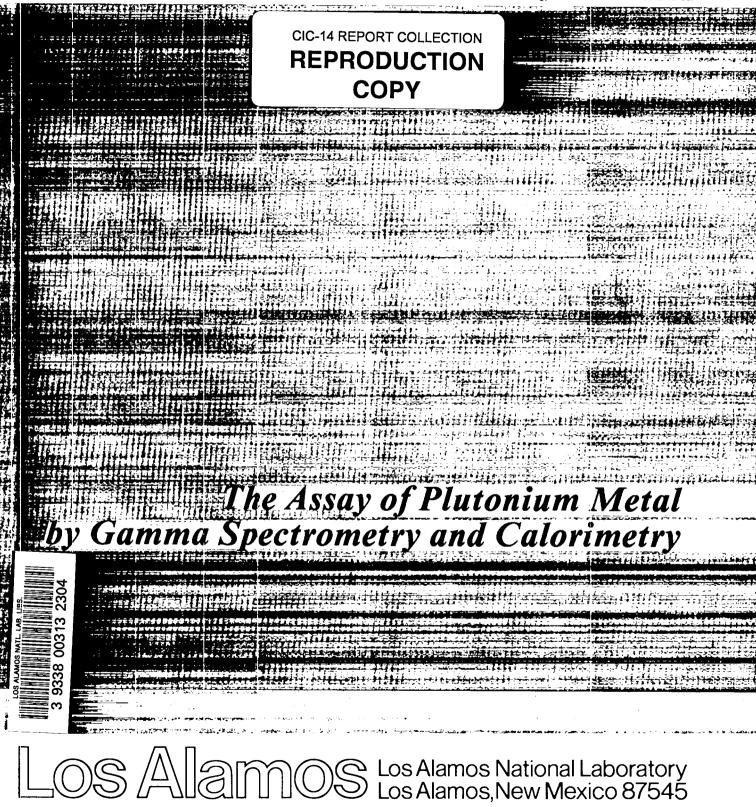


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The Assay of Plutonium Metal Gamma Spectrometry and Calorimetry

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by

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ABSTRACT

The nondestructive assay of plutonium buttons containing 1-3 kg plutonium agrees within 0.2% of the value calculated by chemical assay. These buttons were 4-15% 240 Pu by weight and contained 100 to 10000 ppm 241 Am. The nondestructive assay consisted of a gamma spectrometric determination of the isotopic distribution and a calorimetric determination of the power. Although further study is needed to extend the quantitative results to lower 240 Pu and 241 Am concentrations, the method is a rapid, nonintrusive technique for assaying plutonium.

I. INTRODUCTION

We are always interested in proposals for reliably assaying the plutonium content of materials in the Plutonium Facility at Los Alamos. If we could quickly determine an accurate and precise plutonium value, we could improve the present accounting procedures. The nondestructive assay of plutonium by gamma spectrometry and calorimetry is a promising method for such an assay.¹ The isotopic distribution in typical material, containing 4-15% ²⁴⁰Pu, can be determined accurately in <4 h, and calorimetry can be completed in about the same time. Most shipping cans could be assayed as received with no need for unpackaging.

We determined the plutonium value for 400 metal buttons by gamma spectrometry/calorimetry, chemical assay/calorimetry, and chemical assay/mass measurement (plutonium-factor value). The ²⁴⁰Pu distribution value determined by gamma spectrometry was then used to select button blends for ingots of specified ²⁴⁰Pu composition. An independent value was that given by the shipper, which will be designated as the declared value. We do not know how the declared value was obtained; and, in fact, it may duplicate either the chemical/calorimeter or plutoniumfactor value. We do not have a true plutonium value and do not claim that any one of the four values is more accurate than any other. Although differences in values for individual buttons are interesting, summarizing data is more useful. For the purpose of this report, we will discuss and compare our determinations, the limitations of the present gamma instrument, and the accuracy and precision of the results.

II. EXPERIMENTAL

The gamma instrument is shown in Fig. 1. The intrinsic germanium detector, which is enclosed in lead shielding, is located directly under the sample chamber and beneath the glove box. The sample chamber consists of a 0.6-cm thick, 30-cm diam Lexan window in the glove box floor over the detector and a 2.5-cm-thick, 45-cm-diam lead disk with a variable diameter collimator (2.5-15.0 cm) directly over the window. Sample cans were placed on an adjustable platform over the lead disk. Both the diameter of the collimator and the sample-to-detector distance were varied to optimize counting statistics. The chamber was placed in a glove box to allow the assay of contaminated materials; however, we have used this system, up to now, only on packaged samples.

The electronics, analyzer, and computer for the instrument are approximately 10 m from the detector. A Canberra Series 80 Multichannel Analyzer (Fig. 2), interfaced with a PDP 11/34 computer and printer, is programmed to collect the data.

A sample that consisted of a double can containing the packaged button was placed on the platform in the sample chamber and the sample-to-detector distance adjusted to obtain 10000-12000 counts/s. At lower count rates, the can was placed on the lead shield (Fig. 3). The system was programmed to collect the peak areas at 25 gamma peaks between 125 and 415 keV for a preselected time, usually 3.5-12 h. Each peak is characteristic of the concentration of a given isotope, and by taking ratios of the areas of ²³⁸Pu peaks, ²³⁹Pu peaks, and ²⁴⁰Pu peaks to nearby ²⁴¹Pu peaks and the ratio of the areas of ²⁴¹Am peaks to ²³⁹Pu peaks, the distribution of the isotopes can be calculated. Since ²⁴²Pu has no known usable gamma peaks, its distribution was calculated from the other isotopes by isotopic correlation techniques. Fortunately, the ²⁴²Pu concentration is very small and its estimate can be quite poor without affecting the assay. The ratios, the fractions, the weight per cent of the plutonium isotopes, and the ²⁴¹Am concentration were calculated by the computer. The results were printed and the spectra stored on a floppy disk. The gamma spectrometer was calibrated before these assays with a series of well-characterized plutonium oxides and a plutonium metal button. During the study, the metal button and a 1000-g can of well-characterized PuO2 were occasionally used to ensure that there were no changes in the calibration.

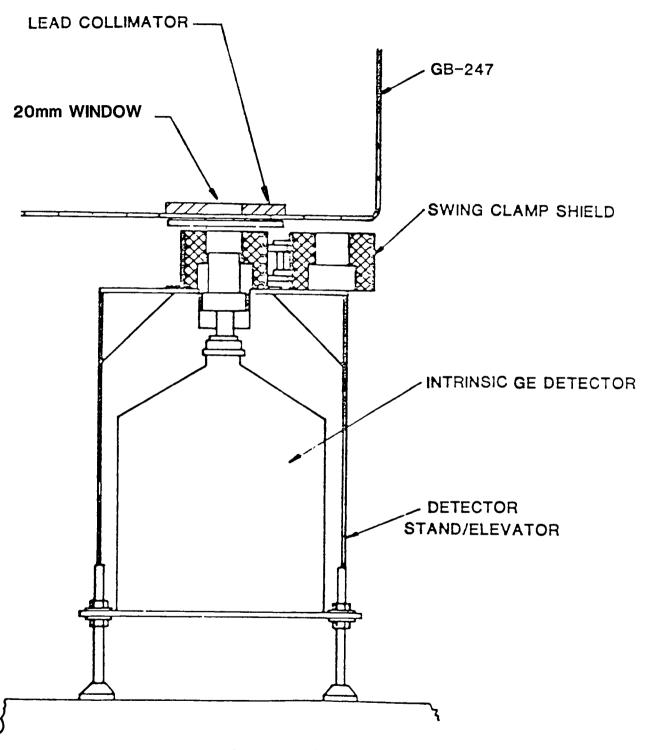


Fig. 1. Gamma detection station.

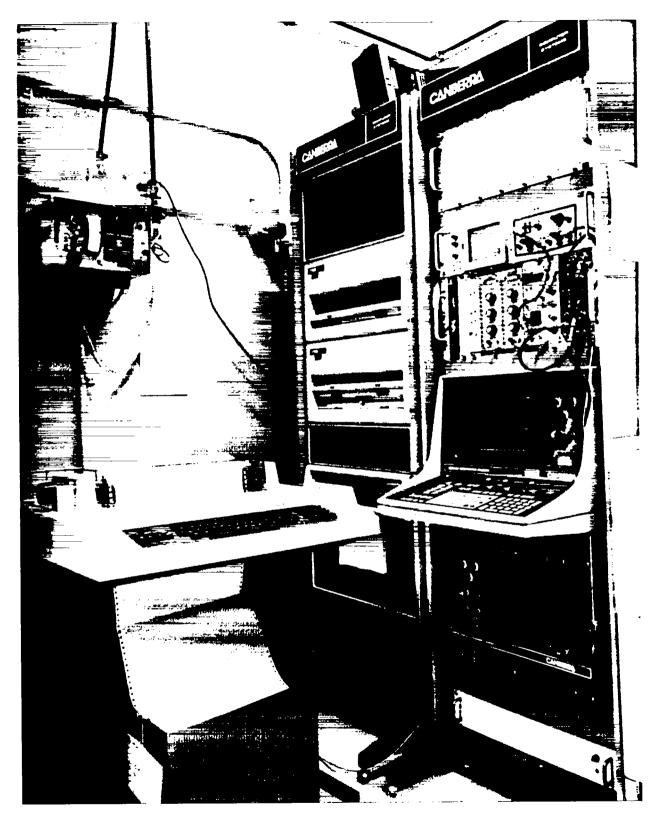


Fig. 2. Computer system.

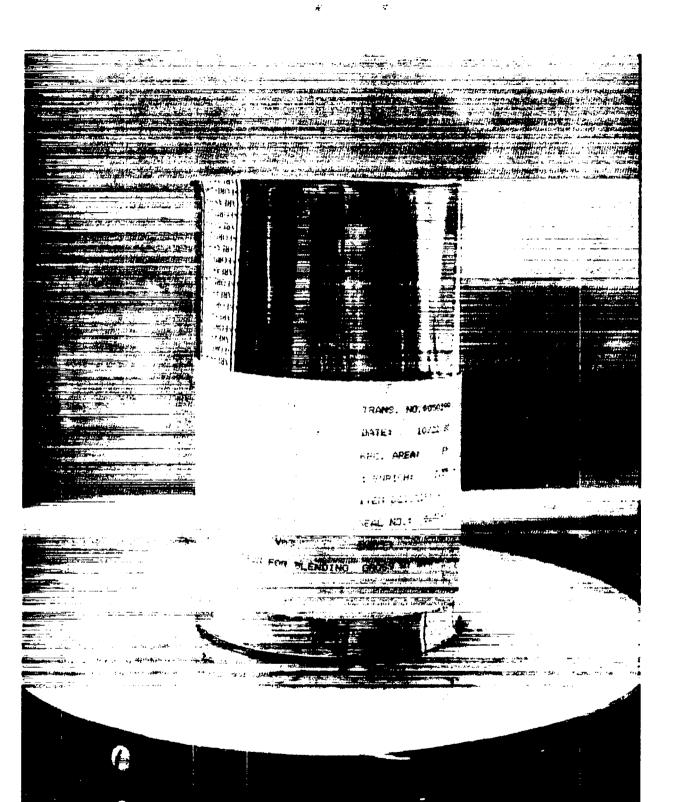


Fig. 3. Can containing plutonium ready for gamma assay.

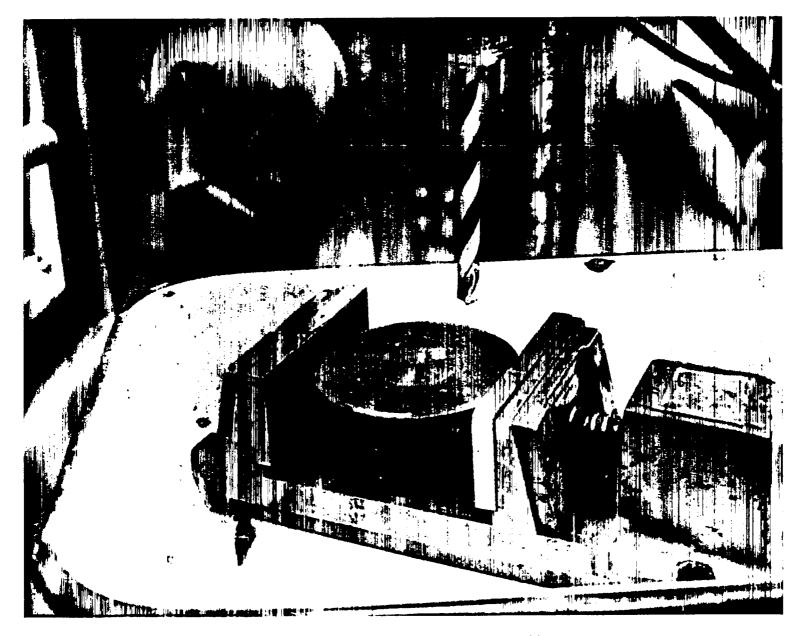


Fig. 4. Button with sample taken by drilling.

After unpacking, a sample was drilled out near the center of the bottom of each button and analyzed (Fig. 4). 'The ²³⁸Pu and ²⁴¹Am concentrations were determined by radiochemistry; the ²³⁹Pu, ²⁴⁰Pu, ²⁴¹Pu, and ²⁴²Pu distributions, by mass spectrometry. The methods were checked with standards from the NBS SRM series for isotopic analysis. The standard deviation in these analyses is <0.2%.² The sample for chemical assay must be representative of the entire button. A typical button is shown before cleaning in Fig. 5. Fifteen 5-g samples were taken from button HRA 147215 to check homogeneity.³ Twelve samples consisted of metal turnings from four main holes (Fig. 6). The other three samples consisted of pieces sheared off the button with a hydraulic chisel at many locations. The data are summarized in Table I. Clearly, the button is homogeneous, and the normal samples from the center of the bottom are representative.

The total plutonium in a button was calculated by dissolving portions of the drilled sample and assaying for total plutonium by controlled potential coulometry. The precision of this assay is >0.1%.⁴ By calculating grams of plutonium per grams of sample, multiplying by the button weight, and adding the plutonium in the residues, a plutonium value was obtained.

The procedure for determining the plutonium value was as follows:

- 1. The packaged weight of the can and its contents were verified by reweighing.
- 2. The plutonium isotopic distribution and the americium concentration were determined by gamma spectrometry.
- The total power of the packaged button was measured with a calorimeter.
- 4. The can was opened and the button was unpackaged, cleaned, and weighed. All loose oxide was collected and weighed. The plutonium in the packaging material was determined by neutron counting.
- 5. A small sample was drilled out near the center of the bottom of the button. The analytical group determined the isotopic distribution, the americium concentration, and the plutonium factor by assaying this material.

The specific power was determined from the isotopic distribution and americium concentration by a computer program.* The plutonium value was then

^{*}This information provided by T. E. Sampson of Los Alamos National Laboratory, December 29, 1981.



Fig. 5. Typical plutonium button before cleaning.

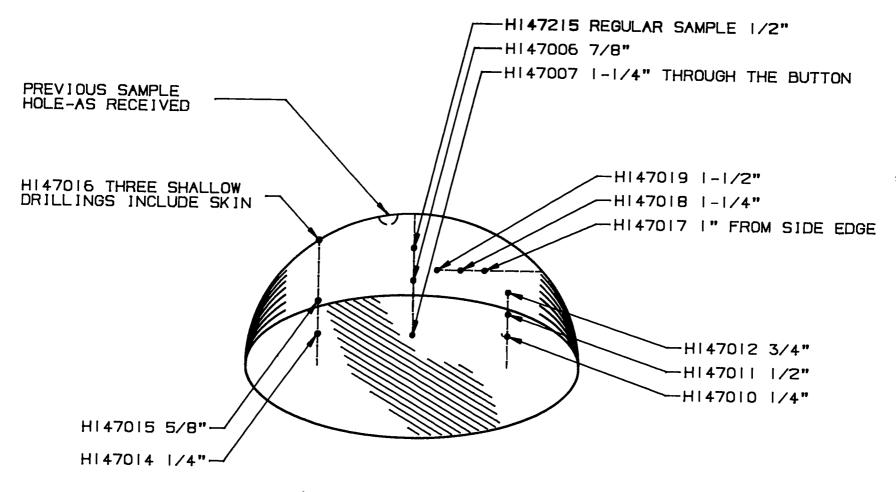


Fig. 6. Multiple sampling of button HRA 147215.

Sample ID							D., 1	alue,g ⁸
10	••••••		leight Perce			241Am,	Chem. Assay	liue,g
HRA 147+	238	239	240	241	242	ppm	+Calor.	Pu Factor
215	0.0081	94.09	5.68	0.202	0.021	1320	2140	2150
006	0.0082	94.10	5.67	0.199	0.020	1330	2139	2147
007	0.0061	94.08	5.69	0.202	0.020	1320	21 39	2146
010	0.0082	94.08	5.69	0.201	0.020	1370	2134	2147
011	0.0085	94.09	5.68	0.200	0.020	1 39 0	2131	2147
012	0.0083	94.09	5.68	0.203	0.021	1370	2134	2148
014	0.0083	94.08	5.69	0.201	0.020	1360	2135	2147
015	0.0083	94.09	5.68	0.199	0.019	1360	2134	2148
016	0.0083	94.09	5.68	0.200	0.021	1350	2134	2136
017	0.0084	94.10	5.67	0.200	0.020	1360	2135	2148
018	0.0086	94.09	5.68	0.199	0.020	1390	2135	2143
019	U.0079	94.09	5.68	0.203	0.021	1320	2141	2147
020	0.0088	94.10	5.68	0.200	0.019	1330	2136	2147
021	0.0081	94.09	5.68	0.200	0.020	1310	2141	2143
U22	0.0079	94.08	5.68	0.203	0.022	1300	2143	2150
x	0.0083	94.0 9	5.68	0.201	0.020	1350	21 37	2146
.s.d. (%)	±3 .0	± 0.00%	±0.1%	±0.7%	±4.0%	± 2.1%	± 0.2%	± 0.2%

TABLE I DATA FROM MULTIPLE SAMPLING OF HRA 147215

*SNM value is 2148 g.

calculated by dividing the total power by the specific power. This resulted in two independent values: one determined by calorimetry and gamma spectrometry; the other, by calorimetry and chemical analysis. A third plutonium value was obtained by multiplying the plutonium factor by the button weight and adding the plutonium in the packaging and that removed as oxide during button cleanup.

III. RESULTS

The approximate distributions of 240 Pu and 241 Am in each button are summarized in Table II. Plutonium values obtained by all three methods differed by <0.7%, as shown in Table III. The average value from three methods, plus the declared value for 409 buttons, was 798521 ± 1396 g, which is equivalent to a standard deviation of 0.2%. The assay of 238 Pu in the first series that we examined, designated the EXCM, was higher by mass spectrometry due to contamination by 238 U. Radioassay determinations were used for 238 Pu on all subsequent chemical assays. The bias was apparently eliminated. The EXCM Series was not included in the totals because of this bias; however, the declared value and the values derived from calorimetry plus gamma spectrometry and from the plutonium factor are in good agreement.

The americium value determined by gamma spectrometry was consistently 10% higher than that determined by radioanalysis. This bias caused a low total plutonium value, which was corrected by dividing all gamma values by an instrument calibration factor of 0.982. We believe that this factor is due to the programming of this particular instrument and could probably be eliminated in another instrument. If the americium concentration was less than 400 ppm, one of the two peaks used to determine the ²⁴¹Am/²³⁹Pu ratio vanished. With our computer program, this caused an erroneously high americium result and, consequently, a low total plutonium value. We substituted a value of 400 ppm americium or used the radioassay value for americium in calculating the total plutonium in such cases, and because the instrumentation factor was largely due to high americium values, we did not apply the factor in such cases. The 2^{40} Pu values at concentrations <3% were usually about 20% higher than normal with this instrument. We had not anticipated receiving material this low in 240 Pu, and we did not have standards in this concentration range for calibration. In addition, for these buttons the 238 Pu concentrations were extremely low (0.005%) and the 241 Am concentrations also were <400 ppm. Our total plutonium values were low for these buttons; the calorimetry/chemical isotopic, plutonium-factor methods and declared values also

were not in good agreement. Therefore, the buttons containing <3% ²⁴⁰Pu are not included in the summary of results. We believe that a gamma instrument programmed for this material would yield acceptable results.

TABLE II

THE APPROXIMATE DISTRIBUTIONS OF ²⁴⁰Pu AND ²⁴¹Pu IN METAL BUTTONS

Series	Composition				
ID	²⁴⁰ Pu (wt%)	²⁴¹ Am (ppm)			
HRA 114	20-14	8000			
HRA 99	10	5000			
HRA 131	8-10	6000			
ARF 147	6	1500			
EXCM	6	1200			
ARF 458	6	100-1500			
ARF 469	6	<500			
ARF 437	6	<500			
ARF 484	6	<400			
ARF 455	5-6	100-300			
ARF 452	4-7	100-500			
ARFSO	<2.5	<500			
ARF 120A	<2.5	<500			

TABLE III

SUMMARY OF RESULTS IN THE DETERMINATION OF Pu VALUES

Series	Number		Pu Value (g	<u>;)</u>			
ID	Buttons	Declared	Cal+Gamma	Cal+Chem	Pu Factor	Average	sd (g) (%)
HRA 114	27	46714	45410	47213	46700	46784	±301 (0.6)
HRA 99	25	88773	88870	88831	86816	88870	±112 (0.1)
HRA 131	26	53365	53393	53334	53236	53382	±151 (0.3)
ARF 147	60	124829	124313	124096	124576	124454	±318 (0.3)
EXCM ^a	21	68389	68614	67773	68498	68068	±869 (1.3)
ARF 458	57	108769	108042	108461	108785	108514	±348 (0.3)
ARF 469	35	67932	67116	67622	68235	67726	±478 (0.7)
ARF 437	49	94681	94439	93953	95151	94556	±449 (0.5)
ARF 484	12	24093	23773	23971	24134	23993	±162 (0.7)
ARF 455	23	43855	43503	43740	43997	43774	±209 (0.5)
ARF 452	23	44270	43811	44217	44477	44194	±279 (0.6)
HRA 12DA	52	102719	102844	102597	102597	102719	±140 (0.2)
Totals	409	800000	796814	798035	799235	798521	±1396 (0.2)

^aEXCM is not included in the totals because the ²³⁸Pu was determined by mass spectrometry, not radiochemistry. This caused low Pu values for the Cal+Chem method.

The average composition of the 45 buttons of the HRA 99 Series is 0.0310 wt% ²³⁸Pu, 90.50 wt% ²³⁹Pu, 8.79 wt% ²⁴⁰Pu, 0.615 wt% ²⁴¹Pu, 0.75 wt% ²⁴²Pu, and 4950 ppm americium. The isotopic distribution for the individual buttons, as determined by gamma spectrometry, is listed in Table IV. The precision of the assay increases with time and with the concentration of the isotope. For example, the estimated precision of the ²³⁸Pu measurement increases from 8.3% at 4.4 h to 2.5% at 14.0 h, and to 1.8% at 40 h for 0.03 wt% 238 Pu. The precision of the 240 Pu assay, based on a much weaker energy peak, increased from 4.2% to 2.1% to 1.2% over the same time period for buttons ~9 wt% ²⁴⁰Pu. On the other hand, the precision of the measurements of both the ²³⁹Pu and ²⁴¹Pu was >0.5% in <3 h. The average assay time for the HRA 99 Series was 10.3 h. The averages for the isotopic data, as determined from the gamma assay, are summarized in Table V. These include the isotopic distribution, the calculated power per gram of plutonium in the button, and the per cent of the specific power contributed by each isotope. The plutonium value is obtained by dividing the power determined by calorimetry by the specific power.

Because the per cent of the total power contributed by each isotope and the precision of each measurement are known, the precision of the contribution of each isotope to the calculation of the plutonium value can be estimated. The 238 Pu contributes 5.61% to the total power even though the concentration is only 0.30%. The estimated precision of the 238 Pu assay is equivalent to a precision of 0.30% in determining contribution to the total power. The major isotope in the buttons is 239 Pu. Because the power generated by individual 239 Pu atoms is far less per gram than that generated by either 238 Pu or 241 Am, the contribution for the power, 55.75%, is much less than the concentration of the isotope. The estimated precision due to the 239 Pu gamma assay is equivalent to 0.17% of the power.

The ²⁴⁰Pu contributed 19.91% of the power and the estimated precision is 0.67%. The power from the ²⁴¹Pu is 0.67% and the precision is >0.01%. The ²⁴²Pu does not contribute significantly to the total power. Finally, the contribution from ²⁴¹Am is 18.06% of the power with an estimated precision of 0.11%. If we assume that the precision of the calorimetric measurement is at least 0.3%, the estimated precision of the plutonium value by gamma spectrometry and calorimetry should be 0.8%. The precision of the ratio of the gamma/calorimetry derived plutonium value to the declared value is actually 0.7%.

TABLE IV

THE ISOTOPIC DISTRIBUTION OF THE HRA 99 SERIES

BY GAMMA SPECTROMETRY ASSAY

Los Alamos			. .		D	
ID	220	220		pic Weight		
HRA 99+	238	239	240	241	242	ppm Am
JA 07	0.0267	90.94	8.42	0.5542	0.0620	4840
X 08	0.0264	91.56	7.83	0.5334	0.0548	4920
NX07	0.0189	90.09	9.26	0.5659	0.0710	5120
NX08	0.0241	90.46	8.86	0.5925	0.0706	5120
NX-9				0.4584		
MA- 9	0.0284	91.47	8.00	0.4364	0.0482	3720
NX09	0.0257	90.24	9.06	0.6052	0.0741	5260
NX11	0.0237	90.45	8.84	0.6152	0.0731	5420
NX12	0.0296	91.06	8.25	0.6032	0.0660	5080
NX13	0.0261	90.47	8.83	0.5993	0.0711	5190
NX14	0.0283	90.85	8.46	0.5989	0.0675	5070
	0.0205	70.05	••••	0.0707	•••••	5070
NX15	0.0408	89.12	9.93	0.7938	0.1092	6520
NX16	0.0459	89.04	9.91	0.8842	0.1215	7160
NX17	0.0464	88.55	10.35	0.9138	0.1327	7410
NX18	0.0471	88.44	10.51	0.8801	0.2301	7240
NX19	0.507	88.97	9.94	0.9144	0.1264	7420
NX20	0.0504	88.74	10.17	0.9138	0.1298	7310
NX21	0.0481	88.35	10.56	0.9139	0.1360	7420
NX22	0.0511	88.32	10.55	0.9540	0.2060	7700
NX24	0.0250	91.38	7.99	0.5456	0.0547	3610
NX35	0.0267	91.03	8.35	0.5389	0.0597	4190
NX36	0.0276	90.90	8.48	0.5362	0.0605	4160
NX37	0.0261	91.36	8.02	0.5377	0.0569	4280
NX38	0.0258	90.53	8.86	0.5295	0.0629	4190
NX 39			7.41		0.0456	4410
NX40	0.0341	92.04		0.4742	0.0430	4250
AZ4U	0.0266	90.43	8.95	0.5350	0.0044	4230
NX41	0.0271	90.82	8.53	0.5572	0.0634	4370
NX42	0.0285	90.32	9.02	0.5643	0.0687	4450
NX43	0.0328	90.11	9.10	0.6791	0.0837	5360
NX44	0.0289	90.30	8.99	0.6084	0.0738	4700
NX45	0.0300	90.36	8.97	0.5696	0.0688	4530
NX 80	0.0273	91.86	7.55	0.5053	0.0498	3000
MX81	0.0245	91.51	7.83	0.5812	0.0598	6470
NX86	0.0254	90.67	8.71	0.5364	0.0625	4150
NX87	0.0273	91.13	8.26	0.5305	0.0580	4060
NX88	0.0260	90.79	8.59	0.5314	0.0609	4050
N X89	0.0276	9 0.84	8.54	0.5309	0.0605	3940
NX90	0.0249	90.76	8.62	0.5305	0.0611	4090
NX91	0.0257	90.91	8.48	0.5295	0.0597	4120
NX92	0.0272	90.91 91.00	8.39	0.5279	0.0588	4000
NX93	0.0253	90.65	8.73	0.5279	0.0627	4000
MA73	0.0233	50.03	0./3	0.000/	0.0027	4110
X X94	0.0266	9 0.30	9.03	0.5727	0.0698	4350
NX95	0.0287	89.65				
WX95			9.65	0.5928	0.0782	4650
X01	0.0266	92.50	6.96	0.4685	0.0419	2910
	0.0461	91.96	7.34	0.6028	0.0575	4030
MX99	0.0275	91.06	8.31	0.5353	0.0590	4270

TABLE V

ISOTOPIC DATA AVERAGES FOR THE HRA 99 SERIES

Isotope	Isotope (W/g)	Isotopic Distribution	Specific Power (W/g) Pu	Per Cent of Total Power
238	0.5672	0.03098%	1.7569×10^{-4}	5.61
239	0.001929	90.50%	17.46×10^{-4}	55.75
240	0.007098	8.786%	6.236 x 10 ⁻⁴	19.91
241	0.003390	0.6152%	0.2085×10^{-4}	0.67
242	0.0001159	0.07584%	0.0009×10^{-4}	0.00
Am-24 1	0.1142	pm بور 4950	5.654×10^{-4}	18.06
Total			31.3253 x 10 ⁻⁴	100.00

TABLE VI

THE RATIOS OF ISOTOPIC VALUES; GS/CHEM FOR THE HRA 99 SERIES

TD		0.00		y/Chemical A		
ID BIRA 99+	238	239	240	241	242	Am
	0.988	0.995	1 050	1 000	0 775	1 000
A07 X08			1.059	1.008	0.775	1.09
	0.912	0.999	1.022	0.994	0.481	1.07
X07	1.129	1.002	0.984	1.006	1.044	1.00
1208	1.053	1.001	0.986	1.016	1.008	1.05
X-9	1.208	0.997	1.039	0.987	1.461	1.19
X09	0.991	0.997	1.035	1.015	1.029	1.18
X11	0.912	1.001	0.991	1.007	0.897	1.05
X12	1.095	1.004	0.959	1.010	0.970	1.04
X13	0.966	0.997	1.029	1.011	1.046	1.05
X14	1.053	1.002	0.978	0.999	0.937	1.07
X 15	1.020	0.998	1.022	1.009	0.628	1.04
X16	0.997	1.004	0.971	1.013	0.552	1.06
X17	0.967	1.001	0.997	1.009	0.561	1.08
X18	1.025	0.998	1.028	1.008	0.593	1.08
X19						
A17	1.056	1.006	0.962	1.018	0.543	1.08
X20	1.050	1.003	0.984	1.013	0.557	1.06
X21	1.023	0.999	1.017	1.004	0.575	1.07
X22	1.022	1.002	0.993	1.015	0.792	1.05
X24	0.967	1.002	0.980	1008	0.545	1.07
X35	1.068	1.001	0.987	1.002	1.328	1.09
X36	1.028	1.000	1.002	1.002	1.345	1.06
X37	1.019	1.005	0.949	1.000	1.264	1.07
X38	0.968	0.995	1.048	0.993	1.430	1.11
X39	1.004	1.000	1.004	1.000	0.449	1.10
X40	1.030	0.995	1.054	0.992	1.431	1.10
IX41	1.062	1.000	0.995	1.000	1.321	1.08
X42						
	1.032	0.996	1.045	0.994	1.164	1.10
X43	1.066	1.002	0.983	1.005	1.087	1.07
X44	1.006	0.998	1.017	0.998	1.229	1.08
X45	1.077	0.997	1.033	0.979	1.324	1.03
X80	1.008	0.996	1.058	0.995	0.754	1.07
X81	0.720	1.000	1.003	0.993	1.107	1.10
X86	0.999	0.997	1.034	1.005	1.421	1.08
X87	1.092	1.001	0.987	1.006	1.350	1.06
X88	1.040	0.998	1.026	1.010	1.128	1.07
X89	1.098	0.899	1.020	1.009	1.407	1.04
X90	0.995	0.998	1.024	1.002	1.421	1.10
X91	1.046	0.999	1.013	1.010	1.389	1.13
X92	1.086	1.000	1.002	1.001	1.368	1.05
X93	0.998	0.997	1.033	1.005	1.394	1.03
N0/	0.00/	0.997	1.033	1.005	1 214	1.05
X94	0.994				1.316	
X95	1.026	0.992	1.079	1.002	1.397	1.05
X 98	0.989	1.000	1.006	1.000	0.607	1.13
XO 1	1.020	0.998	1.040	1.002	0.492	1.06
X 99	1.069	1.001	0.984	0.998	1.312	1.04
Ĩ	1.021	0.999	1.011	1.004	1.027	1.07
.d.(%)	±0.07	±0.003	±0.029	±0.008	±0.345	±0.03

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The ratios of the isotopic values determined by gamma spectrometry to those determined by chemical assay are given in Table VI. The ²³⁸Pu by gamma assay averages 2.1% high, which is equivalent to a 0.1% lower total plutonium value. The 239 Pu ratio is 0.999 so that the average increase in the plutonium value by gamma assay is <0.1% (0.056%). The ²⁴⁰Pu by gamma assay is 1.1% higher than by mass spectrometry, which represents a 0.2% lower total plutonium value. The ²⁴¹Pu and ²⁴²Pu ratios (<0.01%) are not large enough to cause significant changes in the plutonium value. The major differences are due to the high bias in the americium value, 7.9%. This will cause the average plutonium value calculated by the gamma data to be 1.4% lower than that derived from the chemical assay. The total calculated difference in the plutonium value is 1.71% lower for the gamma assay for this series of buttons. In determinations for a large number of buttons, the average was, indeed, 1.82% lower than the declared value. The difference between calculated and actual bias may be due to a small additional bias in the calorimetry.

The plutonium values for the HRA 99 Series are summarized in Table VII. Although individual assays vary, the total plutonium values are in good agreement. These totals are also within 0.2% of the declared value. Where the gamma/calorimetry values have been corrected for instrument bias (1.018 times the raw value), the three methods are equivalent.

IV. CONCLUSIONS

The calorimetry/gamma spectrometry method for determining the plutonium content is attractive because it is rapid, nondestructive, and nonintrusive. If the instruments are calibrated with similar standards, if the same program is employed, and if reported data are standardardized, this technique will provide an excellent procedure for shipper-receiver determination of plutonium value. At Los Alamos, Group Q-1 has completed a second gamma system incorporating the changes indicated by this study. It can be used for routine checks of plutonium content in buttons, oxide, and scrap. If it is used as planned, it should improve resolution of the plutonium content of all plutonium-containing materials.

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		Pu Value				Ratios	
IL.		Calor.+	Calor.+	Pu Factor +	Gamma	Ches	Pu Factor
HRA 99+	Declared	Gamma	Chem.	Oxide + n.c.	Declared	Declared	Declared
JAD7	920	921	927	922	1.001	1.008	1.002
XOB	1775	1786	1775	1771	1.006	1.000	0.998
WX07	2010	2021	1997	2010	1.006	0.994	1.000
XXOS	1945	1959	1952	1937	1.007	1.004	0.996
MX-9	2059	2027	2071	2057	0.985	1.006	0.999
X X09	1852	1853	1892	1853	1,000	1.022	1.000
NO.11	2006	2019	2000	2006	1.006	0.997	1.000
NX12	2041	2053	2033	2041	1.006	0.996	
							1.000
NX13	2020	2017	2012	2019	0.999	0.996	1.000
NX14	1927	1948	1944	1921	1.011	1.009	0.997
EX15	2035	2026	2026	2033	0.996	0.996	0.999
MX16	2000	2004	1997	1994	1.002	0.999	0.997
MA 17	1979	1994	199	1976	1.008	1.007	0.999
MX18	1977	1992	1923	1978	1.008	0.973	1.001
MX19	2004	2011	2020	2003	1.004	1.008	1.000
MX20	2047	2050	2056	2032	1,002	1.004	0.993
83.21	2031	1986	2042	2030	0.978	1.005	1.000
ID.22	1992	1987	1989	1974	0.998	0.999	0.991
MX24	2032	2053	2029	2020	1.010	0.999	0.994
NX35	1930	1951	1949	1934	1.011	1.010	1.002
ID:36	1941	1943	1936	1938	1.001	0.997	
#X30 #X37	1998	2014	1936				0.999
				1997	1.008	0.997	1.000
ND.38	1917	1910	1920	1918	0.996	1.002	1.001
WX 39	1953	1949	1950	1955	0.998	0.999	1.001
10. 40	1778	1769	1782	1776	0.995	1.002	0.999
#X41	1949	1956	1951	1946	1.004	1.001	0.999
IN 42	1907	1896	1910	1903	0.994	1.002	0.998
MX43	1841	1844	1844	1838	1.002	1.002	0.998
30 X=4	1969	1970	1969	1969	1.000	1.000	1.000
XX 45	2092	2068	2072	2092	0.989	0.990	1.000
XX8 0	2081	2072	2070	2083	0.996	0.995	1.001
WX81	269	270	267	269	1.004	0.993	1.000
MX86	2063	2070	2075	2057	1,003	1.006	0.997
JCX 67	2070	2073	2066	2069	1.001	0.998	1.000
NX88	2045	2053	2055	2039	1.004	1.005	0.997
10 089	1993	2003	2001	1977	1.005	1.004	0.992
IDX90	2049	2058	2063	2044	1.005	1.004	0.992
MX91	2029	2036	2050	2023	1.004	1.007	0.998
EX92	2029	2036	2050	2023			0.997
MX92 MX93					1,007	1.004	
	2052	2052	2041	2042	1.000	0.995	0.995
EX 94	2063	2065	2059	2057	1.001	0.998	0.997
XX9 5	2059	2032	2047	2057	0.987	0.994	0.999
X X98	2063	2057	2051	2063	0.997	0.994	1.000
X 01	2038	2038	2017	2036	1.000	0.990	0.999
NX9 9	1903	1903	1885	1903	1.000	0.991	1.000
-	1				I <u> </u>		
Σ	86770	86840	86828 <u>.</u>	86613	1.001	1.000	0.999
Δ¥. Δ¥. ¥		70 0.1	58	-157 - 0.2	s.d. ±.007 s.d. 07%	±.008	±.003
			0.1			0.8%	0.3%

TABLE VII

SUMMARY OF Pu VALUES FOR THE HRA 99 SERIES

X₄≈ 86763 ●.d.(%) ± 104 (0.1)

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tabulated much of the data. The metal fabrication section of Group CMB-11 unpackaged the ingots and sampled them; the count room of Group CMB-11 provided the calorimetry; and the analytical group, Group CMB-1, did all the conventional assays. We wish to thank them all for their help and encouragement.

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