8

Plutonium Isotopic Composition by Gamma-Ray Spectroscopy

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8.1 INTRODUCTION

An accurate measurement of plutonium isotopic composition is usually required to interpret the results of neutron coincidence or calorimetry measurements. Several methods have been developed for determining plutonium isotopic composition by gamma-ray spectroscopy; some of the early approaches are described in Refs. 1 through 5. An American Society for Testing and Materials standard test method has been written for plutonium isotopic analysis using gamma-ray spectroscopy (Ref. 6). Different methods have been developed for different sample types.

This chapter introduces the characteristics of plutonium spectra that influence isotopic measurements, describes useful spectral regions, and presents the principles of spectral analysis important to isotopic measurement. It includes descriptions of typical data collection hardware, details of data analysis methods, and descriptions of several implemented systems with examples of their accuracy and precision.

8.2 BACKGROUND

8.2.1 Decay Characteristics of Plutonium Isotopes

Most plutonium samples contain the isotopes ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, and ^{242}Pu. Americium-241 and ^{237}U are always present as decay products of ^{241}Pu. Table 8-1 lists some of the decay characteristics of these important isotopes.

8.2.2 Decay Characteristics of ^{241}Pu

The decay of ^{241}Pu is shown in Figure 8.1. Because of the long half-life of ^{241}Am, the concentration of the ^{241}Am daughter continues to increase for up to 75 yr. Aged plutonium samples often have very high ^{241}Am content, especially if the initial ^{241}Pu concentration was high.

Table 8-1. Decay characteristics for isotopes useful in plutonium isotopic measurements

Isotope	Half-Life (yr)	Activity (dis/s-g)	Specific Power (MW/g Isotope)
^{238}Pu	87.74 ± 0.04	6.3330×10^{11}	567.57 ± 0.026
^{239}Pu	24 119 ± 26	2.2942×10^{9}	1.9288 ± 0.0003
^{240}Pu	6564 ± 11	8.3971×10^{9}	7.0824 ± 0.0020
^{241}Pu	14.348 ± 0.022	3.8244×10^{12}	3.412 ± 0.002
^{242}Pu	376 300 ± 900	1.4522×10^{8}	0.1159 ± 0.0003
^{241}Am	433.6 ± 1.4	1.2655×10^{11}	114.20 ± 0.50
^{237}U	(6.75 days)	9.4080×10^{7} [a]	- - -

[a] Uranium-237 activity computed assuming ^{241}Pu-^{237}U equilibrium (see Figure 8.1). Alpha branching ratio of ^{241}Pu assumed to be 2.46×10^{-5}.

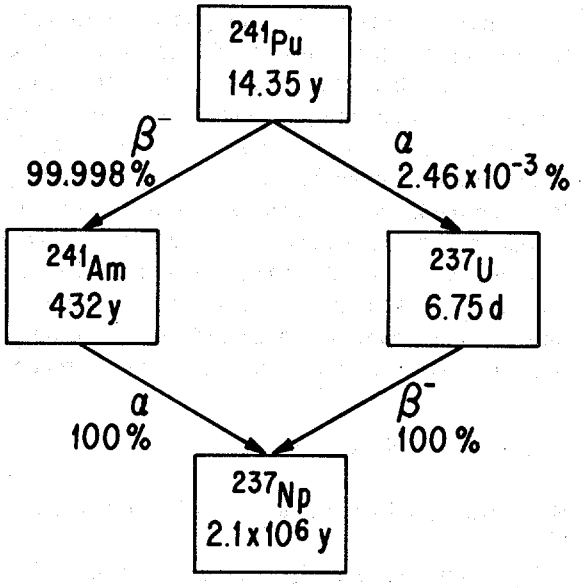

Fig. 8.1 Decay scheme of ^{241}Pu and its daughters.

Because of its short half-life, the ^{237}U daughter rapidly comes into secular equilibrium (Ref. 7) with its ^{241}Pu parent. After approximately 47 days (7 half-lives), gamma rays from the decay of ^{237}U can be used as a measure of ^{241}Pu. Because ^{237}U has several strong gamma rays, it is especially useful for plutonium isotopic measurements. In this chapter the terms ^{241}Pu-^{237}U equilibrium or aged refer to

samples where ^{237}U is in secular equilibrium with ^{241}Pu. Samples in which ^{241}Pu-^{237}U equilibrium does not exist are called freshly separated. For those samples, ^{237}U cannot be used as a measure of ^{241}Pu.

Figure 8.1 shows that both ^{241}Am and ^{237}U decay to the same isotope, ^{237}Np. Both isotopes can populate the same excited states in ^{237}Np and give rise to identical gamma rays. Thus, most of the useful ^{237}U gamma-ray peaks have a contribution from ^{241}Am. The amount of this contribution depends upon the particular gamma ray and the time since americium was last separated from the sample. Figure 8.2 shows the relative contributions for important ^{237}U gamma rays. A correction should be made to ^{241}Pu-^{237}U peaks for their ^{241}Am content.

8.2.3 Determination of ^{242}Pu Concentration

Plutonium-242 has only a few gamma rays, similar in energy and branching ratio to those from ^{240}Pu. However, the long half-life of ^{242}Pu and its low abundance in most plutonium make its detection by gamma-ray measurement impossible. Instead, empirical isotopic correlations (Ref. 8) are used to predict the ^{242}Pu content from the other isotopic fractions. Such predictions generally produce acceptable results for the concentration of ^{242}Pu (typically 0.03 to 5%) found in most plutonium, if process batches have not been mixed and ^{241}Am has neither been added nor removed.

8.2.4 Spectral Interferences

Many regions of the gamma-ray spectrum can contain interfering gamma rays from other isotopes in the sample. For example: very high burnup samples often contain ^{243}Am and its ^{239}Np daughter; aged samples may contain ^{237}Np and its daughter ^{233}Pa; and samples from reprocessed fuel may contain fission products. All the possible interferences cannot be listed here; however, by knowing the history of a sample, the spectroscopist can anticipate possible spectral interferences.

8.2.5 Applications of Plutonium Isotopic Measurements

The principal application of plutonium isotopic measurements is to support other nondestructive assay (NDA) methods in providing the total plutonium content of a sample. Two methods that use plutonium isotopic results are calorimetry and neutron coincidence counting.

Calorimetry uses the isotopic information to calculate the specific power P (W/g Pu) of a sample from the measured isotopic fractions and the known specific power for each isotope (see Chapters 21 and 22).

The response of neutron coincidence counters is a complicated function of all the plutonium isotopes and ^{241}Am. A measurement of isotopic composition is required to convert the coincidence counter response to plutonium mass (see Chapter 16).

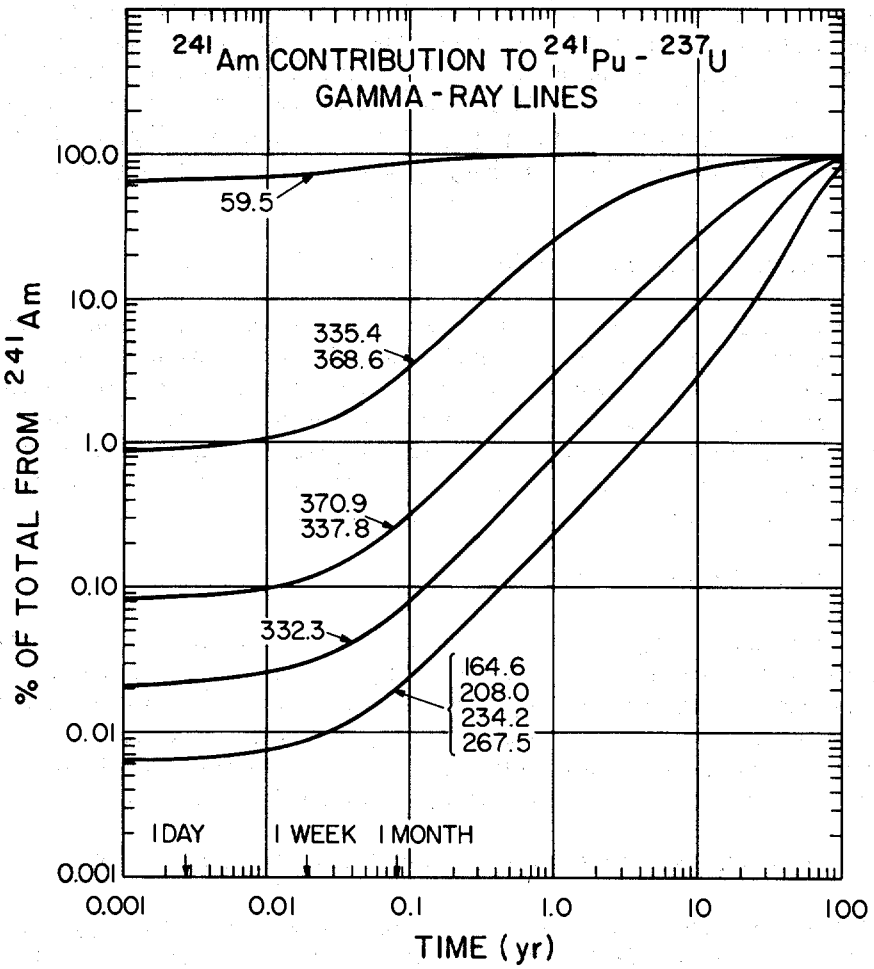

Fig. 8.2 Americium-241 contribution to ^{237}U gamma-ray peaks (energy in keV) as a function of time since separation. Uranium and americium concentrations are zero at t = 0.

8.3 SPECTRAL REGIONS USEFUL FOR ISOTOPIC MEASUREMENTS

This section describes the spectral features that are important for the measurement of plutonium isotopic composition. The descriptions follow the practice used in works of Gunnink et al. (Ref. 9) and Lemming and Rakel (Ref. 10) in which the spectrum is divided into several different regions. The gamma-ray spectrum of plutonium varies greatly with isotopic composition and ^{241}Am concentration. Two sample spectra are shown in Figures 8.3 and 8.4. Figure 8.3 represents low burnup and low ^{241}Am; Figure 8.4 represents intermediate burnup and relatively high ^{241}Am. Similar examples are given for each of the spectral regions to illustrate the variability that may be encountered.

Table 8-2 lists most of the gamma rays that are useful for plutonium isotopic measurements. The list shows that the lower energy gamma rays are more intense than those at higher energies. The lower energy gamma rays should be used whenever possible, however, it is often impossible to use them.

8.3.1 The 40-keV Region

The 40-keV region has been used mainly for analysis of freshly separated solutions from which ^{241}Am and ^{237}U have been removed. If too much ^{241}Am is present, its 60-keV gamma ray overwhelms all other peaks in the region. Usually, the 40-keV region is useful for 15 to 30 days after a separation of americium and uranium. A typical spectrum from a high-burnup reprocessing plant solution (Ref. 11) is shown in Figure 8.5; Table 8-3 lists the peak energies and intensities. When the gamma rays can be measured, the 40-keV region is the most useful region for measuring ^{238}Pu, ^{239}Pu, and ^{240}Pu. The region does not have a measurable ^{241}Pu gamma ray; ^{241}Pu concentration is usually measured from its 148.6-keV gamma ray. Small contributions from ^{241}Pu and ^{237}U interfere with the ^{238}Pu peak at 43.5 keV, the ^{240}Pu peak at 45.2 keV, and the ^{239}Pu peak at 51.6 keV.

Several experimenters have used this region for solution measurements: Gunnink (Ref. 12) and Russo (Ref. 11) have measured freshly separated solutions from a reprocessing plant; Umezawa (Ref. 13) and Bubernak (Ref. 14) have applied these measurements to samples prepared in an analytical laboratory; and Li (Ref. 15) has measured submilligram-sized solid samples with modest ^{241}Am content. Gunnink uses absolute counting techniques and calibrates with known solution standards; Umezawa uses absolute counting with a calibrated detector; Bubernak calibrates with samples of known isotopic composition; and Russo and Li measure isotopic ratios that are independent of calibration standards as discussed in Section 8.4.

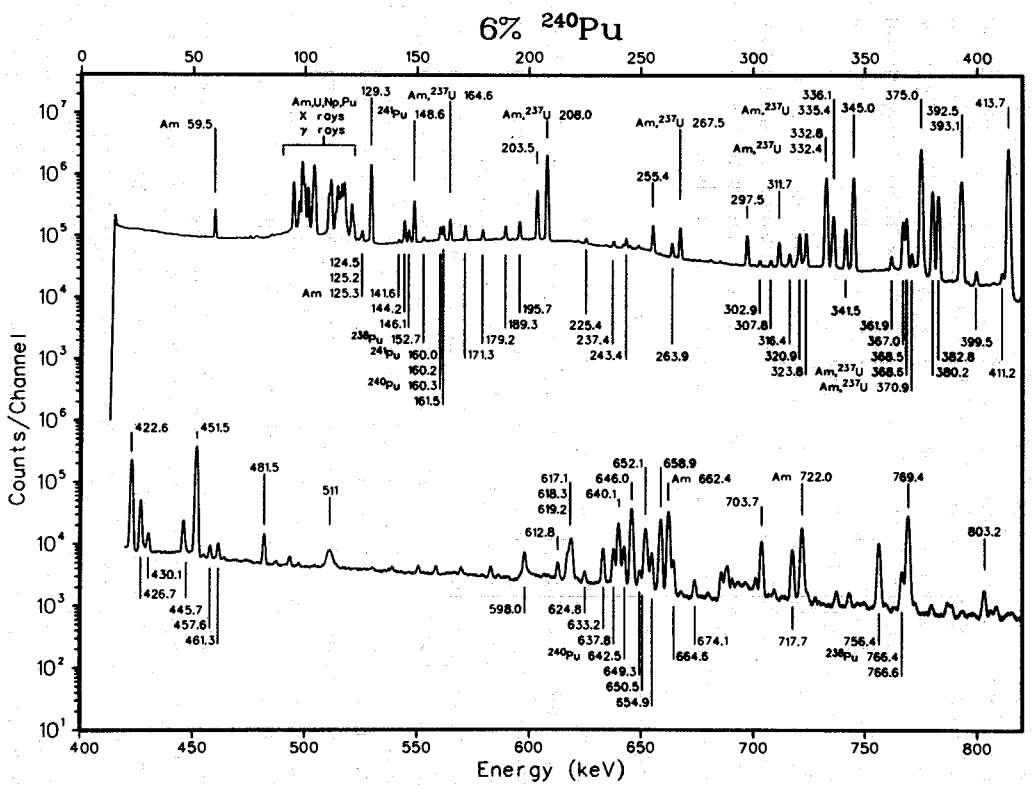

Fig. 8.3 *Gamma-ray spectrum from 500 g of plutonium metal measured with a coaxial germanium detector (at 1332 keV: relative efficiency = 11.7%, FWHM = 1.75 keV). Isotopic composition (wt%): 238, 0.012%; 239, 93.82%; 240, 5.90%; 241, 0.240%; 242, 0.02%; ^{241}Am, 630 μg/g plutonium. Peaks not labeled with a specific isotope are from ^{239}Pu.*

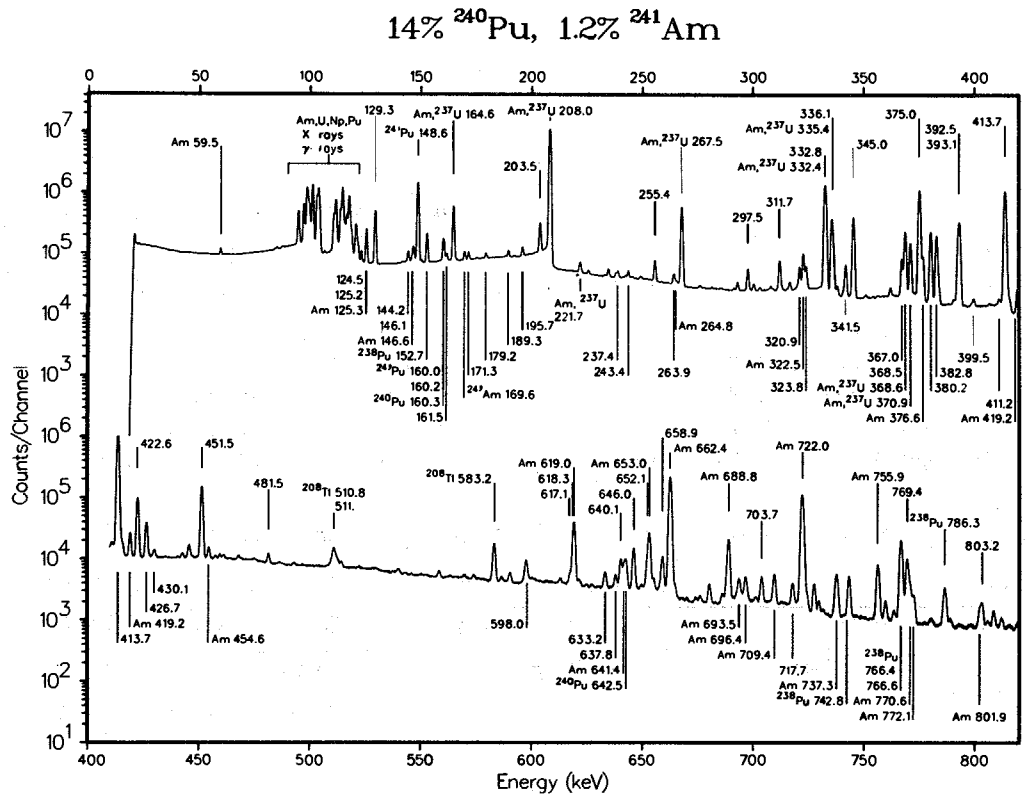

***Fig. 8.4** Gamma-ray spectrum from 530 g of plutonium as PuO_2 measured with a coaxial germanium detector (at 1332 keV: relative efficiency = 10.2%, FWHM = 1.65 keV). Isotopic composition (wt%): 238, 0.202%; 239, 82.49%; 240, 13.75%; 241, 2.69%; 242, 0.76%; ^{241}Am, 11 800 μg/g plutonium. Peaks not labeled with a specific isotope are from ^{239}Pu.*

Table 8-2. Useful gamma rays in various energy regions

Region	^{238}Pu		^{239}Pu		^{240}Pu		^{241}Pu		^{241}Am	
(keV)	(keV)	(γ/s-g)	(keV)	(γ/s-g)	(keV)	(γ/s-g)	(keV)	(γ/s-g)	(keV)	(γ/s-g)
40-60	43.48	2.49×10^{8}	51.63	6.19×10^{5}	45.23	3.80×10^{6}		- - -	59.54	4.54×10^{10}
90-105	99.86	4.59×10^{7}	98.78	2.80×10^{4}	104.24	5.86×10^{5}	103.68	3.86×10^{6}	98.95	2.57×10^{7}
									102.97	2.47×10^{7}
120-450	152.68	6.05×10^{6}	129.29	1.44×10^{5}	160.28	3.38×10^{4}	148.57	7.15×10^{6}	125.29	5.16×10^{6}
			203.54	1.28×10^{4}			164.58[a]	1.73×10^{6}	335.40	6.28×10^{5}
			345.01	1.28×10^{4}			208.00[a]	2.04×10^{7}		
			375.04	3.60×10^{4}			332.35[a]	1.14×10^{6}		
			413.71	3.42×10^{4}			370.93[a]	1.04×10^{5}		
450-800	766.41	1.39×10^{5}	645.97	3.42×10^{2}	642.48	1.05×10^{3}		- - -	662.42	4.61×10^{5}
			717.72	6.29×10^{1}					721.99	2.48×10^{5}

[a]Uranium-237 daughter of ^{241}Pu with ^{241}Pu-^{237}U equilibrium.

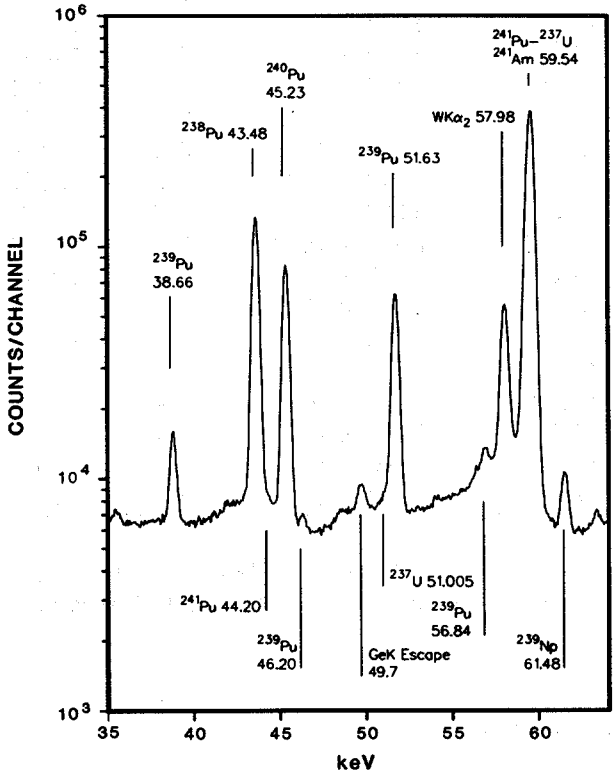

Fig. 8.5 *Gamma-ray spectrum in the 40-keV region from a freshly separated 185-g/L solution of plutonium in nitric acid. Isotopic composition (wt%): 238, 0.649%; 239, 67.01%; 240, 21.80%; 241, 8.11%; 242, 2.44%. The W $K_{\alpha 2}$ x ray arises from the shielding around the detector (Ref. 11).*

Table 8-3. Peak energies and intensities in 40-keV region[a]

Isotope	Energy (keV)	Branching Ratio (photons/dis)	Error (%)	Activity (photons/s-g)
^{239}Pu	38.664	1.050×10^{-4}	1.00	2.4089×10^{5}
^{239}Pu	40.410	1.620×10^{-6}	10.00	3.7165×10^{3}
^{239}Pu	42.060	1.650×10^{-6}	3.00	3.7854×10^{3}
^{237}U	43.430	5.904×10^{-9}	7.00	2.2579×10^{4} [b]
^{238}Pu	43.477	3.930×10^{-4}	0.30	2.4889×10^{8}
^{241}Pu	44.200	4.180×10^{-8}		1.5986×10^{5}
^{241}Pu	44.860	8.360×10^{-9}		3.1972×10^{4}
^{240}Pu	45.232	4.530×10^{-4}	0.20	3.8039×10^{6}
^{239}Pu	46.210	7.370×10^{-6}	10.00	1.6908×10^{4}
^{239}Pu	46.690	5.800×10^{-7}	6.00	1.3306×10^{3}
^{237}U	51.005	8.364×10^{-8}	2.00	3.1987×10^{5} [b]
^{239}Pu	51.629	2.700×10^{-4}	0.20	6.1942×10^{5}
^{239}Pu	54.040	2.000×10^{-6}	1.40	4.5883×10^{3}
^{241}Pu	56.320	2.500×10^{-8}		9.5610×10^{4}
^{239}Pu	56.760	9.750×10^{-9}		3.7288×10^{4}
^{239}Pu	56.838	1.130×10^{-5}	1.00	2.5924×10^{4}
^{237}U	59.536	8.487×10^{-6}	0.20	3.2458×10^{7} [b]
^{241}Am	59.536	3.590×10^{-1}		4.5432×10^{10}
^{237}U	64.832	3.198×10^{-7}	0.50	1.2230×10^{6} [b]

[a]Ref. 9.

[b]Uranium-237 activity computed assuming ^{241}Pu-^{237}U equilibrium. Uranium-237 branching ratio includes 2.46×10^{-5} ^{241}Pu alpha branch to ^{237}U.

8.3.2 The 100-keV Region

The 100-keV region is the most complex region of the gamma-ray spectrum of plutonium. Table 8-4 lists 14 gamma rays and x rays in this region. The uranium x rays arise from plutonium decay and can be used to measure the plutonium isotopes. The neptunium x rays arise from the decay of ^{241}Am and ^{237}U, and the plutonium x rays appear in larger or more concentrated samples from gamma-ray and alpha-particle-induced x-ray fluorescence. The 100-keV region is the only region in which gamma rays from all isotopes are present.

Table 8-4. Peak energies and intensities in 100-keV region[a]

Isotope	Energy (keV)	Branching Ratio (photons/dis)	Error (%)	Activity (photons/s-g)	X Ray
^{238}Pu	94.658	1.050×10^{-6}	1.40	6.6497×10^{5}	U $K_{\alpha 2}$
^{239}Pu	94.658	4.220×10^{-5}	0.25	9.6813×10^{4}	U $K_{\alpha 2}$
^{240}Pu	94.658	6.360×10^{-7}	5.00	5.3406×10^{3}	U $K_{\alpha 2}$
^{241}Pu	94.658	3.030×10^{-6}	0.50	1.1588×10^{7}	U $K_{\alpha 2}$
^{239}Pu	96.130	2.230×10^{-7}	20.00	5.1160×10^{2}	
^{237}U	97.071	3.887×10^{-6}	0.40	1.4865×10^{7}	Np $K_{\alpha 2}$[b]
^{241}Am	97.072	1.180×10^{-5}	2.00	1.4933×10^{6}	Np $K_{\alpha 2}$
^{238}Pu	98.441	1.690×10^{-6}	1.00	1.0703×10^{6}	U $K_{\alpha 1}$
^{239}Pu	98.441	6.760×10^{-5}	0.30	1.5508×10^{5}	U $K_{\alpha 1}$
^{240}Pu	98.441	1.020×10^{-6}	5.00	8.5651×10^{3}	U $K_{\alpha 1}$
^{241}Pu	98.441	4.850×10^{-6}	0.50	1.8548×10^{7}	U $K_{\alpha 1}$
^{239}Pu	98.780	1.220×10^{-5}	3.00	2.7989×10^{4}	
^{241}Am	98.951	2.030×10^{-4}	0.50	2.5690×10^{7}	
Pu[c]	99.530				Pu $K_{\alpha 2}$
^{238}Pu	99.864	7.240×10^{-5}	0.20	4.5851×10^{7}	
^{237}U	101.066	6.199×10^{-6}	0.30	2.3708×10^{7}	Np $K_{\alpha 1}$[b]
^{241}Am	101.066	1.900×10^{-5}	1.40	2.4045×10^{6}	Np $K_{\alpha 1}$
^{241}Am	102.966	1.950×10^{-4}	0.50	2.4677×10^{7}	
^{239}Pu	103.020	2.170×10^{-6}	1.60	4.9783×10^{3}	
^{241}Pu	103.680	1.010×10^{-6}	0.50	3.8627×10^{6}	
Pu[c]	103.748				Pu $K_{\alpha 1}$
^{240}Pu	104.244	6.980×10^{-5}	0.40	5.8612×10^{5}	

[a]Ref. 9.

[b]Uranium-237 activity computed assuming ^{241}Pu-^{237}U equilibrium. Uranium-237 branching ratio includes 2.46×10^{-5} ^{241}Pu alpha branch to ^{237}U.

[c]Plutonium x rays are produced in high-mass or high-concentration samples by gamma-ray- and alpha-particle-induced x-ray fluorescence.

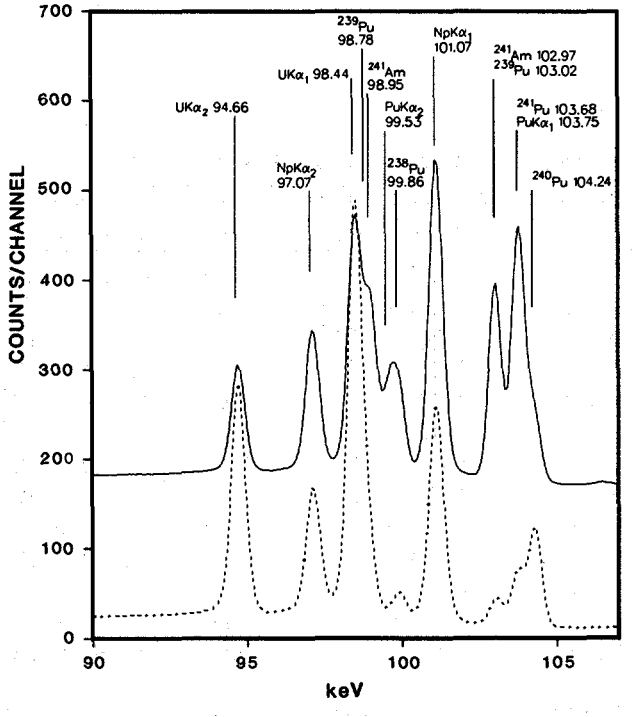

Fig. 8.6 Gamma-ray spectrum in the 100-keV region measured with a HPGe detector with resolution of 490 eV at 122 keV. Solid line: 530 g of plutonium as PuO_2. Isotopic composition (wt%): 238, 0.302%; 239, 82.49%; 240, 13.75%; 241, 2.69%; 242, 0.76%; ^{241}Am, 11 800 μg/g plutonium. Dashed line: 10 mL of 20-g/L solution of plutonium in 1M HNO_3. Isotopic composition (wt%): 238, 0.027%; 239, 91.65%; 240, 7.68%; 241, 0.532%; 242; 0.12%; ^{241}Am, 315 μg/g plutonium.

The strong, overlapping, interfering nature of the spectrum in the 100-keV region is shown in Figure 8.6. The entire isotopic distribution may be determined from this region for low-burnup solution samples (Ref. 4); a spectrum from such a sample is shown at the bottom of Figure 8.6. The complexity of the region requires peak-fitting or response-function (Refs. 4 and 16) analysis. Low-burnup PuO_2 has also been analyzed using response-function methods. High-burnup solutions have been analyzed using response-function methods in this region (Ref. 12), but such analysis is difficult. The increased alpha and gamma-ray activity of high-burnup and high-concentration solutions fluoresces plutonium x rays at 99.53 and 103.75 keV. These

x rays interfere with the gamma-ray peaks at 99.86 keV from ^{238}Pu and 104.24 keV from ^{240}Pu. The intrinsic line shape of x rays is different from that of gamma rays; this difference must be considered in the analysis of this region by response-function methods.

8.3.3 The 125-keV Region

The 125-keV region is used to measure ^{241}Am and ^{239}Pu from gamma rays at 125.29 and 129.29 keV. There are strong interferences to the 125.29-keV ^{241}Am gamma ray from ^{239}Pu lines at 125.21 and 124.51 keV. The ^{241}Am peak area is difficult to measure for ^{241}Am concentrations below a few hundred micrograms per gram of plutonium. For an americium concentration of 500 μg/g Pu, the ^{239}Pu interference contributes over 50% of the 125.3-keV peak area; for an americium concentration of 5000 μg/g Pu, over 90% of the peak area is from ^{241}Am. The ^{239}Pu interference can be removed by peak fitting (Ref. 17) or stripping (Ref. 18). Many measurements use absorbers or filters (see Section 8.5.3) to reduce the count rate of lower energy gamma rays. These absorbers affect the count rate in the 125-keV region. A 0.15-cm-thick cadmium filter transmits only 35% of the incident gamma rays at 125.3 keV.

Table 8-5 lists the gamma-ray energies and intensities in the 125-keV region. Figure 8.7 shows this spectral region; the plutonium K_β x rays at 116.3, 117.3, and 120.6 keV (omitted from Table 8-5) complicate the analysis of this region by making it difficult to find an interference-free background region below 124.5 keV. High-burnup material (solid line, Figure 8.7) generally gives a stronger ^{241}Am peak but a weaker ^{239}Pu peak than low-burnup material does. For high-burnup material, the precision of the ^{239}Pu peak at 129.29 keV is worse because there is less ^{239}Pu in the sample and because the background continuum under the peak is usually higher as a result of the intense ^{241}Pu and ^{237}U gamma rays at higher energies.

Table 8-5. Peak energies and intensities in 125-keV region[a]

Isotope	Energy (keV)	Branching Ratio (photons/dis)	Error (%)	Activity (photons/s-g)
^{239}Pu	119.708	3.000×10^{-7}	2.00	6.8825×10^{2}
^{241}Pu	121.200	6.850×10^{-9}		2.6197×10^{4}
^{241}Am	122.994	1.000×10^{-5}	0.80	1.2655×10^{6}
^{239}Pu	123.620	1.970×10^{-7}	6.00	4.5195×10^{2}
^{239}Pu	124.510	6.130×10^{-7}	3.00	1.4063×10^{3}
^{239}Pu	125.210	7.110×10^{-7}	2.00	1.6311×10^{3}
^{241}Am	125.292	4.080×10^{-5}	0.50	5.1633×10^{6}
^{239}Pu	129.294	6.260×10^{-5}	0.20	1.4361×10^{5}

[a]Ref. 9.

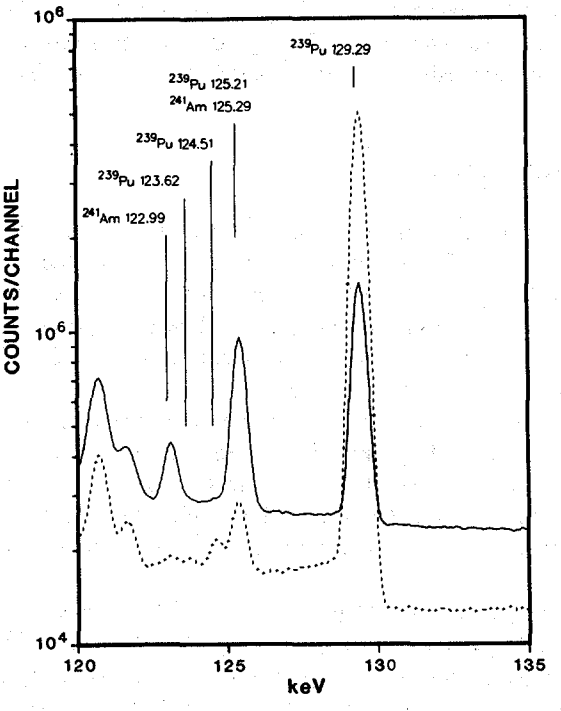

Fig. 8.7 Gamma-ray spectrum of PuO_2 in the 125-keV region measured with a HPGe detector with resolution of 490 eV at 122 keV. Isotopic compositions (wt%): (solid line) 238, 0.378%; 239, 78.89%; 240, 15.28%; 241, 4.42%; 242, 1.04%; ^{241}Am, 14 300 μg/g plutonium; and (dashed line) 238, 0.016%; 239, 93.51%; 240, 6.15%, 241, 0.28%; 242, 0.039%; ^{241}Am, 480 μg/g plutonium.

8.3.4 The 148-keV Region

The two most important peaks in the 148-keV region are the 148.57-keV ^{241}Pu peak and the 152.68-keV ^{238}Pu peak. The 148.57-keV peak is the only useful gamma ray outside of the complex 100-keV region that comes directly from ^{241}Pu. The 152.68-keV peak, although weak, is often the only useful gamma ray from ^{238}Pu above 100 keV. (In some high-burnup samples, the 766.4-keV peak can be used for ^{238}Pu measurement.) The 144.21- and 143.35-keV ^{239}Pu lines are often used in isotopic composition measurements; however, in mixed-oxide samples the ^{235}U gamma ray at 143.76 keV can be an interference. Americium-241 peaks at 146.56 and 150.11 keV complicate window settings and background determination for methods that use simple channel summation procedures to determine peak areas. An additional interference

can arise in systems that use a ^{109}Cd reference source (Refs. 11 and 12). Here, the 88.04-keV ^{109}Cd gamma ray can sum with the 59.54-keV ^{241}Am gamma ray to produce a pileup peak at 147.6 keV. Other sum peaks can interfere with the 152.68-keV ^{238}Pu peak (U $K_{\alpha 2}$ at 94.66 keV plus ^{241}Am at 59.54 keV equals 154.2 keV).

The weak 153-keV ^{238}Pu peak is usually on a high background continuum and yields a poor precision for low-burnup (0.01 wt% ^{238}Pu) material. The precision can be as poor as 10% for typical measurements. It is widely recognized that the branching ratio of the 153-keV gamma ray is approximately 2.5% lower than the value cited in Table 8-6. Branching ratio biases are discussed in Ref. 19. Figure 8.8 shows a plot of the peaks in this region from high- and low-burnup material.

Table 8-6. Peak energies and intensities in 148-keV region[a]

Isotope	Energy (keV)	Branching Ratio (photons/dis)	Error (%)	Activity (photons/s-g)
^{239}Pu	141.657	3.200×10^{-7}	2.00	7.3413×10^{2}
^{239}Pu	143.350	1.730×10^{-7}	4.00	3.9689×10^{2}
^{239}Pu	144.211	2.830×10^{-6}	0.60	6.4925×10^{3}
^{239}Pu	146.077	1.190×10^{-6}	0.60	2.7300×10^{3}
^{241}Am	146.557	4.610×10^{-6}	1.00	5.8340×10^{5}
^{241}Pu	148.567	1.870×10^{-6}	0.30	7.1516×10^{6}
^{241}Am	150.110	7.400×10^{-7}	2.00	9.3648×10^{4}
^{238}Pu	152.680	9.560×10^{-6}	0.50	6.0544×10^{6}

[a]Ref. 9.

8.3.5 The 160-keV Region

For a single detector system not using the 40- or 100-keV region, the 160-keV complex is the only one that can be used for measuring ^{240}Pu. Table 8-7 lists the energies and intensities of the major gamma rays in the 160-keV region. The ^{240}Pu gamma ray at 160.28 keV has strong interferences from ^{241}Pu at 159.95 keV and ^{239}Pu at 160.19 keV. This three-peak complex (Figure 8.9) is only partially resolved from the 161.45-keV ^{239}Pu line. Peak-fitting, peak-stripping, or response-function methods must be used to isolate the ^{240}Pu intensity. The statistical precision of the ^{240}Pu component is seldom measured to better than ~2%.

The intensity of the entire complex increases with increasing burnup, but the fractional contribution from ^{240}Pu decreases. Table 8-8 illustrates how the relative fraction of ^{240}Pu in the 160-keV complex decreases from almost 70% for material with 6% ^{240}Pu to 25% for material with 20% ^{240}Pu. This decrease occurs because ^{241}Pu increases more rapidly than ^{240}Pu as burnup increases.

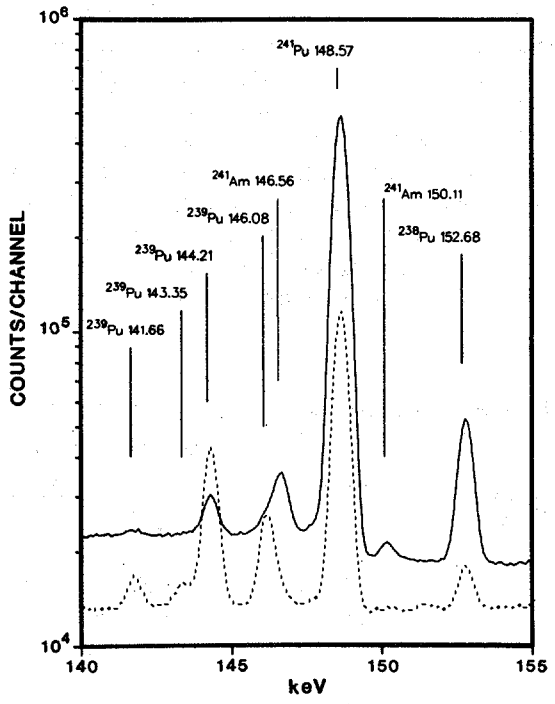

Fig. 8.8 Gamma-ray spectrum of PuO_2 in the 148-keV region measured with a HPGe detector with resolution of 490 eV at 122 keV. Isotopic compositions (wt%): (solid line) 238, 0.378%; 239, 78.89%; 240, 15.28%; 241, 4.42%; 242, 1.04%; ^{241}Am, 14 300 μg/g plutonium; and (dashed line) 238, 0.016%; 239, 93.51%; 240, 6.15%; 241, 0.28%; 242, 0.039%; ^{241}Am, 480 μg/g plutonium.

Table 8-7. Peak energies and intensities in 160-keV region[a]

Isotope	Energy (keV)	Branching Ratio (photons/dis)	Error (%)	Activity (photons/s-g)
^{239}Pu	158.100	1.000×10^{-8}	10.00	2.2942×10^{1}
^{241}Pu	159.955	6.740×10^{-8}		2.5776×10^{5}
^{239}Pu	160.190	6.200×10^{-8}	20.00	1.4224×10^{2}
^{240}Pu	160.280	4.020×10^{-6}	0.70	3.3756×10^{4}
^{239}Pu	161.450	1.200×10^{-6}	0.40	2.7530×10^{3}
^{237}U	164.580	4.526×10^{-7}	0.50	1.7311×10^{6} [b]
^{241}Am	164.580	6.670×10^{-7}	3.00	8.4410×10^{4}
^{241}Am	165.930	2.320×10^{-7}	4.00	2.9360×10^{4}

[a]Ref. 9.

[b]Uranium-237 activity computed assuming ^{241}Pu-^{237}U equilibrium. Uranium-237 branching ratio includes 2.46×10^{-5} ^{241}Pu alpha branch to ^{237}U.

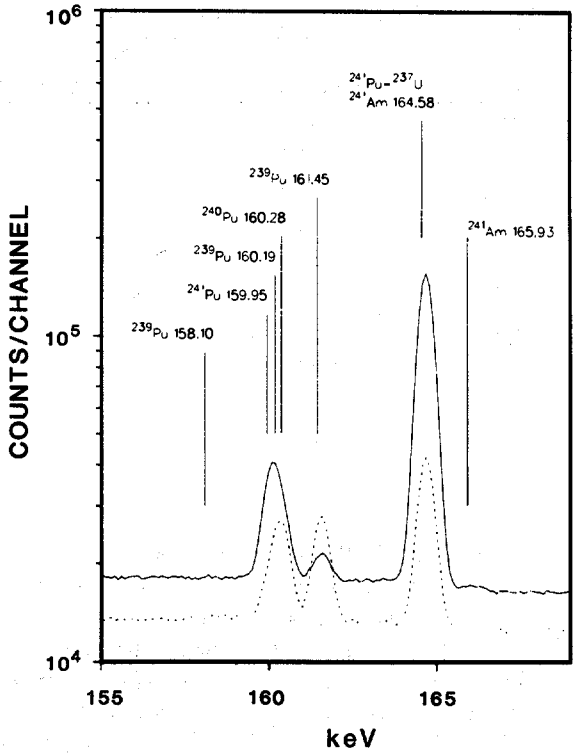

Fig. 8.9 *Gamma-ray spectrum of PuO_2 in the 160-keV region measured with a HPGe detector with resolution of 490 eV at 122 keV. Isotopic compositions (wt%): (solid line) 238, 0.378%; 239, 78.89%; 240, 15.28%; 241, 4.42%; 242, 1.04%; ^{241}Am, 14 300 μg/g plutonium; and (dashed line) 238, 0.016%; 239, 93.51%; 240, 6.15%; 241, 0.28%; 242, 0.039%; ^{241}Am 480 μg/g plutonium.*

Because the 164.58-keV peak comes from the ^{237}U daughter of ^{241}Pu, it can be used for ^{241}Pu only after equilibrium has been attained. A correction is often made for the ^{241}Am contribution to the 164.58-keV peak. The summing of x rays and gamma rays from the 100-keV region with the 59.5-keV ^{241}Am gamma ray can interfere with the 160-keV complex. These sum peaks can be eliminated by using a filter to selectively absorb gamma rays and x rays from the 60- and 100-keV regions before they interact with the detector (Ref. 20). In mixed-oxide samples, the ^{235}U gamma ray at 163.35 keV can interfere with the 164.58-keV peak if the ratio of ^{235}U to ^{241}Pu is greater than approximately 1.5.

Table 8-8. Components of 160-keV complex

Isotope	Weight Percent	Percentage of 160 Complex from Indicated Isotope
239	93.5	4.5
240	6.0	69.1
241	0.3	26.4
239	86.0	1.5
240	12.0	50.4
241	1.5	48.1
239	67.0	0.3
240	20.0	24.6
241	8.0	75.1

8.3.6 The 208-keV Region

The strong ^{241}Pu-^{237}U peak at 208.00 keV dominates the 208-keV region. Usually it is the most intense peak in the spectrum. It has a contribution from ^{241}Am that becomes approximately 1% (relative) after 4 yr. Because this gamma-ray comes from ^{237}U, it can be used only for aged samples. Its strength and freedom from interference make it suitable for shape and energy calibrations for analysis methods using peak fitting or response functions. For mixed oxides, ^{235}U peaks at 202.1 and 205.3 keV can interfere with the 203.54-keV ^{239}Pu line. The ^{239}Pu/^{241}Pu ratio formed with the 203.54/208.00 line pair gives best results for low-burnup material. For high-burnup material, the precision of the 203.54-keV ^{239}Pu peak becomes worse because of the Compton background and the long tail from the very strong 208.00-keV ^{237}U peak. High ^{241}Am (~1%) can cause a few tenths of a percent interference with the 203.5-keV peak; however, the effects of this interference can be removed easily. Table 8-9 and Figure 8.10 list and display the parameters and spectral features of the 208-keV region.

8.3.7 The 332-keV Region

The 332-keV region has contributions from ^{241}Pu-^{237}U, ^{241}Am, and ^{239}Pu as shown in Table 8-10 and Figure 8.11. For high-burnup aged material the ratio of the 345.01-keV ^{239}Pu peak to the 332.35-keV ^{241}Pu-^{237}U peak is useful for measuring the ^{239}Pu/^{241}Pu ratio. Both the 332.35- and 335.40-keV peaks from ^{241}Pu-^{237}U contain very close interferences from ^{239}Pu peaks. In Figure 8.11, the plot of a very low-burnup sample (98% ^{239}Pu) illustrates how close these two interferences are. Table 8-11 gives the relative magnitudes of these ^{239}Pu interferences for different isotopic compositions. After the ^{239}Pu interferences are removed, the two peak complexes are

left with both ^{241}Pu-^{237}U and ^{241}Am components that can be used to measure the ^{241}Pu/^{241}Am ratio (Ref. 3). Peak fitting, peak stripping, or response functions are required to analyze this region.

Table 8-9. Peak energies and intensities in 208-keV region[a]

Isotope	Energy (keV)	Branching Ratio (photons/dis)	Error (%)	Activity (photons/s-g)
^{239}Pu	203.537	5.600×10^{-6}	0.20	1.2847×10^{4}
^{241}Am	203.870	2.900×10^{-8}	6.00	3.6700×10^{3}
^{237}U	208.000	5.338×10^{-6}	0.20	2.0415×10^{7} [b]
^{241}Am	208.000	7.910×10^{-6}	0.50	1.0010×10^{6}

[a]Ref. 9.

[b]Uranium-237 activity computed assuming ^{241}Pu-^{237}U equilibrium. Uranium-237 branching ratio includes 2.46×10^{-5} ^{241}Pu alpha branch to ^{237}U.

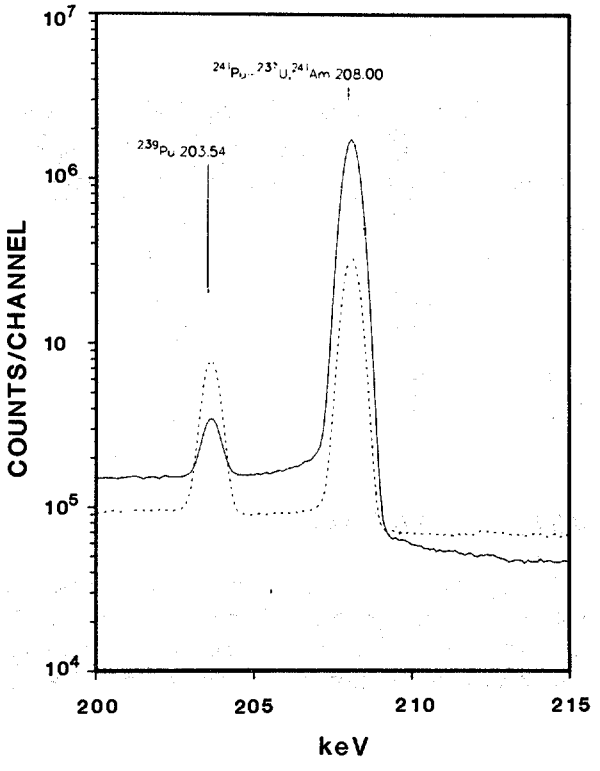

Fig. 8.10 *Gamma-ray spectrum of PuO_2 in the 208-keV region measured with a HPGe detector with resolution of 490 eV at 122 keV. Isotopic compositions (wt%): (solid line) 238, 0.378%; 239, 78.89%; 240, 15.28%; 241, 4.42%; 242, 1.04%; ^{241}Am, 14 300 μg/g plutonium; and (dashed line) 238, 0.016%; 239, 93.51%; 240, 6.15%; 241, 0.28%; 242, 0.039%; ^{241}Am, 480 μg/g plutonium.*

Table 8-10. Peak energies and intensities in 332-keV region[a]

Isotope	Energy (keV)	Branching Ratio (photons/dis)	Error (%)	Activity (photons/s-g)
^{237}U	332.354	2.977×10^{-7}	0.30	1.1384×10^{6} [b]
^{241}Am	332.354	1.490×10^{-6}	0.30	1.8856×10^{5}
^{239}Pu	332.838	5.060×10^{-6}	0.20	1.1608×10^{4}
^{237}U	335.405	2.386×10^{-8}	1.00	9.1258×10^{4} [b]
^{241}Am	335.405	4.960×10^{-6}	0.30	6.2769×10^{5}
^{239}Pu	336.107	1.134×10^{-6}	0.30	2.6016×10^{3}
^{241}Am	337.720	4.290×10^{-8}	5.00	5.4290×10^{3}
^{237}U	337.720	2.189×10^{-9}	5.00	8.3732×10^{3} [b]
^{237}U	340.450	4.059×10^{-10}	20.00	1.5523×10^{3} [b]
^{239}Pu	341.510	6.620×10^{-7}	0.40	1.5187×10^{3}
^{239}Pu	345.014	5.592×10^{-6}	0.20	1.2829×10^{4}

[a]Ref. 9.

[b]Uranium-237 activity computed assuming ^{241}Pu-^{237}U equilibrium. Uranium-237 branching ratio includes 2.46×10^{-5} ^{241}Pu alpha branch to ^{237}U.

Table 8-11. Components of 332 and 336 complexes (^{241}Am neglected)

Isotope	Weight Percent	Percentage of 332 Complex from Indicated Isotope	Percentage of 336 Complex from Indicated Isotope
239	93.5	76.1	89.9
241	0.3	23.9	10.1
239	86.0	36.9	62
241	1.5	63.1	38
239	67.0	7.9	19.3
241	8.0	92.1	80.7

8.3.8 The 375-keV Region

The 375-keV region, shown in Figure 8.12 and Table 8-12, has components from the same isotopes as the 332-keV region: ^{241}Pu-^{237}U, ^{241}Am, and ^{239}Pu. For all isotopes except ^{239}Pu, the branching ratios are lower than in the 332-keV region, so the isotopic information will be less precise. The strong 375.04-keV ^{239}Pu peak is

often used for relative efficiency determination. The ^{241}Am interference at 376.59 keV becomes bothersome above concentrations of a few thousand micrograms per gram of plutonium.

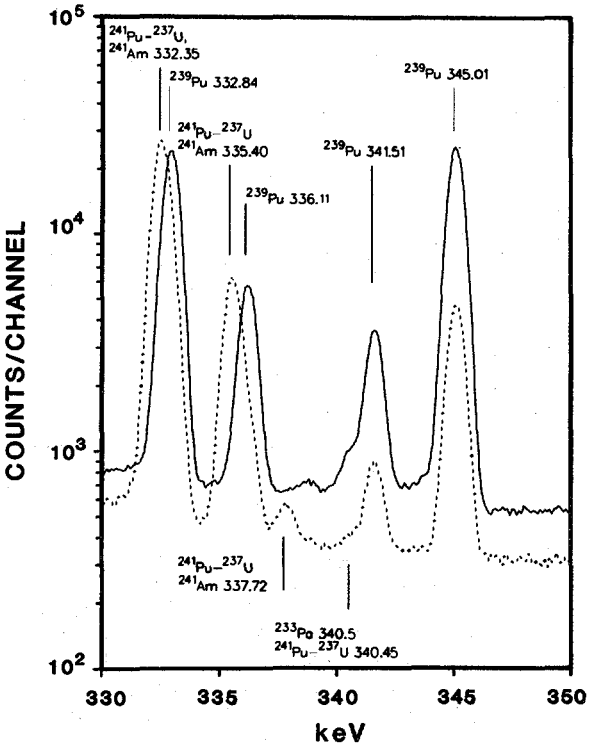

Fig. 8.11 Gamma-ray spectrum of PuO_2 in the 332-keV region measured with a HPGe detector with resolution of 490 eV at 122 keV. Isotopic compositions (wt%): (solid line) 238, 0.0024%; 239, 97.96%; 240, 2.01%; 241, 0.020%; 242, 0.0030%; ^{241}Am, 11 μg/g plutonium; and (dashed line) 238, 0.378%; 239, 78.89%; 240, 15.28%; 241, 4.42%; 242, 1.04%; ^{241}Am, 14 300 μg/g plutonium.

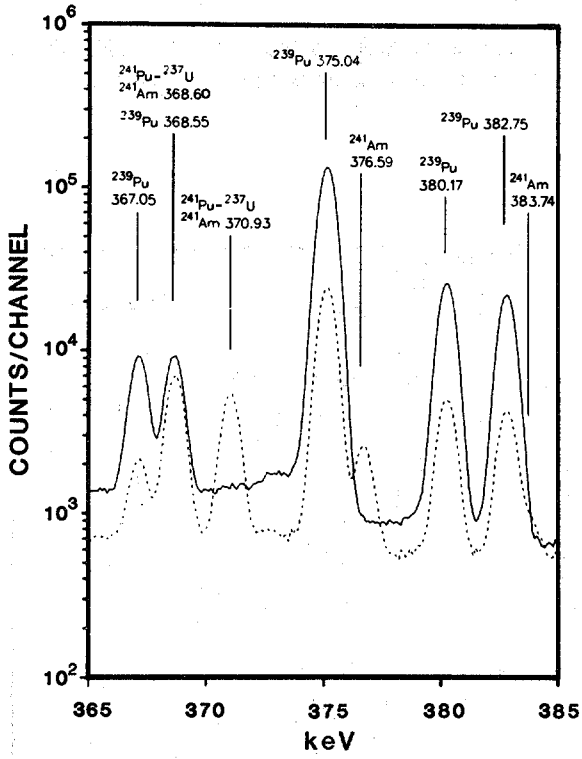

Fig. 8.12 Gamma-ray spectrum of PuO_2 in the 375-keV region measured with a HPGe detector with resolution of 490 eV at 122 keV. Isotopic compositions (wt%): (solid line) 238, 0.0024%; 239, 97.96%; 240, 2.01%; 241, 0.020%; 242, 0.0030%; ^{241}Am, 11 μg/g plutonium; and (dashed line) 238, 0.378%; 239, 78.89%; 240, 15.28%; 241, 4.42%; 242, 1.04%; ^{241}Am, 14 300 μg/g plutonium.

8.3.9 The 640-keV Region

Figure 8.13 and Table 8-13 show the characteristics of the 640-keV region. This is the only region above 160 keV that can be used for measuring ^{240}Pu. The region is useful only for large samples because of the low intensity of the 642.48-keV ^{240}Pu gamma ray. Nearby peaks from ^{239}Pu and ^{241}Am complicate the region. The 645.97- and 662.42-keV peaks are useful for measuring ^{239}Pu and ^{241}Am. The other gamma rays in the region are not generally used but must be considered for peak-fitting or response-function analysis.

Table 8-12. Peak energies and intensities in 375-keV region[a]

Isotope	Energy (keV)	Branching Ratio (photons/dis)	Error (%)	Activity (photons/s-g)
^{239}Pu	367.050	8.650×10^{-7}	0.30	1.9844×10^{3}
^{239}Pu	368.550	9.030×10^{-7}	0.30	2.0716×10^{3}
^{237}U	368.605	1.055×10^{-8}	2.00	4.0360×10^{4} [b]
^{241}Am	368.605	2.170×10^{-6}	0.30	2.7462×10^{5}
^{237}U	370.934	2.713×10^{-8}	1.40	1.0377×10^{5} [b]
^{241}Am	370.934	5.230×10^{-7}	0.80	6.6186×10^{4}
^{239}Pu	375.042	1.570×10^{-5}	0.10	3.6018×10^{4}
^{241}Am	376.595	1.383×10^{-6}	0.70	1.7502×10^{5}
^{239}Pu	380.166	3.051×10^{-6}	0.20	6.9995×10^{3}
^{239}Pu	382.751	2.587×10^{-6}	0.20	5.9350×10^{3}
^{241}Am	383.740	2.820×10^{-7}	1.50	3.5687×10^{4}

[a]Ref. 9.

[b]Uranium-237 activity computed assuming ^{241}Pu-^{237}U equilibrium. Uranium-237 branching ratio includes 2.46×10^{-5} ^{241}Pu alpha branch to ^{237}U.

A large-volume coaxial detector (10% relative efficiency or greater) should be used in the 640-keV region; analysis schemes that also analyze data from the 100 to 400-keV region should use two detectors (Ref. 20).

If fission products such as ^{95}Zr-^{95}Nb and ^{137}Cs are present in the sample, their gamma rays will complicate the analysis of the 640-keV region. A fission product concentration as low as 10 μCi/g Pu can make the analysis of this region impossible.

8.4 MEASUREMENT PRINCIPLES

8.4.1 Measurement of Isotopic Ratios

The photopeak area for any single gamma ray can be written as

$$C(E_j^i) = \lambda^i N^i BR_j^i \varepsilon(E_j) \tag{8-1}$$

where $C(E_j^i)$ = photopeak area of gamma ray j with energy E_j emitted from isotope i

λ^i = decay constant of isotope i ($\lambda^i = \ln 2/T_{1/2}^i$, where $T_{1/2}^i$ is the half-life of isotope i)

N^i = number of atoms of isotope i

BR_j^i = branching ratio (gamma rays/disintegration) of gamma ray j from isotope i

$\varepsilon(E_j)$ = total efficiency for photopeak detection of gamma ray with energy E_j. Includes detector efficiency, geometry, sample self-absorption, and attenuation in materials between the sample and detector.

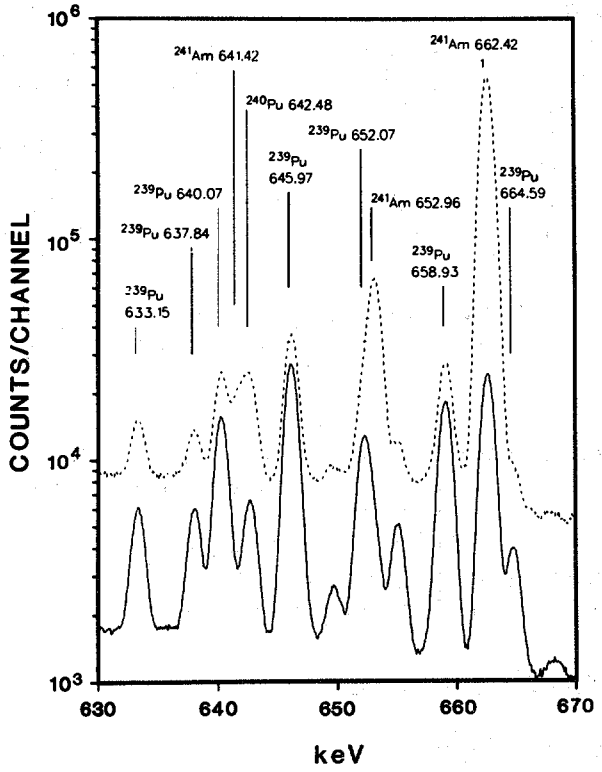

Fig. 8.13 Gamma-ray spectrum in the 640-keV region. Dashed line: 530 g of plutonium as PuO_2 measured with a coaxial HPGe detector (at 1332 keV: efficiency = 10.2%, FWHM = 1.65 keV). Isotopic composition (wt%): 238, 0.302%; 239, 82.49%; 240, 13.75%; 241, 2.69%; 242, 0.76%; ^{241}Am, 11 800 μg/g plutonium. Solid line: 500 g of plutonium metal measured with a coaxial HPGe detector (at 1332 keV: efficiency = 11.7%, FWHM = 1.75 keV). Isotopic composition (wt%): 238, 0.012%; 239, 93.82%; 240, 5.90%; 241, 0.240%; 242, 0.028%; ^{241}Am, 630 μg/g plutonium.

The photopeak area can be written also in terms of the mass of the isotope as

$$C(E_j^i) = \gamma_j^i \, M_i \, \varepsilon(E_j) \tag{8-2}$$

where γ_j^i = photon emission rate of gamma ray j from isotope i in γ/s-g
M_i = mass of isotope i (g) .

Table 8-13. Peak energies and intensities in 640-keV region[a]

Isotope	Energy (keV)	Branching Ratio (photons/dis)	Error (%)	Activity (photons/s-g)
^{241}Am	633.000	1.260×10^{-8}	15.00	1.5945×10^{3}
^{239}Pu	633.150	2.530×10^{-8}	1.20	5.8042×10^{1}
^{239}Pu	637.837	2.560×10^{-8}	1.20	5.8730×10^{1}
^{239}Pu	640.075	8.200×10^{-8}	0.60	1.8812×10^{2}
^{241}Am	641.420	7.100×10^{-8}	4.00	8.9851×10^{3}
^{240}Pu	642.480	1.245×10^{-7}	1.00	1.0454×10^{3}
^{239}Pu	645.969	1.489×10^{-7}	0.40	3.4160×10^{2}
^{239}Pu	649.321	7.120×10^{-9}	7.00	1.6334×10^{1}
^{239}Pu	650.529	2.700×10^{-9}	15.00	6.1942×10^{0}
^{239}Pu	652.074	6.550×10^{-8}	0.60	1.5027×10^{2}
^{241}Am	652.960	3.770×10^{-7}	2.00	4.7710×10^{4}
^{239}Pu	654.880	2.250×10^{-8}	1.20	5.1618×10^{1}
^{239}Pu	658.929	9.690×10^{-8}	0.70	2.2230×10^{2}
^{241}Am	662.420	3.640×10^{-6}	0.30	4.6065×10^{5}
^{239}Pu	664.587	1.657×10^{-8}	1.60	3.8014×10^{1}
^{239}Pu	668.200	3.930×10^{-10}	30.00	9.0160×10^{-1}

[a]Ref. 9.

These two equations may be rearranged to give expressions for the atom and mass ratios of two isotopes. The atom ratio is given by

$$\frac{N^i}{N^k} = \frac{C(E_j^i)}{C(E_m^k)} \times \frac{T_{1/2}^i}{T_{1/2}^k} \times \frac{BR_m^k}{BR_j^i} \times \frac{RE(E_m)}{RE(E_j)} \, . \tag{8-3}$$

The similar expression for the mass ratio is

$$\frac{M^i}{M^k} = \frac{C(E_j^i)}{C(E_m^k)} \times \frac{\gamma_m^k}{\gamma_j^i} \times \frac{RE(E_m)}{RE(E_j)} \, . \tag{8-4}$$

In Equations 8-3 and 8-4, the photopeak areas C(E) are measured and the half-lives $T_{1/2}$, branching ratios BR, and photon emission rates γ are either known or can be calculated from nuclear data. The total efficiency has been expressed in terms of

the relative efficiency RE. Geometry factors cancel and the relative efficiency ratio includes only sample self-absorption, attenuation in materials between the sample and detector, and detector efficiency. The use of an efficiency ratio removes the need for reproducible geometry and makes the isotopic ratio method applicable to samples of arbitrary size, shape, and composition.

A relative efficiency curve can be determined from the measured spectrum of every sample. Equations 8-1 and 8-2 give the following proportionality for gamma rays from a single isotope *i*:

$$\varepsilon(E_j) \propto RE(E_j) \propto \frac{C(E_j^i)}{BR_j^i} \propto \frac{C(E_j^i)}{\gamma_j^i}. \tag{8-5}$$

Because efficiency ratios are used in Equations 8-3 and 8-4, only the shape of the relative efficiency curve is important; either of the ratios given in Equation 8-5 can be used. The peak areas C(E) of strong, interference-free gamma rays from a single isotope are used to define the relative efficiency curve. Curve-fitting (Ref. 17) or interpolation techniques are used to define the relative efficiency at energies between the measured points. To better define the shape of the relative efficiency curve, points from more than one isotope can be used by normalizing one isotope to another (Refs. 17 and 18). Gamma rays from ^{239}Pu and ^{241}Pu-^{237}U are most often used to define the relative efficiency curve in the range from 130 to 450 keV. Figure 8.14 shows two examples of measured relative efficiency curves.

It is advantageous to use closely spaced gamma rays to measure isotopic ratios because the relevant relative efficiency ratios will be near unity. However, the relative efficiency correction must be applied even for closely spaced lines in the 120- to 200-keV region. A typical correction for the 152.7-keV/148.6-keV ratio (^{238}Pu/^{241}Pu) can be 10%.

After appropriate isotopic ratios are measured, it is usually desirable to calculate absolute isotopic fractions. The sum of all isotopic fractions must equal unity. Neglecting ^{242}Pu, this implies that

$$1 = f_{238} + f_{239} + f_{240} + f_{241} \tag{8-6}$$

where f_i is the isotopic fraction of isotope *i*.

Dividing Equation 8-6 by f_{241} gives

$$\frac{1}{f_{241}} = \frac{f_{238}}{f_{241}} + \frac{f_{239}}{f_{241}} + \frac{f_{240}}{f_{241}} + 1 . \tag{8-7}$$

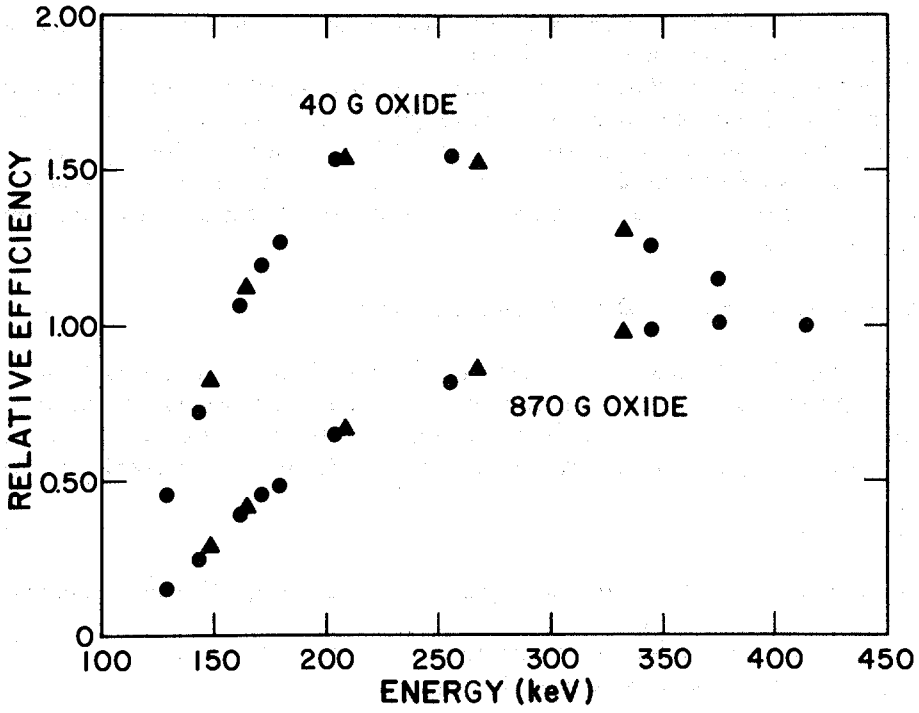

Fig. 8.14 Typical relative efficiency curves for a 200-mm² by 10-mm-deep planar HPGe detector (^{239}Pu peaks are indicated with solid circles and ^{241}Pu-^{237}U peaks with solid triangles). Both curves are normalized to 1.00 at 414 keV. The curve obtained for the 870-g PuO_2 sample shows the effect of higher attenuation.

Equation 8-7 expresses the isotopic fraction of ^{241}Pu (f_{241}) in terms of the three measured ratios f_{238}/f_{241}, f_{239}/f_{241}, and f_{240}/f_{241}. The remainder of the isotopic fractions are obtained from

$$f_i = f_{241} \times \left[\frac{f_i}{f_{241}}\right] \quad i = 238, 239, 240 . \tag{8-8}$$

Section 8.5.3 discusses the incorporation of ^{242}Pu into this analysis. If the ratio of ^{241}Am to any of the plutonium isotopes (usually ^{239}Pu) has been measured, then the absolute fraction of ^{241}Am can be calculated from

$$f_{Am} = f_i \times \frac{f_{Am}}{f_i} . \tag{8-9}$$

Note that this formula gives the weight or atom fraction of ^{241}Am in the sample with respect to total plutonium, not total sample.

The isotopic ratio method may be applied to samples of arbitrary size, shape, and composition. The method works as long as the plutonium isotopic composition and the Am/Pu ratio are uniform throughout the sample. The plutonium may be nonuniformly distributed as long as the above uniformity is present. A method was developed at Mound Laboratory to measure electrorefining salt residues that have a nonuniform Am/Pu ratio (Ref. 21).

8.4.2 Measurement of Absolute Isotopic Mass

Although the ratio method discussed in Section 8.4.1 can be applied to arbitrary samples, a more specialized method can be used to measure samples with reproducible geometries. The absolute measurement of isotope mass has been used by Gunnink and coworkers (Refs. 4, 12, and 16) for solution samples. This method uses the equation

$$C(E_j^i) = K_j^i m^i \tag{8-10}$$

where $C(E_j^i)$ = photopeak area of gamma ray j with energy E_j emitted from isotope i

K_j^i = calibration constant for gamma ray j from isotope i

m^i = mass of isotope i in sample.

Reference standards that have the same geometry as the unknown samples are used to determine the calibration constants. Self-attenuation corrections may be needed to account for differences between calibration standards and unknowns. The size and shape of the samples are carefully chosen to minimize (but not eliminate) self-absorption corrections. Given the mass of each isotope in the sample, the isotopic fractions are obtained from

$$f_i = \frac{m^i}{\sum_{i=238}^{241} m^i} . \tag{8-11}$$

8.4.3 The ^{242}Pu Isotopic Correlation

Plutonium-242 cannot be measured directly because of its low activity, low abundance, and weak gamma rays. Instead, isotopic correlation techniques (Ref. 8) are used to predict the ^{242}Pu abundance from knowledge of the other isotopic fractions. It is well known that correlations exist among the plutonium isotopic abundances because of the nature of the neutron capture reactions that produce the plutonium

isotopes in nuclear reactors. Because these correlations depend upon the reactor type and details of the irradiation history, it is difficult, if not impossible, to find a single correlation that is optimum for all material. Gunnink (Ref. 8) suggests that the correlation

$$242 = \frac{K(240)(241)}{(239)^2} \quad (\text{where } 242 = f_{242}) \tag{8-12}$$

is linear and relatively independent of reactor type. When the isotopic fractions are given in weight percent, the constant K equals 52. One disadvantage to this correlation is that it depends on ^{241}Pu, which decreases in absolute abundance by about 5% per year. The correlation works best if the ^{241}Pu abundance can be corrected to the time of fuel discharge from the reactor. When the discharge time is not known, a partial correction can be made by adding the quantities of ^{241}Am to the ^{241}Pu before computing the correlation. The total gives the ^{241}Pu content at the time of the last chemical separation.

A correlation not involving ^{241}Pu has been suggested (Refs. 8 and 22):

$$242 = \frac{K(240)^3}{(239)^2} \, . \tag{8-13}$$

This correlation is linear for a given reactor type, but the slope K depends on reactor type.

After the isotopic fraction of ^{242}Pu has been determined using a suitable correlation, known value, or stream average, the other isotopic fractions should be corrected using

$$f_i^c = f_i(1 - f_{242}) \tag{8-14}$$

where f_i^c are the normalized isotopic fractions including ^{242}Pu. This correction renormalizes the fractions so that the sum over all the plutonium isotopes equals unity.

8.5 DATA ACQUISITION

8.5.1 Electronics

A detailed discussion of the instrumentation used for gamma-ray spectroscopy is presented in Chapter 4. Plutonium isotopic measurement systems use conventional, high-quality, nuclear instrumentation modules (NIM). Digital gain and zero-point stabilization are required for methods using channel-summation or response-function methods to evaluate peak areas. A multichannel analyzer (MCA) with a 4096-channel memory is used for most applications. Systems that use two detectors require two analog-to-digital converters and an 8192-channel memory.

The extensive data analysis requirements dictate that the MCA be interfaced to a computer. A 16-bit minicomputer with a 32-kword memory is adequate. A disk is needed for program and data file storage. Simple analysis can be done with programmable calculators after peak areas are obtained.

8.5.2 Detectors

All data analysis methods benefit from the use of a detector that gives the best possible resolution and peak shape. These parameters are most important when selecting a detector for a plutonium isotopic system. A planar, high-purity-germanium detector is used in most applications. A detector with a front surface area of 200 mm^2 and a thickness of 10 to 13 mm gives a good trade-off between resolution and efficiency. Such detectors are available commercially with a resolution (full width at half maximum) better than 500 eV at 122 keV. A peak-shape specification of 2.55 or better for the ratio of full width at one-fiftieth maximum to full width at half maximum at 122 keV helps ensure good peak shape. Good detectors give values of 2.5 or below for this parameter. The low efficiency of planar detectors restricts their use to regions below 400 keV. High-quality coaxial detectors can be used in the 100- to 400-keV region, but their lower resolution complicates the analysis of partially resolved peaks using channel-summation methods.

A coaxial detector with a relative efficiency of 10% or higher is required for measurements in the 600-keV region. Again, resolution is important. The very best resolution may negate the need to peak fit the entire 600-keV region (Ref. 17). Resolutions of 1.7 keV or better at 1332 keV are available.

8.5.3 Filters

Filters must be used in nearly all situations to reduce the count rate from the 59.54-keV ^{241}Am gamma ray that dominates the unfiltered spectrum from any aged sample. If the detector is unfiltered, the americium peak will cause unnecessary deadtime and will sum with x rays and gamma rays in the 100-keV region to produce interferences in the 150- to 165-keV region. Typical filters use 0.15 to 0.30 cm of cadmium and 0.025 cm of copper to selectively absorb the 59.54-keV gamma ray. A reasonable rule of thumb is to design the filter to reduce the 60-keV peak height to just less than the peak heights in the 100-keV region. A thicker filter will unnecessarily reduce the intensity of the important plutonium peaks in the 120- to 200-keV area (see Section 8.3.3). A further test for an adequate filter is to check that the region between 153 and 160 keV is flat and contains no sum peaks (Ref. 20). A more complete discussion of filter design is given in Chapter 2.

Little, if any, filtering is needed for freshly separated samples (no ^{241}Am or ^{237}U) when using the 100-keV region or the 40-keV region. If the detector is shielded with lead, the shield is often lined with approximately 0.25 cm of cadmium to suppress lead x rays (72 to 87 keV) that would otherwise appear in the spectrum.

8.5.4 Count Rate and Sample/Detector Geometry

The sample-to-detector distance or detector collimation is varied to achieve the desired count rate. The count rate is usually kept below 20 000 counts/s to maximize resolution. Developments in high-count-rate spectroscopy (Refs. 23 and 24) may soon allow the use of a much higher count rate. Count rates as high as 60 000 counts/s have been documented for plutonium isotopic measurements (Ref. 25).

Solution measurements use a fixed sample geometry and a disposable vial or a refillable cell. The sample thickness is chosen to optimize the measurement with respect to concentration and energy considerations.

If small samples are placed too close to the detector, gamma rays emitted in cascade may cause coincidence sum peaks in the spectrum because of the large solid angle subtended by the detector (Ref. 19). An example of this effect is the coincidence summing of the 129- and 203-keV gamma rays from ^{239}Pu; the sum peaks can interfere with the 332-keV complex from ^{241}Pu-^{237}U. An effect of 1.6% has been noted in the 332-keV region using a planar detector and a sample-to-detector distance of 3 to 4 cm. Large samples are generally placed at a greater distance from the detector, making this effect less important.

Large plutonium samples have high neutron emission rates; 1 kg of plutonium emits 1 to 2 $\times$ 10^5 n/s. High neutron exposure is known to damage germanium detectors and degrade detector resolution. It is difficult to minimize this effect, because, as the sample-to-detector distance is increased, the count time must be increased and the neutron dose remains essentially constant.

8.5.5 Count Time

The count time required to produce the desired precision is a function of the spectral region studied. In the 40- and 100-keV regions, count times of 1000 s to 1 h are usually satisfactory. Count times of 1 or 2 h or longer are often necessary when using gamma rays above 120 keV to measure high-mass samples, although in some situations samples as small as 10 g can be measured to better than 1% in less than 30 min. Small samples (1 to 2 g or less) may require overnight measurement times. For large samples, simple verification of the ^{239}Pu/^{241}Pu ratio may take only a few minutes. Some specific examples are discussed in Section 8.7.

8.6 SPECTRAL ANALYSIS

This section discusses spectral analysis techniques used for the measurement of plutonium isotopic composition. A general and more complete discussion of the methods used to determine photopeak areas is given in Section 5.3.

8.6.1 Region-of-Interest Summation

Region-of-interest (ROI)-summation or channel-summation techniques are often used to determine photopeak areas for plutonium isotopic composition measurements because they are easy to implement, understand, and use. Both linear and smoothed-step-function backgrounds are used. ROI-summation techniques work well to determine the areas of single isolated gamma-ray peaks but are less satisfactory for the analysis of overlapping peaks such as those found in the 125-, 160-, 332-, and 375-keV regions of the plutonium spectrum. When ROI summation is used to obtain the total area of a multiplet, the individual components can be isolated by an integral stripping method using neighboring peaks and known relative efficiency differences. This analysis generally leads to a loss of precision.

Electronic spectrum stabilizers are often included in systems that use ROI-summation techniques. The background windows must be chosen carefully so that they do not include any of the numerous weak plutonium or ^{241}Am gamma-ray peaks. This is particularly critical when measuring materials with high americium concentration.

8.6.2 Peak Fitting

A detailed description of peak-fitting techniques is given in Section 5.3. The techniques developed by Gunnink and coworkers at Livermore (Ref. 26) are widely used for both plutonium isotopic measurements and general gamma-ray spectroscopy. The GRPANL program (Refs. 27 and 28) was developed by Gunnink specifically to analyze the multiple peaks of the plutonium spectrum; it also forms the basis for the area determination routines of the GRPAUT program (Ref. 17) used at Mound Laboratory.

Both GRPANL and GRPAUT use a smoothed-step-function background and a Gaussian function with an exponential tail to describe the photopeak. The equation for the photopeak function is

$$Y_i = Y_0\{\exp[\alpha (X_i - X_0)^2] + T(X_i)\} \tag{8-15}$$

where Y_i = net counts in channel X_i for a single peak
Y_0 = peak amplitude
α = $-4 \ln 2/(\text{FWHM})^2 = 1/2\sigma^2$ where σ is the standard deviation of the Gaussian function
X_0 = peak centroid
$T(X_i)$ = tailing function at channel X_i.

The tailing function is given by

$$T(X_i) = \{A \exp[B(X_i - X_0)] + C \exp[D(X_i - X_0)]\} \times \{1 - \exp[0.4\alpha(X_i - X_0)^2]\}\delta \tag{8-16}$$

where A and C = short- and long-term tailing amplitude
B and D = short- and long-term tailing slope
δ = 1 for $X_i < X_0$
δ = 0 for $X_i \geq X_0$.

The second term brings the tailing function smoothly to zero at X_0 as shown in Figure 8.15. For many applications the long-term tail can be neglected (C = 0); for large multiplets with strong peaks, it should be included.

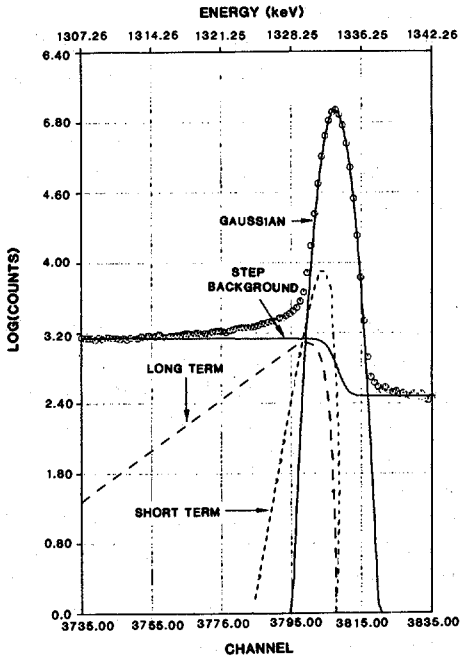

Fig. 8.15 *Gamma-ray photopeak obtained with Ge(Li) detector showing (1) Gaussian, (2) short-term tailing, (3) long-term tailing, and (4) smoothed-step background contributions to spectral peak shape (Ref. 28).*

When all seven parameters in Equations 8-15 and 8-16 are treated as free parameters, the peak-fitting process is usually slow, although today's computers often permit a sufficiently fast analysis. Fortunately, many of the parameters can be predetermined (see Section 5.3). The peak positions X_0 and width parameters α can be determined from two strong, isolated, reference peaks such as the 148- and 208-keV peaks from ^{241}Pu and ^{237}U. Because the gamma-ray branching ratios are well known, the relative intensities of peaks from the same isotope can be fixed.

Experience (Ref. 26) shows that the short-term tailing slope B is constant for a given detector system and should be measured for a high-energy peak where tailing is large. The short-term tailing amplitude A is given by

$$\ln A = k_1 + k_2 E \, . \qquad (8\text{-}17)$$

After B has been fixed, A can be determined from the two peaks that were used to determine the peak positions and width parameters.

If the long-term tailing is zero, the only free parameters are the peak amplitudes Y_0, and the fitting procedure is relatively fast. GRPANL allows other parameters to be free, but this increases the analysis time. The step-function background is determined first and subtracted from the acquired spectrum. GRPANL uses an iterative, nonlinear least-squares technique (Refs. 26 and 27) to fit the residual photopeak activity. Because the method is iterative, the analysis time depends on the number of peaks, the number of free parameters, and the type of computer system. Typically, analysis of a plutonium spectrum containing over 50 peaks in 15 groups in the 120- to 450-keV range takes about 10 min on a Digital Equipment Corporation PDP-11/23 computer or 3 to 4 min on a PDP-11/73. The analysis time is usually much shorter than the data accumulation time.

GRPANL can fit x-ray peaks that have a different intrinsic line shape (Lorentzian) than gamma rays (Ref. 29). This feature is necessary to fit peaks in the 100-keV region.

8.6.3 Response-Function Analysis

Response-function analysis uses the principles discussed in Section 8.6.2 to calculate the shape of the detector response to a particular isotope in a particular energy region. The peak-fitting procedure assigns a separate term with the form of Equation 8-15 to each photopeak in the analysis region and allows some or all of the shape parameters to be free. The response-function analysis uses the same equation but fixes all shape parameters and the relative amplitudes Y_0 of all the peaks from the same isotope; the only free parameters in the fitting procedure are the amplitudes of the isotopes that contribute peaks to the analysis region. The fitting procedure is reduced to a linear least-squares analysis that can be quickly solved.

The peak-shape characteristics of the detector must be known or determined from the spectrum of the sample under study. If the parameters are determined directly from each spectrum, variations that are due to different count rates or instrumental changes are automatically registered. (The procedure used to determine peak positions and shape parameters is discussed in Sections 5.3 and 8.6.2.) Given the shape parameters and positions for all gamma-ray peaks, it is easy to compute the response profile of each isotope in the analysis region. Response-function analysis has been used to fit the complex 100-keV region (Refs. 4, 12, and 16) and the many regions between 120 and 370 keV (Refs. 30 and 31). The formalism of Ref. 29 should be used to compute x-ray line shapes when analyzing the 100-keV region.

8.7 IMPLEMENTED SYSTEMS

This section discusses the characteristics and performance of four plutonium isotopic measurement systems that are in routine use in the United States.

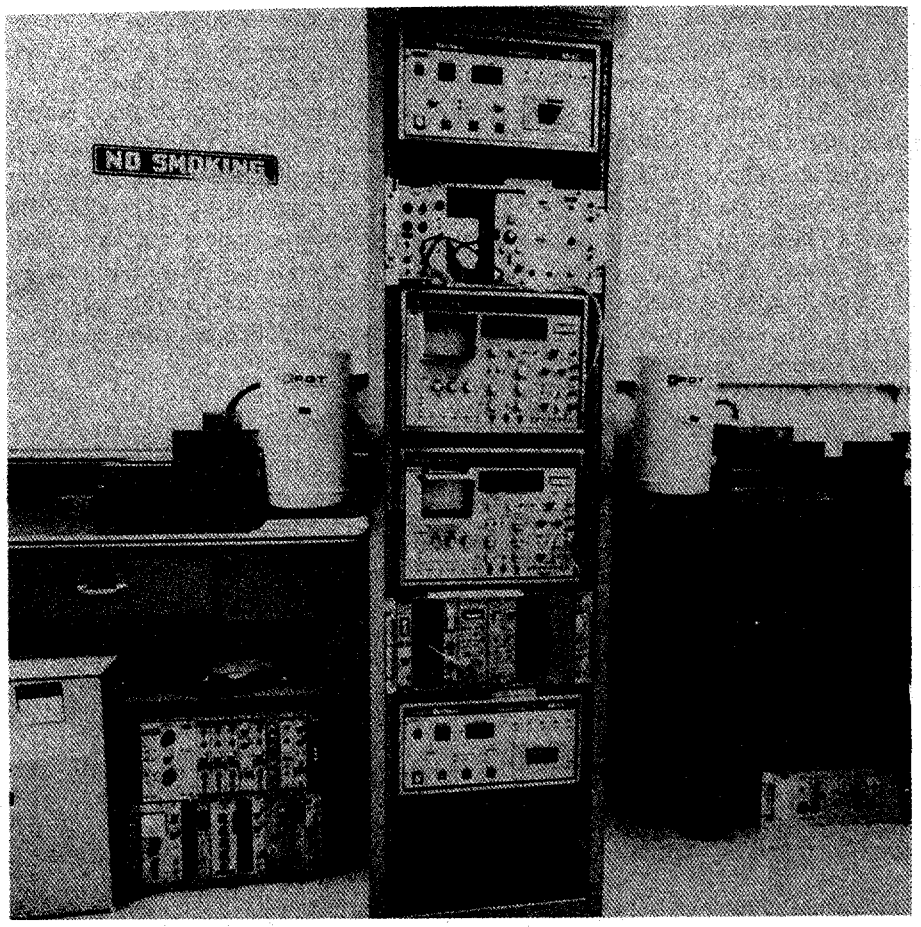

Fig. 8.16 *Plutonium isotopic system implemented at Rockwell-Hanford. (Photo courtesy of R. A. Hamilton, Rockwell-Hanford.)*

8.7.1 Rockwell-Hanford Company

The Rockwell-Hanford Company uses a plutonium isotopic measurement system in conjunction with a calorimeter to measure the plutonium content of solid samples of plutonium oxide, metal, mixed oxide, impure oxide, and scrap (Ref. 32). The system described here was used until 1984; the GRPAUT program, developed at Mound, is now used. The system, shown in Figure 8.16, uses four 300-mm^2 by 7-mm-deep planar HPGe detectors. The sample-to-detector distance is adjusted to give a count rate of 3000 counts/s; samples are counted for 10 000 s. No sample rotation is used. Spectra acquired in the MCA are analyzed in an off-line computer. Selected peaks

in the 120- to 400-keV range are analyzed using the isotope ratio method outlined in Ref. 3. Only the fundamental branching ratios and half-lives are used in the isotope ratio expressions. Bias corrections are not made and ^{242}Pu is not calculated.

Table 8-14 shows part of the large volume of performance data obtained during measurement of 14 standards that span the range from 2 to 24% ^{240}Pu. In Table 8-14 the measured isotope fractions have been used to calculate the rate of heat production in each sample; the definition of specific power and its importance to calorimetry are given in Chapter 21. The measurement precision for the specific power is 0.5 to 1.0% and the bias is of the same order. When applied to determining the specific power, plutonium isotopic measurement by gamma-ray spectroscopy is somewhat forgiving. Biases in one isotope fraction are partially cancelled by the normalization condition that all isotopes must sum to unity.

Table 8-14. Performance of Rockwell-Hanford isotopic system for specific power

	Isotope Fraction (wt%)						
Isotope	1(oxide)	2(metal)	3(oxide)	4(oxide)	5(oxide)	6(metal)	7(metal)
^{238}Pu	0.0003	0.0008	0.028	0.14	0.064	0.069	0.089
^{239}Pu	97.56	93.73	91.64	87.87	86.50	80.77	73.81
^{240}Pu	2.40	6.03	7.65	10.23	11.78	17.10	22.83
^{241}Pu	0.038	0.21	0.569	1.49	1.42	1.66	2.26
^{241}Am	0.059	0.138	0.447	1.26	0.088	1.12	2.13
No. of measurements							
	102	103	102	109	98	103	101
Precision of specific power (% RSD) calculated from measurement reproducibility							
	1.02%	0.72%	0.65%	0.55%	0.84%	0.62%	0.53%
Bias: Specific power from NDA divided by specific power from mass spectrometry values							
	0.9914	0.9921	1.003	1.008	1.016	1.002	1.028

8.7.2 Los Alamos National Laboratory

The Plutonium Processing Facility at Los Alamos has an isotopic composition measurement system that uses an isotopic ratio technique (Refs. 3, 18, and 33). Spectra are acquired with 200-mm^2 by 10-mm-deep planar HPGe detectors. The sample-to-detector distance is adjusted to give the desired count rate that may be as high as 20 000 counts/s. Digital gain stabilization is used so that ROI-summation techniques can be used to evaluate peak areas. The spectral regions between 120 and 400 keV are analyzed to determine isotopic ratios. All isotopic ratios are measured relative to ^{241}Pu. Different gamma-ray ratios are used for aged and freshly separated material; Table 8-15 lists the ratios used in the analysis. Equation 8-12 is used to

estimate ^{242}Pu; a correlation constant of 90 is used for materials with a wide range of reactor histories. The required relative efficiency ratios are determined from a set of strong, clean ^{239}Pu and ^{241}Pu-^{237}U lines that are normalized to each other. Simple linear and quadratic interpolation and extrapolation are used to obtain needed ratios. On-line analysis has been applied to material with 2 to 18% ^{240}Pu and up to 2.0% ^{241}Am. The system components are shown in Figure 8.17; the two detectors can acquire data from two samples simultaneously. The existing analysis program can handle up to four detectors.

Table 8-15. Ratios used in Los Alamos plutonium isotopic system

Isotopic	Gamma-Ray Energy of Samples (keV)	
Ratio	Aged	Freshly Separated
238/241	152.7/148.6	152.7/148.6
239/241[a]	345.0/332.4	129.3/148.6
	203.5/208.0	
240/241	160.3/164.6	160.3/148.6
Am/239[a]	125.3/129.3	125.3/129.3[b]
	169.6/171.3	

[a]A weighted average of the two ratios is used.
[b]The ^{241}Am is usually too low to measure.

Because ROI-summation techniques cannot separate overlapping peaks, clean neighboring peaks are used to subtract interferences. Table 8-16 lists the gamma-ray peaks used for this subtraction. The ^{241}Pu/^{241}Am ratio is determined from the 332-keV region; this ratio is used to subtract the ^{241}Am contribution from the ^{241}Pu-^{237}U peaks at 164.6, 208.0, 267.5, and 332.4 keV.

The fundamental constants in each isotopic ratio equation are adjusted slightly by measuring standard reference materials. This procedure compensates for possible biases in the measured peak areas as might be expected using channel-summation methods where there are close interferences in the 125-, 160-, and 332-keV regions. The calibration standards included a wide variety of plutonium oxide and metal samples with masses ranging from less than 0.5 g to approximately 1 kg.

Figure 8.18 shows the average accuracy of the Los Alamos system; ^{239}Pu, ^{240}Pu, and ^{241}Pu are measured to better than 0.2% and ^{238}Pu and ^{241}Am are measured to a few percent. These values are limited by the accuracy of the standards. The precision of the measurement is shown in Figure 8.19 as a function of count time. Figure 8.20 shows the precision of the specific power measurement, which is used in conjunction with a calorimetry measurement to give total plutonium mass. The specific power can be determined to better than 1% with a 30-min measurement and to about 0.5% with a 2-h measurement.

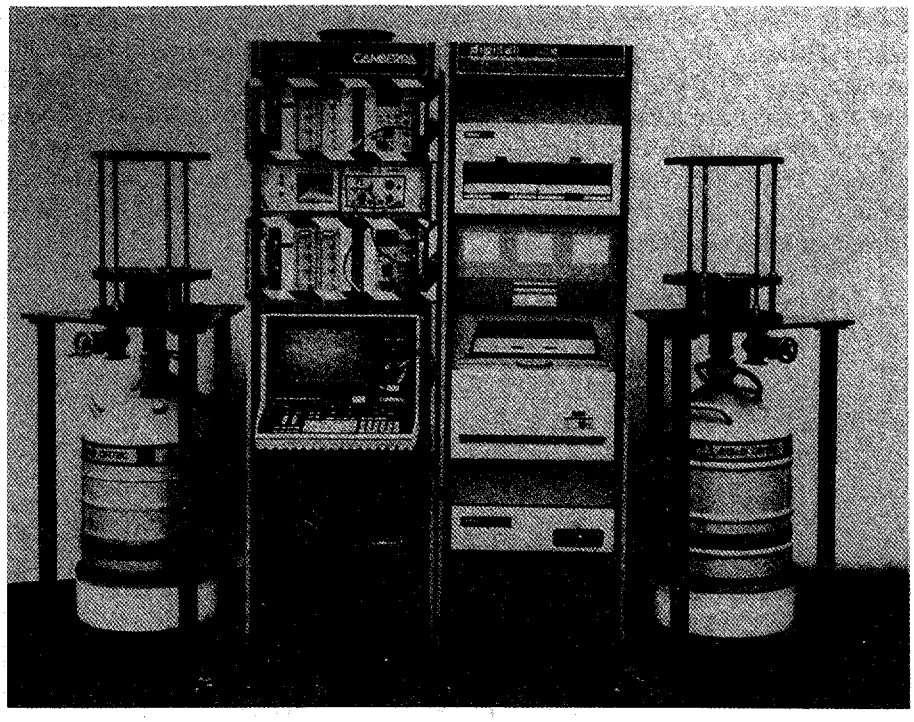

Fig. 8.17 Los Alamos multiple-detector plutonium isotopic system.

Table 8-16. Peaks used for interference subtraction

Region (keV)	Interfering Isotope	Peak Used for Subtraction (keV)
125	^{239}Pu	129.3
160	^{239}Pu	161.5
160	^{241}Pu	164.6[a]
332	^{239}Pu	345.0

[a]For aged material only.

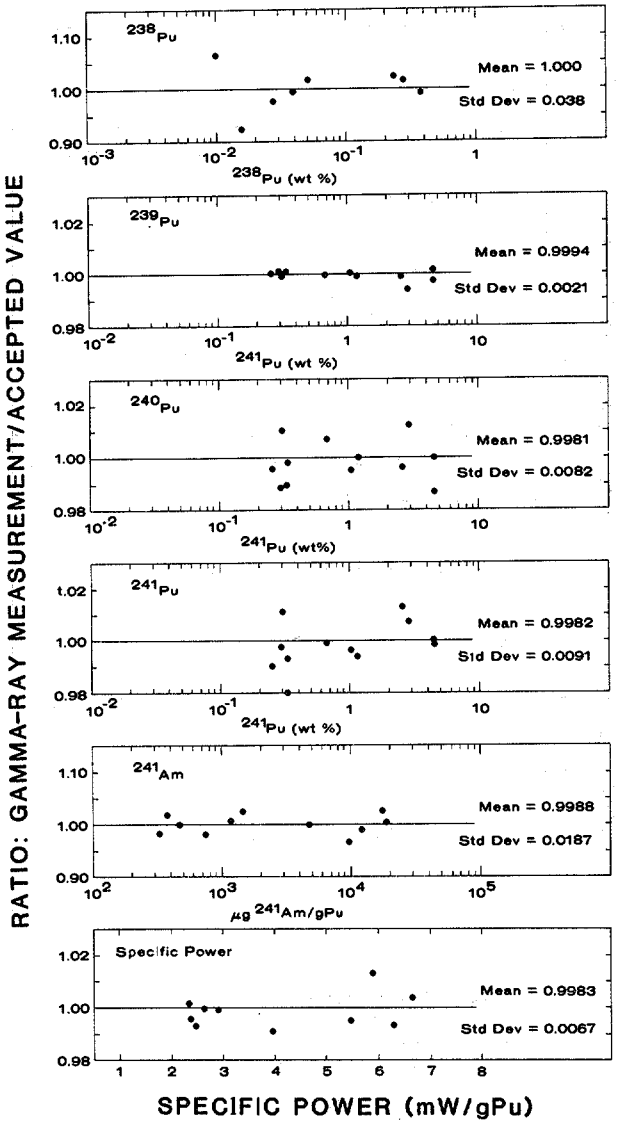

***Fig. 8.18** Accuracy of the Los Alamos plutonium isotopic system for a wide range of material types and isotopic composition.*

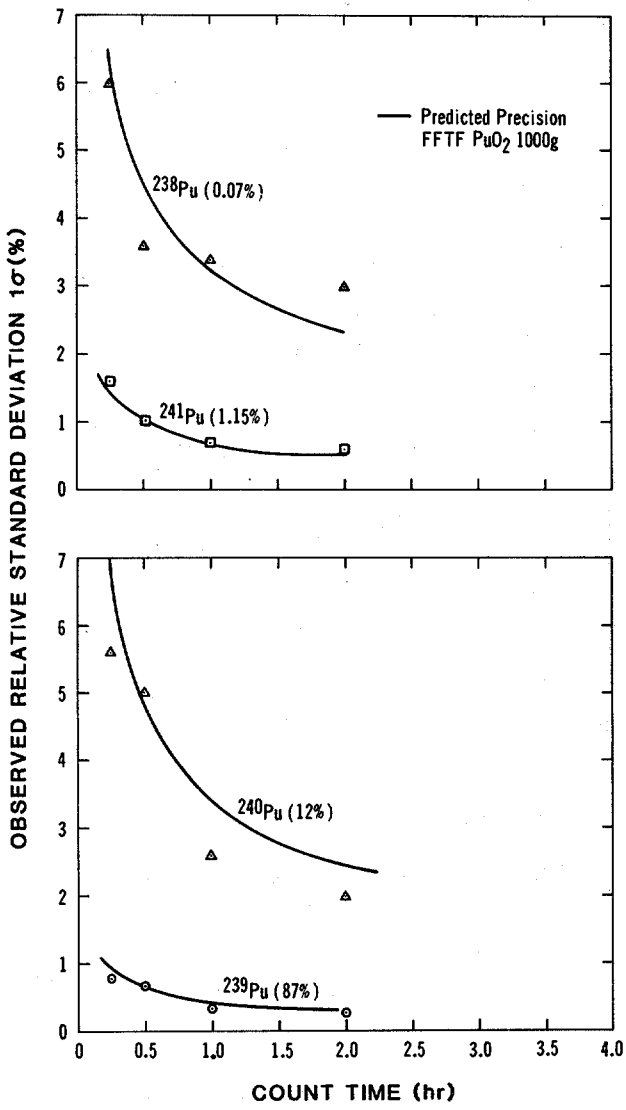

Fig. 8.19 *Precision of the Los Alamos plutonium isotopic system as determined from 30 measurements of a 1-kg sample of PuO_2 (12% ^{240}Pu). The solid line is the precision predicted from counting statistics.*

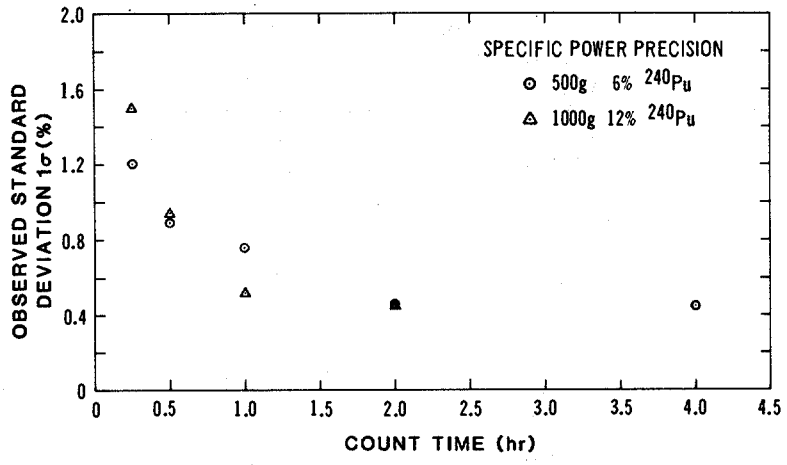

***Fig. 8.20** Precision of the Los Alamos plutonium isotopic system for the determination of specific power as used to interpret calorimeter measurements. Precision calculated from 30 measurements (circles indicate a 500-g sample of plutonium metal with 6% ^{240}Pu, triangles indicate a 1-kg sample of PuO_2 with 12% ^{240}Pu).*

8.7.3 Mound Facility

Investigators at the Mound Facility have carried out plutonium isotopic measurements in support of their calorimetry effort for some time. They use the peak-fitting programs GRPANL and GRPAUT described in Section 8.6.2. Long-term tailing is not used. The slope of the short-term tail B is fixed during initial detector characterization. The peak amplitudes Y_0 and widths α are free parameters; in some cases the short-term-tail amplitude is a free parameter. Over 50 peaks are fit in the region from 120 to 450 keV; the 640-keV region is also analyzed. Table 8-17 lists the ratios used in the GRPAUT program. A relative efficiency curve is determined from ^{239}Pu, ^{241}Pu-^{237}U, and ^{241}Am peaks; weighted least-squares techniques are used to fit the measured points to a smooth curve (see Equation 8.18). Equation 8-13 is used to estimate ^{242}Pu.

$$\ln \varepsilon_i = A_0 + \sum_{j=1}^{2} A_j / E_i^j + \sum_{j=1}^{3} A_{j+2} (\ln E_i)^j + A_6 \delta_6 + A_7 \delta_7 \qquad (8\text{-}18)$$

where ε_i = relative efficiency
E_i = gamma-ray energy
A_6, A_7 = constants to normalize ^{241}Pu-^{237}U and ^{241}Am points to ^{239}Pu
δ_6, δ_7 = 1 for ^{241}Pu and ^{241}Am, and 0 otherwise.

Table 8-17. Peak ratios calculated in GRPAUT

Ratio	Energy(keV)
241/239	208/203[a]
241/239	148/144
241/239	165/161
241/239	148/129[a]
238/239	153/144
238/241	153/148[a]
240/239	160/161
240/241	160/165
240/241	160/148[a]
Am/239	125/129
Am/239	335/345[a]
Am/239	369/375[a]
Am/239	662/646
Am/239	772/718

[a]Ratios used to calculate isotopic fractions. Weighted averages are used where appropriate.

The GRPAUT program has been used in several applications. Reference 20 describes a two-detector method that uses a planar detector in the 120- to 300-keV region and a coaxial detector in the 300- to 700-keV region. The coaxial detector is used to measure the 642.48-keV ^{240}Pu and 662.42-keV ^{241}Am gamma rays; the detector is heavily filtered to absorb gamma rays with energy below 100 keV. The precision for a 50 000-s count time is ~2% for both the 160/148 ^{240}Pu/^{241}Pu ratio and for the 642/646 ^{240}Pu/^{239}Pu ratio. For large samples, the 662/646 ^{241}Am/^{239}Pu ratio is significantly more precise than the 125/129 ^{241}Am/^{239}Pu ratio.

Figure 8.21 shows a system developed at Mound for the simultaneous calorimetric and gamma-ray spectroscopic assay of plutonium (Refs. 34 and 35). This system reduces operator radiation exposure through less sample handling and also reduces data transcription errors. Table 8-18 summarizes the measurement results obtained using the transportable assay system at three nuclear facilities. The measured samples contained from a few hundred grams to 2 kg of plutonium oxide and metal; most samples had a nominal ^{240}Pu concentration of 6%. Count times were generally 10 000 to 50 000 s. The total plutonium mass was measured with an accuracy of better than 1% and a precision of 1 to 3%.

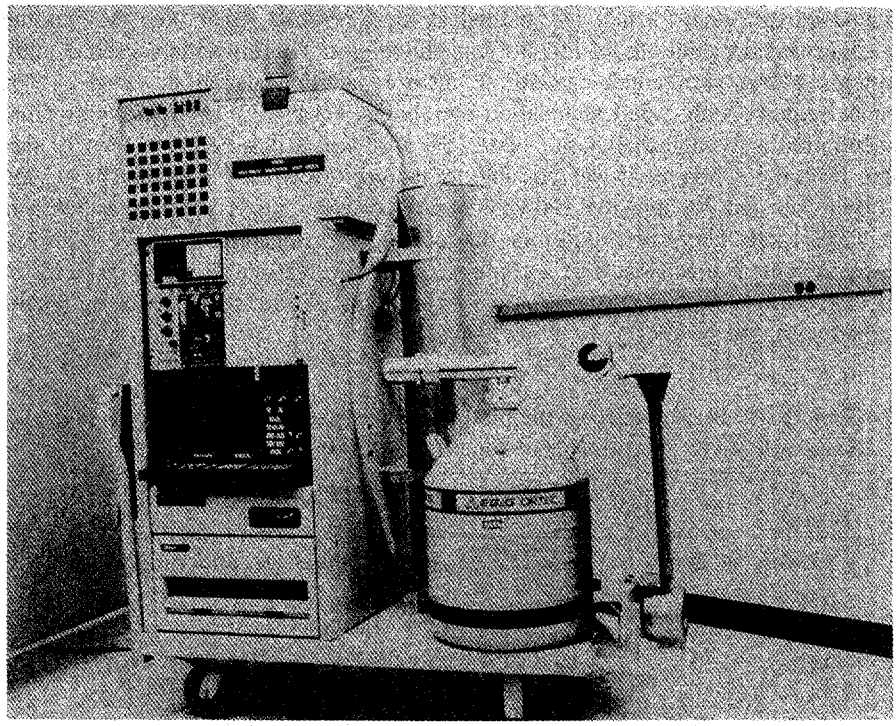

Fig. 8.21 *Transportable calorimeter with simultaneous calorimetry/gamma-ray spectroscopy capability. (Photo courtesy of J. G. Fleissner, Mound Facility.)*

Table 8-18. Simultaneous assay with transportable calorimeter and gamma-ray spectroscopy system[a]

Site	No. of Samples	Total Plutonium Assay Average Ratio Measured/Accepted	Precision of Ratio (%)
1	18	0.997	1.7
2	20	1.007	0.7
3	13	1.00	3.0

[a]Ref. 35.

8.7.4 Lawrence Livermore National Laboratory

The response-function analysis method has been used extensively by Gunnink and coworkers at Lawrence Livermore National Laboratory. In the early 1970s, they

installed an instrument using this method at the Savannah River Plant to measure low-concentration solutions of low-burnup plutonium. The instrument used a high-resolution planar germanium detector to measure a 10-mL solution sample. The samples required only small attenuation corrections because of their low plutonium concentration and small thickness (1 cm). Because americium had been freshly separated from the solutions, it was possible to analyze the 40-keV region. The system analyzed both the 40- and the 100-keV regions; these regions yield measurements with significantly better precision than that obtainable using regions above 120 keV. The measurement precision with a count time of only 10 min is within a factor of 2 of that customarily assigned to mass spectrometry. Table 8-19 lists some results obtained with this system.

Table 8-19. Performance of the plutonium isotopic assay system at the Savannah River Plant (response-function method)[a]

Isotope	Abundance (%)	Av Bias (%)	Precision (%)
1. Freshly separated solutions, 10-min count time, 3 g/L plutonium:			
238	0.008	5.6	4.7
239	93.46	0.048	0.049
240	5.88	0.75	0.72
241	0.65	0.96	1.9
2. Aged solutions, 60-min count time, 4 g/L plutonium:			
238	0.018	7.6	5.7
239	90.92	0.14	0.09
240	8.40	1.6	0.94
241	0.661	0.64	0.61
Am	- - -	0.08	0.26
Total Pu	5.4 g/L	0.46	0.35

[a]Ref. 4.

Gunnink developed a similar instrument to measure high-burnup (~20% ^{240}Pu), high-concentration (~250-g Pu/L) reprocessing plant solutions (Ref. 12). The instrument is installed at the reprocessing plant in Tokai-Mura, Japan. The 40-keV region is analyzed when measuring freshly separated solutions. In addition, the 148-keV peak and the 94-keV U $K_{\alpha 2}$ x-ray peak (which has contributions from all the plutonium isotopes) are used to measure ^{241}Pu, and the 129-keV gamma-ray peak is used to measure ^{239}Pu. Peak areas are determined using simple ROI-summation techniques; interfering peaks are stripped out channel by channel before summing. For aged solutions, response-function methods are applied to the 100-keV region. The 208- and

59-keV peaks are used to determine the energy calibration and the peak-shape parameters. Numerous interferences are stripped out of the 100-keV complex before fitting the response functions. The instrument is calibrated with known solution standards to measure the absolute concentration of each isotope. Equation 8-12 is used to estimate ^{242}Pu.

The very thin sample cell (~1 mm thick) shown in Figure 8.22 allows a 0.25-mL sample to be viewed by the detector. The cell is mounted on the detector face as shown in Figure 8.23, and the plutonium solution to be measured is pumped into the sample cell from a glovebox. Fresh solutions are counted for 15 to 30 min; aged solutions for 30 min to 1 h. Table 8-20 summarizes the performance of the Tokai system when measuring both freshly separated and aged process solutions.

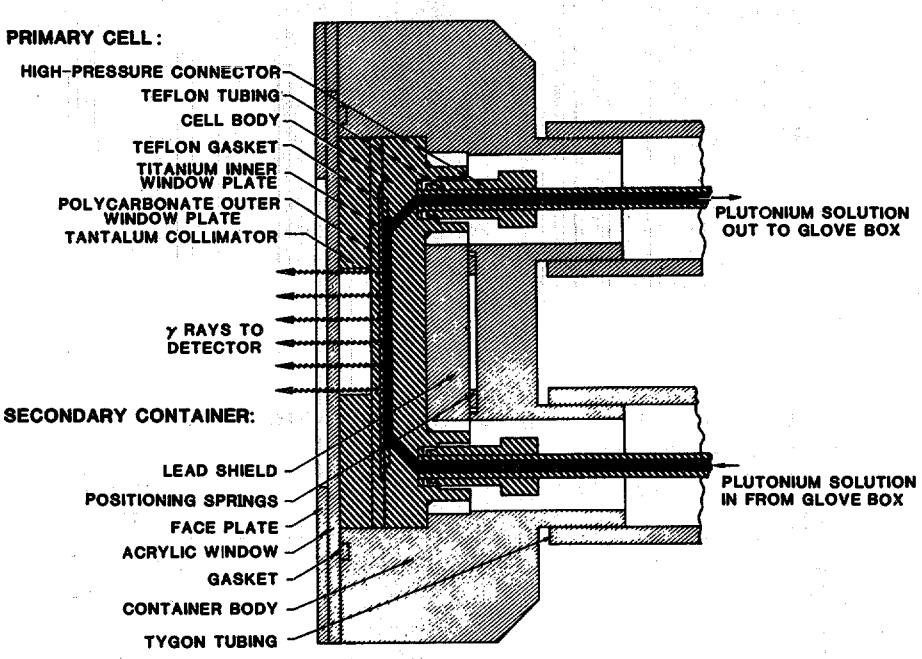

Fig. 8.22 *Sample cell used with the plutonium isotopic assay system installed at the Tokai-Mura reprocessing plant in Japan (Ref. 12). The cell fits over the front of a HPGe detector; plutonium solutions are pumped to the cell from a glovebox in the analytical laboratory. (Photo courtesy of R. Gunnink, Lawrence Livermore National Laboratory.)*

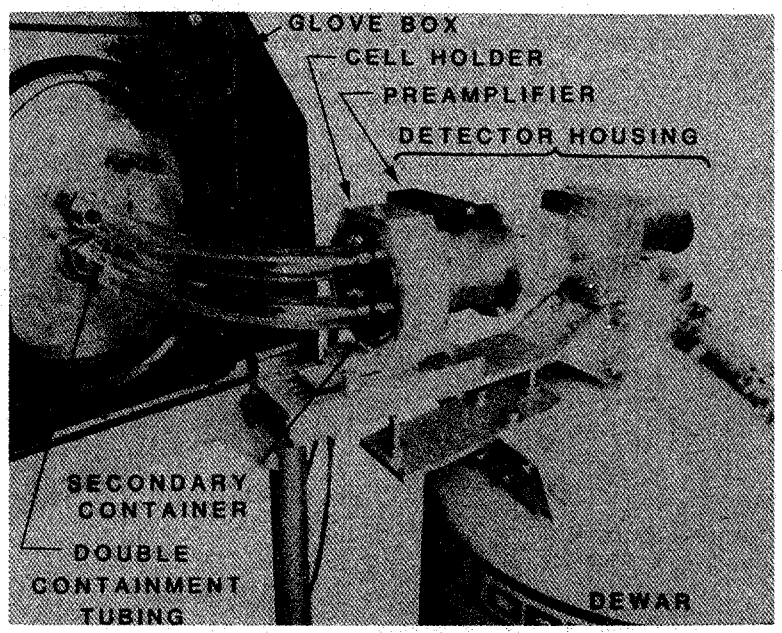

Fig. 8.23 HPGe detector and sample cell used with plutonium isotopic assay system installed at the Tokai-Mura reprocessing plant in Japan (Ref. 12). (Photo courtesy of R. Gunnink, Lawrence Livermore National Laboratory.)

Table 8-20. Performance of Tokai isotopic assay system; 94 samples, 130– to 270-g/L plutonium[a]

Isotope	Abundance (%)	Bias (%)	Precision (%)
238	0.5 – 1.0	0.1 – 0.8	0.4 – 0.7
239	60 – 75	0.01 – 0.3	0.08 – 0.4
240	17 – 23	0.02 – 0.4	0.2 – 1.3
241	5 – 11	0.02 – 0.8	0.2 – 0.8

[a]Ref. 12.

Response-function techniques have also been applied by Ruhter and Camp (Refs. 30 and 31) to solid samples using gamma rays in the 120- to 450-keV region. They developed a portable instrument for the use of safeguards inspectors at the International Atomic Energy Agency.

Another system is described by Gunnink in Ref. 36; it makes use of all the data available in the spectrum, accommodates two detectors (high and low energy), and analyzes data with both response functions and ROI peak integration. This system can obtain better precision than other systems can when measuring arbitrary samples because of its response-function analysis of the 100-keV region.

8.7.5 Summary of Measurement Precision

For all techniques discussed, the measurement precision is influenced most by the spectral region analyzed; a higher precision is obtained when measuring the lower energy regions that have higher gamma-ray emission rates. Table 8-21 summarizes the measurement precision attainable for different energy regions. Measurement accuracy is usually commensurate with precision.

Table 8-21. Typical measurement precision (%)

Region (keV)	Count Time	238	239	240	241	Am	Specific Power
40	10-30 min	0.3-5	0.05-0.5	0.2-1.0	0.2-1.0	- - -	- - -
100	30-60 min	0.3-5	0.05-0.5	0.2-1.0	0.2-0.8	0.1-1.0	0.1-1.0
>120	1-4 h[a]	>1-10	0.1-0.5	1-5	0.3-0.8	0.2-10	0.3-2

[a]With high-count-rate systems these precisions can be realized in less than 30 min.

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