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<sup>242</sup>Pu Isotopic Verification

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UNITED STATES DEPARTMENT OF ENERGY CONTRACT W-7408-ENG. 36

LA-8372-MS Informal Report

UC-15

Issued: May 1980

# <sup>242</sup>Pu Isotopic Verification

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### 242Pu ISOTOPIC VERIFICATION

by

T. E. Sampson, S. Johnson, and K. Kroncke

### **ABSTRACT**

We report on measurement methods being developed and implemented to verify nondestructively the isotopic composition of certain batches of plutonium highly enriched in  $^{242}\mathrm{Pu}$ . High-resolution gamma-ray spectroscopy techniques are used to measure the isotopic ratios of  $^{238}\mathrm{Pu}/^{241}\mathrm{Pu}$  and  $^{239}\mathrm{Pu}/^{241}\mathrm{Pu}$ , which are minor constituents of the enriched  $^{242}\mathrm{Pu}$ . These ratios uniquely verify the specific batch of  $^{242}\mathrm{Pu}$  and can indicate with high sensitivity if the  $^{242}\mathrm{Pu}$  has been contaminated with plutonium of the other isotopic compositions. Data are presented that illustrate the successful application of these techniques to samples of arbitrary geometry ranging in mass from  ${\sim}0.1$  g to  ${>}2$  kg.

### I. INTRODUCTION

The presence at Los Alamos Scientific Laboratory (LASL) of significant quantities of plutonium highly enriched in  $^{242}$ Pu has made it necessary to implement measurement methods for distinguishing between  $^{242}$ Pu and other plutonium isotopic compositions.

The majority of the  $^{242}$ Pu at LASL is enriched to either  $^{\circ}83\%$   $^{242}$ Pu or  $^{\circ}95\%$   $^{242}$ Pu. A method is desired that will distinguish between the 83% and 95% mixtures and that will also verify that the sample has not been cross-contaminated with plutonium of any other isotopic composition.

This report gives a preliminary account of the gamma-ray spectroscopic methods that are being be used to verify this

material at the LASL TA-55 Plutonium Facility. Since this work is ongoing, changes can be expected in the techniques and results as we gain experience with this new material.

### II. MINOR ISOTOPIC RATIOS FOR 242 Pu

Gamma rays at 44.92, 103.50, and 158.80 keV are reported from  $^{242}$ Pu. These gamma rays are relatively weak because of the long half-life of  $^{242}$ Pu (376 000 y) and have strong interferences from gamma rays and x rays from other plutonium isotopes. Specific interferences are  $^{240}$ Pu at 45.23 keV,  $^{241}$ Am at 102.97 keV,  $^{239}$ Pu at 103.02 keV,  $^{241}$ Pu at 103.68 keV, PuK $_{1}$  at 103.75 keV,  $^{241}$ Pu at 159.96 keV,  $^{239}$ Pu at 160.19 keV, and  $^{240}$ Pu at 160.28 keV. Direct spectroscopy on the  $^{242}$ Pu gamma rays from the  $^{242}$ Pu isotopic mixtures at LASL is not feasible at this time.

However, certain minor isotope ratios that can be measured by gamma-ray spectroscopy techniques seem to be useful in verifying this material. In Table I we show typical isotopic compositions

TABLE I

Pu ISOTOPIC COMPOSITION FOR Pu

FUELS AND LASL 242Pu MIXTURES

Pu Specifications	238 <sub>Pu</sub>	239 <sub>Pu</sub>	240 <sub>Pu</sub>	241 <sub>Pu</sub>	242 <sub>Pu</sub>
Weapons Grade	0.01	93.6	5.9	0.40	0.015
FFTF Feed	0.08	86.1	11.7	1.90	0.20
Spent LWR	1.10	61.6	20.9	12.6	3.8
LWR Recycle	3.3	41.1	27.6	17.6	10.4
242 <sub>Pu</sub> "High"	0.49	0.56	2.46	1.49	94.98
242 <sub>Pu</sub> "Low"	1.09	1.34	10.11	5.07	82.37

for plutonium fuels and the  $^{242}$ Pu mixtures at LASL. The  $^{242}$ Pu isotopics are averaged over the entire batch. Individual  $^{242}$ Pu samples may exhibit isotopics different from the averages.

In Table II the minor isotope ratios  $^{241}$ Pu/ $^{238}$ Pu and  $^{239}$ Pu/ $^{241}$ Pu are presented. The  $^{241}$ Pu/ $^{238}$ Pu ratio is straightforward to measure with a precision of near 1%. Also, this ratio was constant for all the LASL  $^{242}$ Pu feed material in both enrichment classes. The measurement of this ratio can be used to distinguish between the two classes of  $^{242}$ Pu. This ratio is significantly different for the  $^{242}$ Pu material than for either weapons grade plutonium or Fast Flux Test Facility (FFTF) feed, which are commonly found at LASL.

Because of the low  $^{239}$ Pu content of the  $^{242}$ Pu mixtures, the  $^{239}$ Pu/ $^{241}$ Pu ratio is very sensitive to contamination with grades of plutonium with high  $^{239}$ Pu content. The measurement of this ratio assures that contamination has not taken place.

TABLE II
ISOTOPIC RATIOS

Pu Specification	$\frac{241_{\text{Pu}}/238_{\text{Pu}}}{}$	239 <sub>Pu</sub> /241 <sub>Pu</sub>
Weapons Grade	40.0	234.0
FFTF Feed	23.75	45.3
Spent LWR	11.45	4.89
LWR Recycle	5.33	2.34
242 <sub>Pu</sub> "High"	3.04	0.376
242 <sub>Pu</sub> "Low"	4.65	0.264

For the  $^{242}\text{Pu}$  material this ratio is an order of magnitude or more different from that expected for any other plutonium material available in quantity.

The initial feed showed some variability in the  $^{239}$ Pu/ $^{241}$ Pu ratio for both  $^{242}$ Pu grades, arising mainly from variable  $^{239}$ Pu. Most of the feed for both grades exhibited a  $^{239}$ Pu/ $^{241}$ Pu ratio of  $^{\sim}0.23$ . About 25% of the feed in each batch had higher  $^{239}$ Pu content leading to the averages in Table II. For most of the  $^{242}$ Pu feed the  $^{239}$ Pu/ $^{241}$ Pu ratio was not significantly different between the two grades. Because this ratio is measured with an uncertainty of  $^{\sim}10\%$ , it can not be reliably used at this time to distinguish between the two grades of  $^{242}$ Pu.

As the feed material in each grade becomes mixed and blended via reprocessing and recycling, the  $^{239}$ Pu/ $^{241}$ Pu ratios may approach the average values of Table II. If this happens, this ratio may also be able to distinguish between the two grades as well as flagging  $^{239}$ Pu contamination.

In Figs. 1 and 2 the sensitivity of the two isotopic ratios for sensing contamination with weapons grade plutonium is shown.

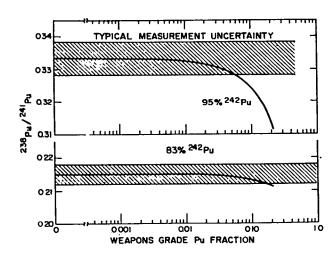


Fig. 1. 238<sub>Pu/241Pu</sub> ratio is insensitive to contamination with weapons grade Pu.

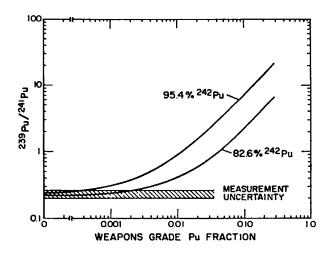


Fig. 2. 239<sub>Pu</sub>/241<sub>Pu</sub> ratio is sensitive to contamination with weapons grade Pu.

The  $^{238}$ Pu/ $^{241}$ Pu ratio (Fig. 1) is very poor for detecting contamination with weapons grade plutonium. However, the  $^{239}$ Pu/ $^{241}$ Pu (Fig. 2) ratio is sensitive to contamination with a fraction of weapons grade plutonium around 0.1%.

The combination of these two ratios provides a very good means of verifying the batch and isotopic purity of the bulk of the  $^{242}$ Pu material at LASL. These techniques are quite specific for the current  $^{242}$ Pu at LASL. Any additional  $^{242}$ Pu mixtures would have to be studied to see if these techniques are applicable.

# III. GAMMA-RAY SPECTROSCOPIC MEASUREMENT OF ISOTOPIC RATIOS WITH APPLICATION TO <sup>242</sup>Pu

The intrinsic self-calibration method used here to measure isotopic ratios by gamma-ray spectroscopy was developed by Parker and Reilly in 1974. This technique is gaining wide acceptance.  $^{2-4}$  Other references may be found in the above references. The main feature of this technique is that it is applicable to samples of arbitrary geometry without recourse to standards.

This method determines isotopic ratios from the areas of close-lying peaks in the gamma-ray spectrum from the sample of interest. The ratio of isotope 1 to isotope 2 is given by

$$\frac{N_1}{N_2} = \frac{C_1}{C_2} \times \frac{T_1(\frac{l_2}{l_2})}{T_2(\frac{l_2}{l_2})} \times \frac{B_2}{B_1} \times \frac{\varepsilon_2}{\varepsilon_1} \quad , \tag{1}$$

where

N = relative number of atoms of indicated isotope,

C = photopeak counts from gamma ray from indicated
 isotope,

 $T(\frac{1}{2})$  = isotope half-life,

B = branching ratio of indicated isotope (gamma rays/ disintegration), and ε = relative efficiency including detector efficiency, sample self-absorption, attenuation in media between sample and detector, and sample-detector geometry.

C is measured from the gamma-ray spectrum.  $T(\frac{1}{2})$  and B are taken from known nuclear data. The branching ratios of Gunnink<sup>5</sup> are used for B.  $\varepsilon$  is determined from a relative efficiency curve found from the spectrum under study by a curve of photopeak area/branching ratio for gamma-ray lines from one of the isotopes in the sample. For  $^{242}$ Pu samples we determine this curve by using  $^{241}$ Pu and  $^{241}$ Pu- $^{237}$ U lines at 148.6, 164.6, 208.0, 267.5, 332.4 and 370.9 keV.

The shape of the relative efficiency curve depends mainly on

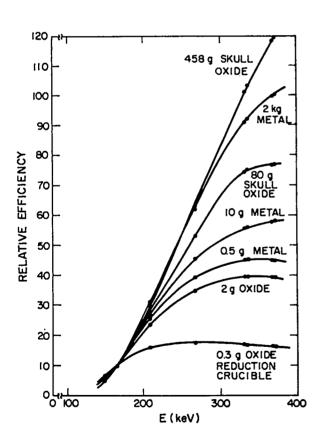


Fig. 3.
Typical relative efficiency curves determined from <sup>241</sup>Pu, <sup>241</sup>Pu, <sup>241</sup>Pu ines in <sup>242</sup>Pu samples. Smooth curves are drawn to guide the eye.

the detector, sample size and configuration, and filtering. The family of relative efficiency curves in Fig. 3 were measured with a 14% coaxial intrinsic germanium detector with filtering from 0.51-mm Ta, 0.76-mm Cd, and 0.38-mm brass. They are normalized to 10.0 for the 164.6-keV gamma ray from 237U.

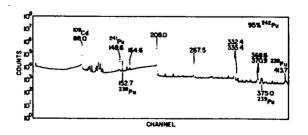


Fig. 4.
Gamma-ray pulse-height spectrum from 10-g metal disk of 95% 242Pu. Coaxial intrinsic Ge detector with 14% relative efficiency.

In Fig. 4 the gamma-ray pulse-height spectrum from a 10-g metal disk of 95%  $^{242}$ Pu is shown. Filtering is as described above. Most of the prominent lines in the gamma-ray spectrum arise from the  $^{237}$ U daughter of  $^{241}$ Pu. The 148.6-keV line from  $^{241}$ Pu and the 152.7-keV  $^{238}$ Pu line are strong. The usually strong  $^{239}$ Pu lines at 375 and 414 keV are quite weak here because of the low  $^{239}$ Pu content (0.45%) of the sample. The  $^{239}$ Pu line at 129.3 keV is not useful because of the high Compton continuum from the 208-keV line and from neutron background in the detector.

The  $^{238}$ Pu/ $^{241}$ Pu and  $^{239}$ Pu/ $^{241}$ Pu ratios to be used for verification are determined from the 150-keV region and 370 keV-region, respectively.

The <sup>238</sup>Pu/<sup>241</sup>Pu ratio is determined from the 152.7-keV <sup>238</sup>Pu peak and the 148.6-keV <sup>241</sup>Pu peak. The spectrum is clean in this region and this ratio is straightforward to measure. However, the steepness of the relative efficiency curve makes the relative efficiency correction rather large, typically 20-30%. Figure 5 shows typical data in this energy range.

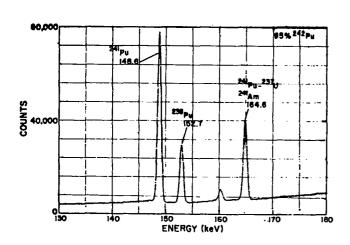


Fig. 5.
Typical data from 150-keV region used to determine 238 Pu/241 Pu from 152.7/148.6-keV ratio.

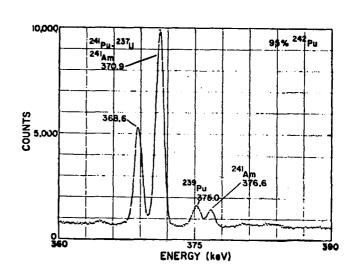


Fig. 6.
Spectral features of 370-keV region used to determine 239pu/241pu ratio at 375.0/370.9 keV.

The  $^{239}$ Pu/ $^{241}$ Pu ratio is measured at 375.0 keV ( $^{239}$ Pu) and 370.9 keV ( $^{241}$ Pu- $^{237}$ U) (Fig.6). This region is more complex. The 370.9-keV peak must be stripped from its neighbor at 368.6 keV. The 375.0-keV peak has a high energy shoulder from  $^{241}$ Am at 376.6 keV. The 413.7-keV line from  $^{239}$ Pu is also visible and useful in some cases. It, however, rides on the tail of a peak at 416 keV from pileup of the intense 208-keV line.

To measure the  $^{239}$ Pu/ $^{241}$ Pu ratio at 375/370.9, one must be certain that the 370.9-keV line from  $^{237}$ U is in equilibrium with its  $^{241}$ Pu parent.  $^{237}$ U grows in with a 6.75 day half-life. Ninety-nine percent of equilibrium intensity is achieved in 45 days. Cases where there is no  $^{241}$ Pu- $^{237}$ U equilibrium are discussed in Sec. IV.

# IV. 242 Pu VERIFICATION PROBLEMS AT THE LASL TA-55 PLUTONIUM FACILITY

In general, the verification problems at TA-55 divide into two classes, (1) those involving aged material ( $^{5}45$  days since chemical separation of uranium) where  $^{241}$ Pu- $^{237}$ U equilibrium is present, and (2) those involving fresh material where there is no  $^{241}$ Pu- $^{237}$ U equilibrium. The nonequilibrium case is discussed next.

## A. Verification of Freshly Separated <sup>242</sup>Pu Mixtures

The general procedures discussed in Sec. III apply only to material where there is equilibrium between  $^{241}$ Pu and  $^{237}$ U. If this does not exist, the 370.9-keV line from  $^{237}$ U can not be used to indicate  $^{241}$ Pu and hence the  $^{239}$ Pu/ $^{241}$ Pu ratio at 375.0/370.9 will not be valid. Also, the relative efficiency point at 148.6 keV from  $^{241}$ Pu will not lie on the same curve as all the other points (164.6, 208.0, 267.5, 332.4, 370.9 keV) that arise from  $^{237}$ U. This makes the determination of the relative efficiency at 148.6 and 152.7 keV more difficult, because the relative efficiency curve will now have to be extrapolated to

148.6 and 152.7 keV from the nearest points at 164.6 and 208.0 keV. The curvature in typical relative efficiency curves (Fig. 3) in this region makes this extrapolation unreliable.

Fortunately the geometry of the freshly separated <sup>242</sup>Pu mixtures enables the use of another technique. Verifications on freshly separated material are desired at three points in the recovery process. The first place is the metal button formed from the reduction of PuO2. This button is about 76 mm diameter with a thickness in the middle of about 13 mm tapering to less than half of that at the outer rim. The second verification point is a cylindrical metal ingot formed by melting and casting several buttons. This ingot is about 60 mm diameter by about 76 mm high. The third verification point is the output of the electrorefining process, which yields a donut-shaped metal piece about 25 mm high with a 14-mm-thick by 100-mm-OD ring. All of these three forms have in common a metal section of \$13 mm inches in thickness. With the mean free path of  $\alpha\text{-phase}$  plutonium being 1.5 mm and 1.8 mm at 375 and 414 keV, respectively, we see that all three metal forms are "infinitely thick" for these and other lower energy gamma-ray emissions. This condition means that the isotopic enrichment can be measured directly without recourse to ratios of gamma-ray intensities. A discussion of the principles of enrichment measurements may be found in Ref. 6 and references therein.

Enrichment measurements require standards for calibration. A desirable metal standard would be 38-50 mm diameter by 13 mm thick and would require between 300 and 500 g of <sup>242</sup>Pu. This is too much <sup>242</sup>Pu to tie up in permanent calibration standards. Two alternate methods of enrichment calibration will be studied. The first will use an actual <sup>242</sup>Pu metal part (button, ingot, or donut) with mass spectrometry measurement of its isotopic composition. This will be a transient standard because it will disappear after the measurement. The second method will use permanent standard of weapons grade plutonium and a 1% weapons grade Pudepleted uranium alloy. These will be permanent standards that could be used in a measurement control program if desired.

The enrichment measurements will measure  $^{239}$ Pu content using the 375- and 414-keV gamma rays.  $^{241}$ Pu and  $^{238}$ Pu will also be measured at 148.6 keV and 152.7 keV using this method.

A fixed reproducible geometry is necessary for these measurements. Also, pileup and deadtime effects arising from count-rate variations must be accounted for. A 109Cd source is used to normalize the data and correct for count-rate-associated effects. The analysis software also corrects for the decay of the 109Cd source between the enrichment calibration measurement date and the measurement date of an unknown sample. To allow for small differences between the geometry of the permanent standards and that of the unknowns, we have incorporated an additional adjustable parameter in the enrichment calculation expression.

If the geometry is not constant between the enrichment calibration and the measurement of an unknown, the enrichment results will be invalid. However, the isotopic ratio of  $^{238} \mathrm{Pu}/^{241} \mathrm{Pu}$  can still be measured at 152.7/148.6 keV. This is done independently of the enrichment measurement to provide additional verification capability. The use of thick samples and the low energy of the  $^{238} \mathrm{Pu}$  and  $^{241} \mathrm{Pu}$  gamma rays should make it possible to perform this ratio measurement with a single multiplicative calibration constant. This means that we do not have to worry about extrapolation of the relative efficiency curve for the nonequilibrium measurements.

As of this writing no  $^{242}$ Pu metal has been produced at LASL that requires nonequilibrium or enrichment measurements. The testing and refinement of the enrichment techniques will have to await production of freshly separated metal.

## B. Verification of <sup>242</sup>Pu Mixtures with <sup>241</sup>Pu-<sup>237</sup>U

in Equilibrium. Details and Results

Equilibrium  $^{242}$ Pu mixtures are verified using the principles discussed in Secs. II and III. For measurements on equilibrium material where gamma-ray lines from  $^{237}$ U can be used for  $^{241}$ Pu verification, the sample may be of arbitrary geometry and composition. The sample spectrum is self-calibrating with

gamma-ray lines at 164.6, 208.0, 267.5, 332.4, 335.4, 368.6, and 370.9 keV from  $^{237}$ U and 148.6 keV from  $^{241}$ Pu being used to define the relative efficiency curve.

The  $^{238}$ Pu/ $^{241}$ Pu ratio is determined from the 152.7-keV  $^{238}$ Pu line and 148.6-keV  $^{241}$ Pu line. The relative efficiency at 152.7 keV is determined by a quadratic fit to the relative efficiency points at 148.6, 164.6, and 208.0 keV.

For the  $^{239}$ Pu/ $^{241}$ Pu ratio at 375.0/370.9 keV the relative efficiency at 375.0 keV is determined from a linear extrapolation of the efficiency points at 332.4 and 370.9 keV.

Before the relative efficiency points are computed, all  $^{237}\text{U}$  lines are corrected for their  $^{241}\text{Am}$  content. This correction is made using  $^{241}\text{Pu}/^{241}\text{Am}$  ratios computed from pairs of neighboring lines that each have contributions from both isotopes. The 332-335 complex is used 1 to determine the correction for the 164-, 208-, 267-, 332-, and 335-keV lines. In a similar fashion the 368-371 complex is used to determine the correction for the 368- and 371-keV lines.

For material that is  $\sim$ l yr old this correction is only a few tenths of a percent for the 164-, 208-, 267-, and 332-keV lines. The correction is about 3% for 37l keV whereas the lines at 335 and 368 keV have larger corrections of about 25%.

At the present time the  $^{241}$ Pu/ $^{241}$ Am ratios from the 332 and 368 complexes are not as consistent as expected. The agreement gets poorer as the  $^{241}$ Am content gets lower. Some of the branching ratios used may be in error. However, the  $^{\sim}25\%$  corrections that have been applied to the 335- and 368-keV lines in Fig. 3 do seem to bring these points into agreement with the remainder of the curve.

We have been able to accumulate a reasonable amount of data on a wide range of samples that have attained  $^{241}\mathrm{Pu}^{-237}\mathrm{U}$  equilibrium. The relative efficiency curves for some of these samples have been shown in Fig. 3. In Table III a comparison of measured  $^{238}\mathrm{Pu}^{/241}\mathrm{Pu}$  ratios to those found from mass spectrometry is shown. The average of all the mass spec/measured ratios has been used to normalize this ratio. This normalization parameter is

TABLE III
ISOTOPIC RATIOS 238pu/241pu

Sample	Measured	Mass Spec	Measured/ Mass Spec
0.5 g 95% metal	0.3270 <u>+</u> 0.0028	0.3312 0.3254 0.3307	0.987 1.005 0.989
2 g 83% oxide	0.2199 <u>+</u> 0.0012	0.2234 0.2222	0.984 0.990
2 g 95% oxide	0.3335 <u>+</u> 0.0019	0.3312 0.3254 0.3307	1.007 1.025 1.008
10 g 95% metal	0.3374 <u>+</u> 0.0015	0.3421 0.3380	0.986 0.998
20 g 95% oxide	0.3289 <u>+</u> 0.0014	0.3312 0.3254 0.3307	0.993 1.011 0.995
2 kg 95% metal	0.3364 <u>+</u> 0.0032	0.3360 0.3373	1.001 0.997
2 kg 95% metal	0.3354 <u>+</u> 0.0032	0.3367 0.3329	0.996 1.008
80 g 95% oxide	0.3368 <u>+</u> 0.0025	0.3544 0.3370	0.950* 0.999
240 g 95% oxide	0.3324 <u>+</u> 0.0035	0.3312 0.3254 0.3307	1.004 1.022 1.005
	•	mean std. dev	1.000 0.011

<sup>\*</sup>omitted from average

1.0317. That this parameter is not 1.0 is attributed to an error in the published  $^5$  branching ratio for the 152.7-keV gamma ray of  $^{238}$ Pu. These measurements of this branching ratio discrepancy are in good agreement with those of other investigators.  $^{3,4}$ 

Examination of Table III shows the following: (1) estimated precision ranges from 0.5-1.0%, (2) the experimental precision is about 1% and is probably consistent with the estimated precision considering that the mass spectrometry values are not without errors, and (3) mass spectrometry measurements of this ratio are not significantly better than the gamma-ray measurements. Indeed the mass spectrometry values have been plagued by problems with possible sample inhomogeneity. The gamma-ray measurements average over a much greater volume than the mass spectrometry values.

Sample inhomogeneity will show up in a more pronounced fashion in the  $^{239}$ Pu/ $^{241}$ Pu ratios shown in Table IV. Because the initial feed shows variablity in the  $^{239}$ Pu/ $^{241}$ Pu ratio, poor mixing could result in inhomogeneous samples. Also, during sample preparation the small mass spectrometry samples are very susceptible to contamination with weapons grade plutonium. These factors, plus the fact that the gamma ray measurement is imprecise because of the weak  $^{239}$ Pu peak at 375 keV, lead to a much greater spread in the measured/mass spec values for the  $^{239}$ Pu/ $^{241}$ Pu ratios in Table IV. No normalization has been applied to the measured/mass spec ratio.

More measurements need to be compared to mass spectrometry results to understand fully if mixing and contamination are serious problems.

### V. LIMITATIONS OF VERIFICATION METHODS

In Sec. II we stressed that the methods described here are useful for the specific  $^{242}$ Pu mixtures now at LASL. These methods are not designed to work on highly enriched  $^{242}$ Pu rich mixtures of arbitrary isotopic composition.

The major limitation that currently could affect verification measurements is the lack of a proven technique to verify

TABLE IV

ISOTOPIC RATIOS 239Pu/241Pu

Sample	Measured	Mass Spec	Measured/ Mass Spec
2 g 83% oxide	0.2614 <u>+</u> 0.0055	0.2920 0.2942	0.895 0.889
10 g 95% metal	0.3412 <u>+</u> 0.0077	0.3266 0.3094	1.045 1.103
2 kg 95% metal	0.3880 <u>+</u> 0.0130	0.3853 0.3889	1.007 0.998
2 kg 95% metal	0.3950 <u>+</u> 0.0130	0.3894 0.3922	1.014 1.007
0.5 g 95% metal	0.4960 <u>+</u> 0.0150	0.4561 0.4819 0.4605	1.087 1.029 1.077
2 g 95% oxide	0.4740 <u>+</u> 0.0110	0.4561 0.4819 0.4605	1.039 0.984 1.029
20 g 95% oxide	0.4584 <u>+</u> 0.0083	0.4561 0.4819 0.4605	1.005 0.951 0.995
240 g 95% oxide	0.4520 <u>+</u> 0.0140	0.4561 0.4819 0.4605	0.991 0.938 0.981
		mean std. dev	1.003 0.056

nonequilibrium ( $^{241}$ Pu- $^{237}$ U) samples of arbitrary geometry. The major problem arises in trying to determine the relative efficiency curve at 148 and 152 keV when the nearest points available are 164, 208, and 267 keV. An extrapolation from 164 and 208 keV to 148 and 152 keV will be unreliable because of the curvature of the relative efficiency curves in this region (see Fig. 3). This case is still under study.

Large samples (>500 g or so) present a problem from the high neutron background associated with the spontaneous fission of  $^{242}$ Pu. This neutron background degrades the quality of the data by increasing backgrounds and adding tailing to the peaks. Consequently, the results will be somewhat poorer for large samples than for small samples.

For all samples the precision of the results is affected by the length of time the sample is counted. The  $^{238}$ Pu/ $^{241}$ Pu ratio at 152/148 keV can be determined very quickly (a few hundred seconds) for rough verification purposes. The  $^{239}$ Pu/ $^{241}$ Pu ratio is much more difficult and takes longer, especially with large high neutron background samples. The count time is variable and we have used 1000-3000 seconds for routine assays at TA-55. For these count times the lower limit of sample mass that is feasible to count is probably somewhat less than 0.1 q.

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