.

We have now gathered sufficient data to solve Eq. (11) for S_V . For 86-keV photons, $\mu_s = 7.6 \times 10^{-3} \text{ cm}^{-1}$. Therefore, $Z = 29 \text{ cm}$ (obtained from empirical data), $R_0 = 29 \text{ cm}$, $a = 79 \text{ cm}$, and $\theta_1 = \theta_2 = 24^\circ$ when the detector is at the center of drum. Thus,

$$S_V = \frac{(4)(480 \text{ photon/cm}^2\text{-sec})(100 \text{ cm})}{(29^2)(2)(0.44)} = 260 \text{ photon/sec-cm}^3,$$

and the specific activity is

$$SA = \frac{(260 \text{ photon/sec-cm}^3)(0.208)}{(10.4)(0.14)(3.7 \times 10^7)} = 10^{-6} \text{ Ci/g},$$

where 0.14 represents the yield of the 86-keV photon

In practice, the determination of ϕ is made from the count observed on the instrument. This involves many factors such as detector efficiency, detector resolution, backscatter contributions, detector size, count time, and background contribution (which varies with the radon concentrations in air). Therefore, any calculations of levels of minimum detectability made here will have a large uncertainty.

In the presence of a background of 20 $\mu\text{R/hr}$, typical of levels inside concrete structures, the minimum detectability of the TRU in waste that has been packaged at a density of 50 kg/m^3 in a 55-gal fiberboard drum and using a 3×3 in. NaI detector at 1 m from the center of the drum is

$$SA = (2.9 \times 10^{-10}) \phi / f .$$

Here, f = photon yield per disintegration and ϕ = photon flux density above natural background of 20 $\mu\text{R/hr}$ as determined from the count data. (We assume that photon-flux density contributed by the TRU present in the drum is detectable when the peak value of the photopeak is 20% above the background level.

In the presence of a 20- $\mu\text{R/hr}$ background, the following TRU isotopes can be detected at the 10-nCi/g level by this method, assuming a 1000-sec count: ^{233}U , ^{237}Np , ^{244}Pu , ^{241}Am , ^{243}Am , ^{243}Cm , ^{245}Cm , ^{248}Cm , ^{250}Cm , ^{247}Bk , ^{249}Cf , ^{252}Cf , and ^{254}Es . If the limit is raised to 1 $\mu\text{Ci/g}$, ^{236}Pu , ^{238}Pu , ^{239}Pu , $^{242\text{m}}\text{Am}$, ^{242}Cm , and ^{252}Es can also be detected. If a shorter count time is used, the minimum detectable level will be higher.

The calculations of the minimum detectable levels of specific activity are based on an ideal case where only one TRU isotope is considered to be present. In a real case however, several TRU isotopes are present and the Compton scattering from higher-energy photons within the energy region of interest degrades the sensitivity of detection. Several techniques are available though to minimize the Compton contribution including housing the NaI detector in a large-volume graded shield, collimating the photons entering the detector, and using coaxial detectors with anticoincidence counting.

The main difficulty with this gamma-counting method is that its sensitivity drops significantly as the background level increases. Above a background of 10 mR/hr , the counting system cannot accurately detect the photons. Table 4 lists the minimum detectable specific activity in a 55-gal

drum with a packing density of 50 kg/m^3 as measured with a 3×3 in. NaI detector in the presence of a gamma background. Only ^{241}Am (60 keV) and ^{239}Pu (375 keV) are shown.

Table 4. Minimum detectable specific activities (Ci/g) in gamma backgrounds.

TRU Isotope	Gamma Background Level (mR/hr)				
	0.02	0.10	1.0	5.0	10.0
^{241}Am	1.3×10^{-10}	4.3×10^{-10}	4.3×10^{-9}	1.5×10^{-8}	4.3×10^{-8}
^{239}Pu	1.9×10^{-5}	1.4×10^{-4}	1.4×10^{-3}	4.6×10^{-3}	1.4×10^{-2}

Depending on the TRU isotopes present in the waste, techniques can be adopted to reduce the effect of a gamma background. For example, if photon energies below 100 keV are to be counted, a very thin NaI detector can be used in front of a thick crystal and only counts made in anticoincidence are registered. With such schemes, the background effects can be reduced fivefold.¹⁰ Also, Ge(Li) detectors of high resolution used with coaxial anticoincident systems can reduce background effects. Because of their smaller surface area and volume, solid state detectors are less sensitive. However, by assembling arrays of Ge(Li) detectors, researchers have been able to increase the sensitivities of solid state detectors to that of a good NaI detector.

Another difficulty with gamma counting is the photon attenuation that occurs in the waste matrix. Packing densities of 250 kg/m^3 are not uncommon in waste drums and packing densities are not uniform throughout the container volume. Attenuation of the low-energy photons can be appreciable and difficult to determine. Drums with high packing densities can probably not be measured with photon-counting methods.

Prompt Gamma Rays

An average of seven prompt gammas are produced per fission. These prompt gammas have energies of 0.3 to 10.5 MeV and the total energy carried away by the photons is about 7.2 MeV/fission. In the energy range of 0.3 to 1.0 MeV, 4.55 prompt photons are produced per fission whereas in the energy range of 3.0 to 4.0 MeV, only 0.161 photons are produced per fission.¹¹

Therefore, it is not feasible to count photons in coincidence pairs above 1.5 MeV because about six of the seven photons per fission are emitted

below this energy level and are thus undetectable. (The discriminator must be set at 1.5 MeV to overcome the background interference.) Sometimes a prompt gamma is counted in coincidence with a prompt neutron to differentiate between the prompt photon or neutron and the background radiations.

However, it is not necessary to take such a sophisticated approach to count the prompt gammas from waste drums. All of the aged fission products and TRU gamma emissions are below 3 MeV/photon. By setting the low-level energy discriminator at 3 MeV, we can reject all photons emitted by the fission products and TRU isotopes and count only the prompt gammas from fission. Approximately 0.25 photons per fission have energies greater than 3 MeV.¹¹ Prompt-gamma counting can be conducted with the same instrumentation used to detect the decay gammas. However, it may be necessary to place a lead window in the collimator to reduce the background photon-flux density that enters the detector to prevent the counting of low-energy photons because of pulse pile-up.

Equation (10) may again be used to calculate the photon-flux density ϕ entering the detector. We shall consider only the photons with energy > 3 MeV. The following data is known: $R_0^2 = 853 \text{ cm}^2$, $a + A = 100 \text{ cm}$, $B \approx 1$, and $F(\theta_1, b_2) + F(\theta_2, b_2) = 0.32$ for $\theta_1 = \theta_2 = 24^\circ$ and with a 2-cm lead window in the collimator. Solving Eq. (11) for S_V as before and inserting the above known values, $S_V = 146 \phi$. For spontaneously fissioning isotopes, S_V is related to P_V (the number of atoms of a specific TRU isotope per cubic centimeter) by the equation,

$$S_V = \lambda_{SF} P_V Y.$$

Since

$$P_V = \frac{(3.7 \times 10^{10})(C_g)M}{\lambda_r V_c},$$

then

$$S_V = \frac{(3.7 \times 10^7)Y(C_g)M}{V_c} \frac{T(R)_{1/2}}{T(SF)_{1/2}},$$

or

$$\phi_{SF} = (2.53 \times 10^7) \frac{Y(C_g)M}{V_c} \frac{T(R)_{1/2}}{T(SF)_{1/2}},$$

where Y = the photon yield per fission, C_g = the curies per gram of TRU, M = the mass of waste (kg), V_c = the volume of waste container (m^3), and $T(R)_{1/2}$ and $T(SF)_{1/2}$ are as defined previously.

For induced fission,

$$S_V = \sigma_f(nv)P_V Y.$$

Making the appropriate substitution, we find

$$\phi_{IF} = \frac{(1.68 \times 10^{-9})(Cg)M'\sigma_f(nv)T(R)}{V_c}^{1/2},$$

where σ_f is the fission cross section (barn), nv is the neutron-flux density (n/cm^2 -sec), and the other symbols are as defined before.

The observed count c for any given photon is related to ϕ by the equation

$$c = \phi e P_K A t,$$

where e = the total absolute efficiency of the detector and counting system determined by calibration, P_K = the peak-to-total ratio determined by calibration, A = the area of the detector normal to the incident photon flux (cm^2), and t = the time of the count (sec).

In practice, the barrel is rotated during the count as it is scanned from top to bottom to minimize the effect of a nonuniform distribution of TRU in the waste. The minimum count that can be determined is about 100 counts for photons of energy > 3 MeV because the background at this energy level is essentially zero. The TRU isotopes undergoing spontaneous fission that can be detected at the 10-nCi/g level are ^{248}Cm , ^{250}Cm , and ^{252}Cf . If the level is raised to 1 $\mu Ci/g$ then ^{244}Pu , ^{246}Cm , ^{248}Cf , and ^{250}Cf can also be detected. However, if we subject the waste material to a neutron-flux density of 10^6 n/cm^2 -sec, only ^{233}U can be detected at the 10-nCi/g level by counting the prompt gammas from induced fission. If the limit is raised to 1 $\mu Ci/g$, then we can also detect ^{239}Pu , ^{242m}Am , ^{248}Cm , and probably ^{249}Cf (see Table 5). Because of the counting technique used, these levels can probably be detected in the presence of a 1-R/hr background.

The main difficulty encountered with this method is the uncertainty associated with the product of σ_f , nv , and Y . However, calibrations made with standards of each isotope of interest should overcome this difficulty. A problem may arise from the fact that gammas created by the (n,γ) reaction on the material undergoing irradiation may create a high-energy gamma background. Such a background would seriously degrade the sensitivity of the method, contributing an undeterminable bias to the count. Whether the gamma background

Table 5. Summary of minimum-detection levels^a for all TRU isotopes by non-destructive testing at a packing density of 50 kg/m³.

TRU	SF Neutron ^b (CI/g)	Kr-Xe(SF) ^c (CI/g)	Kr-Xe(1F) ^d (CI/g)	X Ray or Gamma Ray ^e (CI/g)	Delayed Neutrons ^f (CI/g)	²⁵² Cf ^g (CI/g)	Prompt Gamma (SF) ^h (CI/g)	Prompt Gamma (1F) ⁱ (CI/g)
²³³ U	-	-	3×10^{-9}	9×10^{-9}	5×10^{-9}	-	-	1×10^{-8}
²³⁷ Np	3×10^{-1}	3×10^2	2×10^{-6}	5×10^{-10}	1×10^{-5}	-	4×10^{-1}	5×10^{-5}
²³⁸ Pu	9×10^{-4}	8×10^{-1}	2×10^{-4}	7×10^{-7}	9×10^{-4}	-	1×10^{-1}	3×10^{-3}
²³⁸ Pu	4×10^{-4}	4×10^{-1}	7×10^{-5}	1×10^{-6}	3×10^{-4}	-	5×10^{-2}	9×10^{-4}
²³⁹ Pu	2×10^{-1}	2×10^2	5×10^{-9}	1×10^{-7}	2×10^{-8}	-	1×10^{-1}	7×10^{-8}
²⁴⁰ Pu	2×10^{-5}	2×10^{-2}	2×10^{-4}	9×10^{-2}	9×10^{-4}	-	2×10^{-3}	2×10^{-3}
²⁴² Pu	1×10^{-7}	1×10^{-4}	1×10^{-6}	-	6×10^{-6}	-	2×10^{-5}	2×10^{-5}
²⁴² Pu	2×10^{-10}	2×10^{-7}	-	4×10^{-10}	-	-	3×10^{-8}	-
²⁴¹ Am	3×10^{-1}	1×10^2	7×10^{-5}	1×10^{-10}	3×10^{-4}	-	4×10^1	9×10^{-4}
²⁴² Am	-	-	1×10^{-7}	8×10^{-8}	5×10^{-7}	10^{-8}	-	1×10^{-6}
²⁴³ Am	-	-	2×10^{-4}	1×10^{-10}	8×10^{-4}	but	-	2×10^{-3}
²⁴² Cm	1×10^{-5}	1×10^{-2}	4×10^{-2}	2×10^{-7}	2×10^{-1}	probably	1×10^{-3}	6×10^{-1}
²⁴³ Cm	-	-	5×10^{-6}	3×10^{-9}	2×10^{-5}	10^{-6}	-	6×10^{-5}
²⁴⁴ Cm	5×10^{-7}	5×10^{-4}	-	6×10^{-6}	-	for all	6×10^{-5}	-
²⁴⁵ Cm	-	-	5×10^{-9}	1×10^{-9}	3×10^{-8}	isotopes	-	7×10^{-8}
²⁴⁶ Cm	2×10^{-9}	2×10^{-6}	-	-	-	-	3×10^{-7}	-
²⁴⁷ Cm	-	-	-	-	-	-	-	-
²⁴⁸ Cm	7×10^{-12}	7×10^{-9}	-	10^{-8} SF γ	-	-	8×10^{-10}	-
²⁵⁰ Cm	3×10^{-13}	3×10^{-10}	-	10^{-8} SF γ	-	-	8×10^{-11}	-
²⁴⁷ Bk	-	-	-	2×10^{-9}	-	-	-	-
²⁴⁸ Cf	1×10^{-8}	1×10^{-5}	-	-	-	-	1×10^{-6}	-
²⁴⁹ Cf	3×10^{-6}	3×10^{-3}	2×10^{-7}	6×10^{-10}	7×10^{-7}	-	3×10^{-4}	2×10^{-6}
²⁵⁰ Cf	9×10^{-10}	9×10^{-7}	2×10^{-5}	-	1×10^{-4}	-	1×10^{-7}	3×10^{-4}
²⁵¹ Cf	-	-	4×10^{-8}	-	2×10^{-7}	-	-	5×10^{-7}
²⁵² Cf	1×10^{-11}	2×10^{-8}	-	10^{-8} SF γ	-	-	3×10^{-9}	-
²⁵² Es	-	-	-	4×10^{-8}	-	-	-	-
²⁵⁴ Es	6×10^{-7}	6×10^{-4}	-	2×10^{-9}	-	-	8×10^{-5}	-

Footnotes for Table 5

^a Minimum-detection levels are used here as the minimum levels that appear to be obtainable with reasonable effort. Because none of the methods (except gamma-ray analysis) has been demonstrated, these values are calculated estimates. A conservative approach was taken in the calculations and thus we believe that, in most cases, these values can be bettered.

^b Determined by coincidence counting of spontaneous-fission prompt neutrons.

^c Determined by measurement of the build-up of noble-fission-gas concentration as spontaneous fission takes place in a sealed container. Analysis is made of ⁸⁸Kr concentration after equilibrium has been established.

^d Determined by measurement of the amount of noble fission gases produced from induced fission by an irradiating neutron flux of 10^6 n/cm²-sec. An irradiation time of 30 min and a decay time of 2 hr are assumed; ⁸⁸Kr is the chosen isotope.

^e Determined by gamma-ray analysis of one selected photon energy in presence of a natural background of 20 μ R/h. Count time is taken as 1000 sec in each case.

^f Determined by counting delayed neutrons after container is subjected to a neutron irradiation-flux density of 10^6 n/cm²-sec until equilibrium is established for ⁸⁷Br (~10 min). Delayed-neutron yield data for thermal fissions are not available for isotopes other than ²³³U and ²³⁹Pu. Values for other isotopes are calculated from estimated yield values.

^g Determined after sealing drum for 10 d followed by purging with nitrogen and measuring the helium concentration.

^h Determined by counting prompt gammas from spontaneous fission. Only photons of >3 MeV are counted after passing through a collimator and 2-cm thick lead window.

ⁱ Determined by counting prompt gammas from induced fission by thermal or epithermal neutrons with a flux density of 10^6 n/cm²-sec. Only photons with energies of >3 MeV are counted after they pass through a collimator and 2-cm thick lead window.

is great enough to significantly degrade the detection sensitivity will have to be determined experimentally.

DETECTION BY NEUTRON ACTIVATION

Even a cursory study of this method reveals that, for nondestructive testing, this is not a practical approach. When given a reasonable neutron flux and irradiation time, the sensitivity is unacceptably low.

Summary Discussion

The nondestructive analytical methods described in this report can all be grouped into four basic detection schemes, neutron counting, photon counting, fission-gas production measurements, and helium production measurements. In the past five years, considerable research has been conducted on nondestructive testing of Special Nuclear Material in connection with the Safeguards Program.⁷ We have reviewed some of the methods developed for the Safeguards program to measure small quantities of TRU in waste material.

Table 5 is a summary of the minimum theoretical detection limits that are possible for each alpha-emitting TRU isotope ($T_{1/2} > 100$ d) for each of the eight viable detection methods. Where blank spaces exist, insufficient data were available on the isotope to calculate a detection level. A conservative approach was taken for the calculations. Therefore, these levels should be achievable with instrumentation and techniques that are available today. In fact, we believe that with some research and operating experience, the limits for some of the isotopes can be lowered by as much as a factor of five. The major advancements in the next few years most probably will be in the area of solid-state detectors. However, we do not believe that tenfold increases in sensitivity will be readily achievable with these developments.

Three of the eight methods presented in this report involve irradiating the waste material with thermal or epithermal neutrons at a flux density around 10^6 n/cm²-sec. Computer neutron diffusion codes have shown that a reasonably uniform flux (within $\pm 20\%$) can be obtained over the volume of a 55-gal waste container using the special configuration of neutron sources shown in Fig. 1. An estimate of the costs to build and operate a neutron irradiation and counting facility is itemized in Appendix A.

A suggested test procedure for neutron counting and krypton or xenon sampling is given in Appendix B. In this procedure, five separate neutron counts are made on each waste container. The neutron-count data yield the spontaneous-fission rate of the material in the container, the delayed-neutron count rate decay, and the effect of the waste matrix on the neutrons. The total time for the count analysis for each waste container is approximately 60 min.

Krypton and xenon sampling take about 2 hr but sampling can be conducted in parallel with several containers sampled simultaneously. The average production rate is then approximately 6 containers per 8-hr shift or 80 min per container. The minimum crew required is one trained technician to operate the neutron generators, take the counts, and collect the samples plus a scientist to calibrate the systems as well as analyze and interpret the data. The data analysis perhaps can be done with an on-line minicomputer, requiring only part-time scientific support. Because of the hazard associated with the neutron generators, elaborate safety procedures and interlocks are needed to prevent radiation injury to the operators.

Ten of the twenty seven TRU isotopes can be detected at the 10-nCi/g level using these neutron methods. The main uncertainty with neutron counting arises from the effect of the neutron background [e.g., from the (α, n) reactions] on the detection sensitivity. A pilot study needs to be made before a commitment is made to build a full scale facility. The added analytical complexity from the presence of several TRU isotopes in the waste can be handled with a computer program. The ratios of the various isotopes must be determined by other means; neutron counting cannot identify the specific isotopes present.

The helium production measurement method is not as sensitive as neutron or gamma counting but it appears to have considerable potential and is worthy of further research. We estimate that one trained technician could test six to eight containers per 8-hr shift after the containers have been prepared, sealed, and stored for ≥ 10 d to permit helium buildup. An itemized cost estimate for two different helium detection facilities is given in Appendix C.

The main uncertainty involved with this method is whether the waste container can be sealed tightly enough to contain the helium. Any leak of helium will cause the calculated amount of TRU in the container to be less than what was actually present. A possible solution is to construct a vacuum

chamber large enough to hold one or more waste containers and which can be pumped down to $\leq 10^{-4}$ Torr. Helium measurements can be made within a few hours after pump-down to obtain the helium leak rate. Calculations then give the alpha-activity level, averaged over the number of containers in the chamber.

Where ^{239}Pu or ^{233}U alone are the major TRU isotopes to be tested, the production of krypton or xenon by induced fission appears to be an attractive method. As an estimate, one technician can test about four containers per 8-hr shift. He also can prepare the molecular sieves which are then sent to the radiochemistry laboratory for the noble-gas analysis. A cost estimate for such an irradiation and sampling facility is given in Appendix D. The major problem with this method is the complexity of interpreting the gas-analysis data.

In the absence of a gamma-ray background, photon counting is a feasible method. One trained technician can count ten to twelve containers per 8-hr shift. Part-time scientific support is required to calibrate the equipment and to review the analysis methods. Appendix E gives the estimated cost for such a facility. The major uncertainty with this method arises from the effect of the waste matrix as it attenuates the low-energy photons before they are counted. The nonuniform distribution of TRU in the waste also causes problems.

The remaining three methods (krypton and xenon production from spontaneous fission, prompt-gamma counting from spontaneous, and from induced fission) are not attractive, mainly because of low sensitivity. If the minimum detection level is raised to 1 $\mu\text{Ci/g}$, prompt-gamma counting from induced fission provides a method for measuring ^{233}U and ^{239}Pu .

Conclusions

The following conclusions on the measurement of small quantities of TRU in waste material using nondestructive testing techniques were reached as a result of this study.

- No simple method exists for measuring the quantity of a TRU in a package of waste in the presence of a gamma-ray background greater than a few milliRoentgen/hr.
- When an interfering background is not present, gamma-ray spectroscopy provides the best method for identifying and quantifying the TRU present in the waste. Several such systems have been developed and are in use

today. Major research efforts will continue toward the development of greater sensitivity and better resolution. However, improvements within the next few years are not expected to provide an increase in sensitivity to reduce the detection level of TRU by tenfold.

- Monitoring the production of helium from the alpha decay of the TRU is an untried method that has the potential to measure all TRU isotopes present in waste. The major technical difficulty with this method involves the naturally occurring helium in the air. However, detection sensitivity of the technique appears to be close to the 10-nCi/g level. The potential of this method should justify the necessary research effort to develop helium-production monitoring into a practical technique.
- The only other viable measurement method at the 10-nCi/g level involves complex procedure of spontaneous-fission neutron counting, neutron irradiation, delayed-neutron counting, and krypton-xenon sampling and analysis. The delayed-neutron counting is not required because it does not add new data. However, it does provide a confirmation of the krypton-xenon sampling results. Using the outlined procedure, ten TRU isotopes can be measured at the 10-nCi/g level including ²³⁹Pu. Elaborate counting and electronic circuits are required to operate the system as well as to collect and analyze the data. This method has not yet been demonstrated experimentally on small quantities of TRU and technical difficulties are more pronounced with this technique than with the other practical methods.
- Excluding the gamma-analysis method, neither of the other two proposed methods will identify the TRU isotopes present. In the neutron irradiation and counting as well as in the krypton-xenon sampling methods, a foreknowledge of the TRU isotopes present and their relative abundance is required ever to quantify the amount of TRU present. If this foreknowledge is unattainable, these methods are useless.
- Counting prompt neutrons from induced fission is not feasible, even when counting in coincidence pairs, because interference from the source neutrons completely masks the prompt-neutron count. No technique has been discovered to overcome this difficulty.
- Raising the minimum detection level to 1 µCi/g will permit the detection of all 27 TRU isotopes by the helium detection method and 19 of the

isotopes by gamma-ray spectroscopy (in the absence of an interfering background).

- If the minimum-detection level were raised to 1 $\mu\text{Ci/g}$, measurement of the prompt fission gammas would be feasible, even in the presence of a 1-R/hr gamma background; additional energy discrimination could be accomplished with shields. However, the isotopes thus detected can be measured at lower activity levels by other methods.
- Lowering the minimum-detection level to 1 nCi/g will make it impossible to measure all TRUs except the five isotopes having the highest spontaneous-fission rates (i.e., ^{244}Pu , ^{248}Cm , ^{250}Cm , ^{250}Cf , and ^{252}Cf).

It is our opinion that the helium-production measurement offers the most promise if gamma-ray analysis is not possible because of interfering background levels. From an economic standpoint, the helium method is very attractive. Data interpretation is simple and straightforward and can be carried out by a technician.

Acknowledgments

The author is indebted to those at LLL who answered questions, provided information and reference material, and offered suggestions. Particular thanks to Charles T. Prevo who investigated several counting and irradiation facility designs through neutron diffusion codes.

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Appendix A

Estimated Cost of a Neutron Irradiation and Counting Facility

These estimated costs for a shielded neutron irradiation and counting facility are based on 1976 labor and equipment costs.

SHIELDED FACILITIES

Structure, 10 ft wide × 15 ft long × 10 ft high with 2-ft reinforced concrete walls and floor	\$ 25,000
Moderator assembly	\$ 5,000

NEUTRON SOURCES

2 neutron generators, 190 KV at 2 mA providing 2×10^{11} n/sec	\$ 50,000
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COUNTING EQUIPMENT

36 neutron detectors	\$ 8,000
Electronics and supporting hardware	\$ 32,000

TOTAL ESTIMATED COSTS	\$ 120,000
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Appendix B

Suggested Neutron Counting Procedure for Krypton and Xenon

- (1) Place a small ^{252}Cf source at the bottom center of the empty neutron counting well. Record a 5-min count of coincidence pairs.
- (2) Insert the fiberboard waste drum into the well counter without disturbing the small ^{252}Cf source. Record a 5-min count of coincidence pairs.
- (3) Remove the small ^{252}Cf source without disturbing the waste drum. Record a 5-min count of coincidence pairs.
- (4) Bypass the coincidence gate and record a straight 5-min count of background from spontaneous-fission neutrons and (α, n) -reaction neutrons.
- (5) Turn on the neutron generators and irradiate waste drum with a flux density of $10^6 \text{ n/cm}^2\text{-sec}$ for at least 10 min.
- (6) Turn off neutron generators and immediately begin a time history of the neutron count rate until the level reaches the background determined in Step (4) (no longer than 15 min).
- (7) Remove the waste drum from the well counter and move to the krypton-xenon sampling equipment. Connect sampling system and collect krypton and xenon on the molecular sieve. Count the sieve after collection.
- (8) Analyze data.

Appendix C

Estimated Cost of a Helium-Production Measurement System

Two cost estimates are given. One is for the nitrogen-gas purge method of preparation. The other one is for a vacuum method of preparation. Estimates are based on 1976 labor and equipment costs.

NITROGEN GAS PURGE METHOD

Liquid nitrogen purge and gas transfer system*	\$ 5,000
Highly sensitive helium-leak detector	\$ 12,000
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TOTAL COST	\$ 17,000

VACUUM METHOD

Vacuum chamber, pumps, and supporting equipment	\$ 15,000
Highly sensitive helium-leak detector	\$ 12,000
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TOTAL COST	\$ 27,000

*Most facilities have large storage tanks of liquid nitrogen as supply for other uses in the plant. We assume here that the liquid nitrogen can be obtained from this storage. These costs include only the equipment necessary to transfer the liquid nitrogen to the waste area and the plumbing to purge the waste containers.

Appendix D
Estimated Cost of an Irradiation and Sample Collection Facility
for Krypton and Xenon Production

Estimates are based on 1976 labor and equipment costs.

SHIELDED FACILITIES

Structure, 10 ft wide × 15 ft long × 10 ft high with 2-ft walls and floor	\$ 25,000
Krypton-xenon monitoring facilities shielded from irradiation facilities (including plumbing, gas transfer, and sampling systems)	\$ 10,000
Moderator assembly	\$ 5,000

NEUTRON SOURCES

2 neutron generators, 190 kV at 2 mA providing 2×10^{11} n/sec	\$ 50,000
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ELECTRONICS

Completely automated photon counting, recording, and analysis system (including on-line minicomputer).	\$ 50,000
TOTAL COST	\$ 140,000

Appendix E

Estimated Cost of a Photon Counting Facility

Estimates are based on 1976 labor and equipment costs.

MECHANICAL HARDWARE

Automated drum scanning system with collimated detector shield	\$ 20,000
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ELECTRONICS

Completely automated photon counting, recording, and analysis system (including on-line minicomputer)	<u>\$ 50,000</u>
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TOTAL COST	\$ 70,000
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