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IN QUE FOR ASSAY OF L 10 BOTTLE
GONUM NITRATE II, ADDITIONAL
APPROXIMATIONS AND CONSIDERATIONS

J. E. Gine
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Aerojet Nuclear Company

NATIONAL REACTOR TESTING STATION

Idaho Falls, Idaho — 83401

MASTER

PREPARED FOR THE

U. S. ATOMIC ENERGY COMMISSION

IDAHO OPERATIONS OFFICE UNDER CONTRACT AT(10-1)-1375

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A TECHNIQUE FOR ASSAY OF L-10 BOTTLES OF PLUTONIUM NITRATE, II,
ADDITIONAL MEASUREMENTS AND CONSIDERATIONS

J. E. Cline
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AEROJET NUCLEAR COMPANY

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ABSTRACT

Extensive assay measurements were recently made of L-10 bottles of plutonium nitrate using the technique previously described in IN-1433. The measurements were made at the ARCHO facility at Hanford. One hundred seven bottles were analyzed in this new series. As a result of these analyses and some further considerations of precisions of the technique, minor modifications have been made to the previous analytical techniques.

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A TECHNIQUE FOR ASSAY OF L-10 BOTTLES OF PLUTONIUM NITRATE, II,
ADDITIONAL MEASUREMENTS AND CONSIDERATIONS

I. INTRODUCTION

In the management and safeguarding of fissionable material, it is highly desirable to have available methods of fast, accurate and non-destructive assay for the elements and isotopes present in the materials. A special need has existed for a procedure for the assay of plutonium stored as a solution in "load-out" bottles. Perhaps equally as important as an absolute assay of the material would be a method of identifying bottles. Such identification would rapidly provide information for the purpose of inventory or determination that tampering or material diversion had not occurred.

Presented here is a technique which can provide both an absolute assay of the plutonium content and a means of identification for the bottles. The method uses non-destructive Ge(Li) spectrometry and can yield a complete analysis in approximately 15 minutes. The elements of the technique have been presented previously (1). Included in the present report are some considerations regarding the accuracy of the technique, a description of the current measurements, and the results of the analyses.

II. DESCRIPTION OF THE TECHNIQUE

The basic technique as described in the previous report [1] is included here for the sake of completeness and continuity.

The principal gamma rays emitted in the decay of ^{239}Pu and ^{241}Pu which are used in the analysis are listed in Table I. If there were no absorption of the emitted gamma rays in the plutonium nitrate-nitric acid solution, the measured intensity of the 413-keV gamma ray would be linearly proportional to the total grams of ^{239}Pu present. A similar relation would exist for the 267-keV gamma ray and the ^{241}Pu content. However, severe self-absorption of these relatively low-energy gamma rays does occur and the problem becomes to find a mechanism for determining the absorption which is a function of the plutonium concentration.

TABLE I

<u>Nuclide</u>	<u>Gamma Ray Energy (KeV)</u>
^{239}Pu	129.27
	413.71
^{241}Pu	148.56
^{237}U (in equilibrium with ^{241}Pu)	267.53
^{238}Pu	152.58
^{240}Pu	160.27

Figure 1. Relative gamma-ray intensities per gram ^{239}Pu vs plutonium concentration.

If it is assumed that the plutonium concentration is constant throughout a bottle, that the density of nitric acid is constant from bottle to bottle, and that the wall thickness of the bottle is uniform in a single bottle and constant from bottle to bottle, it is possible to calculate the relationship between the relative counting rate for a gamma ray of a given energy and the plutonium concentration. One can further calculate (1) the relationship between the ratio of the relative counting rates for a low- and a high-energy gamma ray and the plutonium concentration

$$\text{i.e. } \left(\frac{I_{129\text{-keV}}}{I_{413\text{-keV}}} \right)_{\text{observed}} = f(\text{plutonium concentration}) .$$

Since the gamma-ray absorption coefficient in plutonium is quite energy dependent, a measurement of this ratio can be a sensitive measurement of the plutonium concentration.

Figure 1 shows the calculated relative counting rate of the 413-keV gamma ray as a function of plutonium concentration. Shown for comparison is the relative counting rate of the 129-keV transition. Use of the former curve yields more accurate results because of the lower dependence upon concentration. Figure 2 shows the observed ratios of the intensity of the 129-keV transition to that of the 413-keV transition and of the 149-keV transition to the 267-keV transition, both as a function of plutonium concentration. It is possible then to determine the plutonium concentration through a measurement of the ratio of two gamma-ray intensities and using this concentration, to determine the relative counting rate for the 413-keV gamma ray and, hence, determine total ^{239}Pu .

Figure 2. Gamma-ray intensity ratios as a function of plutonium concentration.

III. UNCERTAINTIES AND ACCURACIES

The uncertainties assigned to the results of the assay are easily determined if they are based solely upon the statistical uncertainties in the number of events in the 148-, 267-, and 413-keV peaks. In this propagation, a 1% statistical uncertainty in the ratio of intensities of the 148- and 267-keV gamma rays results in an uncertainty in the plutonium concentration of from 1.7 to 3.3%. An uncertainty of this magnitude in the concentration causes an uncertainty in total plutonium which is nearly negligible when compared with the statistical uncertainty in the intensity of the 413-keV transition.

Inaccuracies arising from other sources could easily dominate the true accuracy of the measurements. Of great importance is the availability of accurate standards with which to calibrate the system. Among the more obvious of other potential sources of inaccuracy are 1) variations in plutonium concentration within the bottle, 2) variations in the thickness of the containment bottle or vessel, 3) variations in liquid level from bottle to bottle, 4) variations in nitric acid density from bottle. These items will be discussed in order.

1) No variations in concentration within a bottle are expected in these solutions and no precipitation of the plutonium from solution has been observed. Hence this is not presently considered to be a serious problem. 2) The thickness of the container walls (nominally 0.100 in. for L-10 bottles) could be changed by 0.050, i.e. by 50% and the resultant change in either measured ratio of gamma-ray intensities would be no more than 1.5% and the measured intensity of the 413-keV transition would change about 3%. Such a variation in the wall thickness is considered extreme and unlikely. 3) Variations in the liquid level between bottles do in fact exist. Such variations would not significantly affect the concentration determination but could affect the total plutonium assay if the detector were located close to the bottle. For this reason, we have used large detectors in these measurements and have located the detector no closer than 2.5 meters from the 1.25-meter tall bottle. 4) If the normality of the nitric acid solution, normally 6, were different from this value by 1 unit, the density of nitric acid would differ by about 3% from the normal value. This would have the effect of changing the observed gamma-ray intensity ratios by less than 1.6% and of changing the observed intensity of the 413-keV transition by only 1.3%.

None of the above sources of error appear to be very large. Therefore, it is felt that the results of the measurements should be precise to $\pm 5\%$, both for plutonium concentrations and for total plutonium. Isotopic ratio measurements have accuracies which are dependent upon the value of the isotopic ratio, but for $^{241}\text{Pu}/^{239}\text{Pu}$ measurements, a precision of about 2% is readily obtained. For $^{238}\text{Pu}/^{239}\text{Pu}$, however, due to the low relative intensity of the 152-keV transition in the data, the precision is considerably less and can be as large as $\pm 30\%$ for relatively low concentrations of ^{238}Pu . The assay for ^{240}Pu , because of the large interference to the intensity determinations of the gamma rays from this isotope from the gamma rays of similar energies emitted by the other

isotopes of plutonium, has very low precision ($\sim 30\%$) and is probably best done by neutron coincidence counting.

IV. MEASUREMENTS

A Ge(Li) spectrometer was transported to the ARHCO facility. The volume of the detector used for these measurements was approximately 30 cc. The experimental configuration is shown in Figure 3. L-10 bottles, some enclosed in the 0.074-inch thick wall Atomic and Space Development Authority (ASDA) stainless jacket, were lowered into position inside the tube which had 0.234-inch thick walls. The gamma rays from the bottles were counted for 400-seconds live time. The 0.25-inch thick plate glass over the window prevented any plutonium contamination from the bottles from entering the room containing the spectrometer and operating personnel. Because of the high gamma counting rates observed in the experiments, as high as 28,000 counts per second, it would have been advantageous to increase the distance from the bottles to the detector. Space limitations, however, prevented this. Therefore, some rather significant corrections had to be made to the measured gamma-ray intensities to account for losses due to random pulse pile up. These losses, unaccounted for by the spectrometer livetimer, amounted to 20% at 26,000 c/s.

A typical pulse-height spectrum of the gamma rays from an L-10 bottle in this series is shown in Figure 4. A total of 107 bottles was analyzed in the measurements. These covered a wide range of plutonium concentrations and relative isotopic abundances. The data in Figure 4 were taken of a bottle having relative isotopic concentrations of 73.5% ^{239}Pu and 8.9% ^{241}Pu . The peaks in the spectrum corresponding to gamma rays of 129-, 148-, 267-, and 413-keV energy are as indicated. In this example, the statistical accuracy in the integration of the 129-keV peak is limited to nearly 7%. This was caused by the heavy absorption of the 129-keV transition in the stainless jacket and the plate glass and by the large Compton distribution from the very intense 208-keV transition from ^{241}Pu which underlies the 129-keV peak. Therefore, in this particular measurement, a higher degree of accuracy was obtained for the plutonium concentration through the use of the 148- and 267-keV transitions rather than through the 129- and 413-keV transitions. The total ^{239}Pu content was then obtained through the 413-keV transition.

The data from the measurements are shown in Table II. Columns 1 through 6 give the values supplied by ARHCO. Some of these values were measured by them and some were obtained from National Fuel Services (NFS) or ASDA. Bottles marked with a single asterisk denote those which were counted in the ASDA 0.074-inch wall stainless jacket. Columns 7 through 10 give the experimental values obtained in the present analyses from the gamma-ray spectra. Where appropriate, these values have been corrected for absorption in the ASDA jacket, and the corrected values are listed in columns 11 and 12. The average values for a given series of bottles are listed in the corrected-intensity columns. An entry in column 13 indicates that a sample was drawn from the bottle and mass

TABLE II

ARHCO DATA						NRTS DATA										
Bottle Number	Ship T.U.	Conc g/l	Isotopic Abundance			Uncorrected				Absorbion Corrected		ICPP Conc g/l	ARHCO New Value T.U.	Uncorrected 413/g-239	Corrected 413/g-239	ICPP %238
			%239	240	241	148/267	129/148	129/152	413	148/267	413					
NSP-18-1	1091	149.0	88.689	7.911	2.804	4.009	***0.258	2.065	11263			- - -	- - -	11.64		- - -
2	1276	149.0	88.689	7.911	2.804	4.527	0.373	- - -	13028			152.3	- - -	11.51		0.25
3	1287	149.0	88.689	7.911	2.804	4.427	0.427	- - -	13203			- - -	- - -	11.57		- - -
4	1290	149.0	88.689	7.911	2.804	4.278	0.397	1.565	13271			- - -	- - -	11.62		- - -
5	1290	149.0	88.689	7.911	2.804	4.562	0.413	- - -	13121			- - -	- - -	11.49		- - -
	- - -	- - -	- - -	- - -	- - -	- - -	**0.403	**1.815	- - -	**4.361		- - -	- - -	- - -	**11.56	- - -
BRP-30-12-1	1431	209.0	82.099	13.222	3.901	3.823	0.227	- - -	12481			- - -	- - -	10.63		- - -
2	1716	209.0	82.099	13.222	3.901	3.824	0.263	- - -	15545			- - -	- - -	11.04		- - -
3	1792	209.0	82.099	13.222	3.901	3.748	0.256	- - -	16127			- - -	- - -	10.97		- - -
4	1804	209.0	82.099	13.222	3.901	3.820	0.265	- - -	16277			- - -	- - -	11.00		- - -
5	1819	209.0	82.099	13.222	3.901	- - -	- - -	- - -	- - -			- - -	- - -	- - -		- - -
6	1826	209.0	82.099	13.222	3.901	3.835	0.244	- - -	16253			- - -	- - -	10.85		- - -
7	1774	209.0	82.099	13.222	3.901	3.706	0.258	- - -	16327			- - -	- - -	11.53		- - -
8	1806	209.0	82.099	13.222	3.901	3.825	0.261	- - -	16247			218.4	- - -	10.97		0.25
9	1780	209.0	82.099	13.222	3.901	3.382	0.275	- - -	16832			- - -	- - -	10.61		- - -
10	1803	209.0	82.099	13.222	3.901	3.730	0.258	- - -	15685			- - -	- - -	10.91		- - -
11	1799	209.0	82.099	13.222	3.901	3.844	0.258	- - -	16096			- - -	- - -	11.22		- - -
12	1450	209.0	82.099	13.222	3.901	3.744	0.274	2.551	15978			- - -	- - -	***13.44		- - -
	- - -	- - -	- - -	- - -	- - -	- - -	**0.258	- - -	- - -	**3.790		- - -	- - -	- - -	**10.97	- - -
YAE-6-63	1739	205.5	73.518	14.977	8.926	3.677	0.112	1.004	18534			- - -	- - -	14.50		- - -
64	1744	205.5	73.518	14.977	8.926	3.729	0.102	0.904	18260			212.3	1715	14.49		1.19
65	1793	205.5	73.518	14.977	8.926	3.758	0.106	1.217	18733			- - -	- - -	14.21		- - -
66	1776	205.5	73.518	14.977	8.926	3.775	0.112	0.834	18449			- - -	- - -	14.12		- - -
*	67	1765	205.5	73.518	14.977	8.926	0.114	1.024	15208	3.775	18113	- - -	- - -	11.71	13.95	- - -
*	68	1785	205.5	73.518	14.977	8.926	0.112	0.876	15443	3.764	18393	- - -	- - -	11.77	14.02	- - -
*	69	1747	205.5	73.518	14.977	8.926	0.120	0.986	14518	3.802	17291	- - -	- - -	11.31	13.47	- - -
*	70	1775	205.5	73.518	14.977	8.926	0.114	0.846	15308	3.713	18232	- - -	- - -	11.73	13.97	- - -
*	71	1687	205.5	73.518	14.977	8.926	0.117	0.998	15145	3.693	18038	- - -	- - -	12.21	14.54	- - -
*	72	1684	205.5	73.518	14.977	8.926	0.127	1.091	13739	3.707	16363	- - -	- - -	11.10	13.22	- - -

ARHCO DATA						NRTS DATA											
Bottle Number	Ship T.U.	Conc g/l	Isotopic Abundance			Uncorrected				Absorbion Corrected		ICPP Conc g/l	ARHCO New Value T.U.	Uncorrected 413/ g-239	Corrected 413/ g-239	ICPP %238	
			%239	240	241	148/ 267	129/ 148	129/ 152	413	148/ 267	413						
* YAE-6-73	1681	205.5	73.518	14.977	8.926	3.404	0.120	1.067	15142	3.869	18034	- - -	- - -	12.25	14.59	- - -	
	- - -	- - -	- - -	- - -	- - -	- - -	**0.114	**0.986	- - -	**3.759	- - -	- - -	- - -	- - -	**14.10	- - -	
* YAE-15-8	1714	206.1	72.207	15.057	9.926	3.190	0.109	0.779	14315	3.625	17049	- - -	- - -	11.56	13.77	- - -	
* 9	1718	206.1	72.207	15.057	9.926	3.190	0.098	0.797	14652	3.626	17451	- - -	- - -	11.81	14.06	- - -	
* 10	1723	206.1	72.207	15.057	9.926	3.293	0.111	0.822	14147	3.743	16849	- - -	- - -	11.37	13.54	- - -	
* 11	1712	206.1	72.207	15.057	9.926	3.300	0.092	0.775	14908	3.750	17755	- - -	- - -	12.06	14.36	- - -	
* 12	1739	206.1	72.207	15.057	9.926	3.747	0.096	0.896	18169			214.0	1700	14.46		1.08	
* 13	1758	206.1	72.207	15.057	9.926	3.376	0.084	0.660	14582	3.837	17368	- - -	- - -	11.49	13.69	- - -	
	- - -	- - -	- - -	- - -	- - -	- - -	**0.098	**0.788	- - -	**3.721	- - -	- - -	- - -	- - -	**13.98	- - -	
* CDE-9-28	1965	228.6	73.277	16.212	8.475	3.352	0.126	- - -	15412	3.810	18356	- - -	- - -	10.70	12.75	- - -	
* 29	1958	228.6	73.277	16.212	8.475	3.300	0.124	1.194	15758	3.750	18768	- - -	- - -	11.17	13.31	- - -	
* 30	1967	228.6	73.277	16.212	8.475	3.263	0.113	1.524	16041	3.709	19105	- - -	- - -	10.93	13.02	- - -	
* 31	1962	228.6	73.277	16.212	8.475	3.242	0.111	1.173	15670	3.685	18663	- - -	- - -	10.90	12.98	- - -	
* 32	1964	228.6	73.277	16.212	8.475	3.384	0.135	1.823	15503	3.846	18464	236.0	1915	10.77	12.83	0.60	
* 33	1966	228.6	73.277	16.212	8.475	3.309	0.102	- - -	- - -	3.761	- - -	- - -	- - -	- - -	- - -	- - -	
* 34	1906	228.6	73.277	16.212	8.475	3.171	0.164	1.878	15080	3.604	17960	- - -	- - -	10.80	12.86	- - -	
* 35	1907	228.6	73.277	16.212	8.475	3.146	0.126	1.594	14980	3.575	17841	- - -	- - -	10.72	12.77	- - -	
	- - -	- - -	- - -	- - -	- - -	- - -	**0.125	**1.531	- - -	**3.718	- - -	- - -	- - -	- - -	**12.93	- - -	
* CDE-13-55	1702	195.6	71.123	17.016	9.435	3.354	0.104	1.119	13888	3.812	16541	- - -	- - -	11.46	13.65	- - -	
* 56	1709	195.6	71.123	17.016	9.435	3.282	0.110	1.088	14102	3.730	16795	- - -	- - -	11.60	14.82	- - -	
* 57	1644	195.6	71.123	17.016	9.435	3.306	0.108	1.054	13292	3.757	15831	- - -	- - -	11.37	13.54	- - -	
* 58	1507	195.6	71.123	17.016	9.435	3.395	0.122	1.339	11912	3.858	14187	- - -	- - -	11.11	13.23	- - -	
* 59	1338	195.6	71.123	17.016	9.435	3.912	0.106	1.175	12902			203.2	1324	13.56		0.85	
* 60	572	195.6	71.123	17.016	9.435	3.457	0.123	- - -	3653	3.929	4351	- - -	- - -	8.97	***10.69	- - -	
	- - -	- - -	- - -	- - -	- - -	- - -	**0.112	**1.155	- - -	**3.833	- - -	- - -	- - -	- - -	**13.56	- - -	

ARHCO DATA						NRTS DATA										
Bottle Number	Ship T.U.	Conc g/l	Isotopic Abundance			Uncorrected				Absorption Corrected		ICPP Conc g/l	ARHCO New Value T.U.	Uncorrected	Corrected	ICPP %238
			%239	240	241	148/267	129/148	129/152	413	148/267	413			413/g-239	413/g-239	
30-11-195	1977	231.1	89.403	9.442	1.013	4.102	0.994	- - -	18074			- - -	- - -	10.27		- - -
30-12-196	1923	1	89.403	9.442	1.013	4.595	1.015	- - -	18164			- - -	- - -	10.55		- - -
197	790	93.5	89.403	9.442	1.013	6.157	0.896	- - -	8735			- - -	- - -	12.47		- - -
198	1847	215.8	89.403	9.442	1.013	4.496	1.015	- - -	17894			- - -	- - -	10.84		- - -
199	1784	209.2	89.403	9.442	1.013	4.524	0.876	- - -	18574			- - -	- - -	11.61		- - -
31-1-200	1883	228.8	89.403	9.442	1.013	3.960	0.828	- - -	16309			- - -	- - -	9.62		- - -
-3-201	1962	213.9	89.403	9.442	1.013	4.031	1.126	- - -	18329			- - -	- - -	10.37		- - -
-202	2032	234.6	89.403	9.442	1.013	3.731	0.884	- - -	18301			239.6	- - -	10.17		0.060
	- - -	- - -	- - -	- - -	- - -	- - -	**0.954	- - -	- - -			- - -	- - -	- - -		- - -
Man-29-8-																
54	1711	196.9	86.446	11.588	1.653	3.958	0.599	- - -	16040			- - -	- - -	10.84		- - -
56	1635	218.2	86.446	11.588	1.653	4.004	0.585	- - -	15684			- - -	- - -	11.08		- - -
61	2084	256.3	86.446	11.588	1.653	4.071	0.628	- - -	18909			- - -	- - -	10.55		- - -
62	1882	213.3	86.446	11.588	1.653	3.781	0.703	- - -	17256			217.4	- - -	10.59		0.060
30-3-141	1692	217.0	86.446	11.588	1.653	3.937	0.633	- - -	15599			- - -	- - -	10.65		- - -
5-152	1736	206.9	86.446	11.588	1.653	3.789	0.668	- - -	16636			- - -	- - -	11.07		- - -
154	1753	217.3	86.446	11.588	1.653	4.148	0.633	- - -	15091			- - -	- - -	9.95		- - -
155	1817	214.0	86.446	11.588	1.653	3.938	0.616	- - -	17312			- - -	- - -	11.01		- - -
156	1763	216.9	86.446	11.588	1.653	4.239	0.686	- - -	16410			- - -	- - -	10.76		- - -
6-162	690	139.7	86.446	11.588	1.653	4.707	0.541	- - -	6595			- - -	- - -	11.05		- - -
	- - -	- - -	- - -	- - -	- - -	- - -	**0.629	- - -	- - -			- - -	- - -	- - -		- - -
DAA-31-4-																
11	1493	167.5	85.256	11.215	2.997	4.011	0.338	- - -	14875			- - -	- - -	11.68		- - -
12	1433	167.5	85.256	11.215	2.997	4.290	0.393	- - -	14605			- - -	- - -	11.95		- - -
13	1393	167.5	85.256	11.215	2.997	3.925	0.359	- - -	13656			169.6	- - -	11.49		0.35
15	630	167.5	85.256	11.215	2.997	4.162	0.388	- - -	5976			- - -	- - -	11.12		- - -
16	763	167.5	85.256	11.215	2.997	***4.581	0.403	- - -	6345			- - -	- - -	***9.74		- - -
17	1491	167.5	85.256	11.215	2.997	4.276	0.320	- - -	14569			- - -	- - -	11.46		- - -
19	1465	167.5	85.256	11.215	2.997	4.253	***0.120	- - -	14028			- - -	- - -	10.59		- - -

Figure 3. Drawing of counting configuration.

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Figure 4. Pulse-height spectrum of the gamma rays from an L-10 bottle of plutonium nitrate.

spectrometry measurements were made on the sample to determine the plutonium concentration and relative isotopic abundances. The value of the plutonium concentration as determined from these measurements is given in column 13. The relative abundance for ^{238}Pu as determined by the Idaho Chemical Processing Plant (ICPP) is listed in the last column. The relative abundances for the other Pu isotopes were not significantly different from those listed in columns 4 through 6 and are not listed. For six bottles, those indicated by values in column 14, new total plutonium values were obtained. These were obtained by ARHCO through reweighing of the solution and a determination of plutonium concentration in units of grams Pu/gram solution. The values for the uncorrected and corrected intensity of the 413-keV transitions per gram of ^{239}Pu are given in columns 15 and 16, respectively. These values are based on a value for total plutonium as given in column 2 and a value for the relative abundance of ^{239}Pu as given in column 4.

As can be seen from the data in Table II, the average deviation of any individual value of $[I_{148}/I_{267}]$ or of $[I_{413}/\text{gram } ^{239}\text{Pu}]$ in any group from the average value for that group is about 2%. This was to be expected on the basis of arguments presented in Section III, above. Figure 5 shows a plot of the measured intensity ratio, $[I_{148}/I_{267}]$, as a function of plutonium concentration. The values shown on the figure represent the average values in Table II for series of bottles whose concentrations have been verified by chemical analysis at the ICPP. These individual points are identified as shown on the figure. The curve drawn through these values is a calculated curve, normalized to the data. Figure 6 shows the data on 23 YKH and MAN series bottles whose reported plutonium concentrations were not verified. The curve is that from Figure 5. The reason for this large scatter of points off the curve is not known.

Figure 7 shows the intensity $[I_{413}/\text{gram } ^{239}\text{Pu}]$ as a function of plutonium concentration. Again, the values represent the averages for a series of bottles whose concentrations had been verified. The solid curves represent calculated shapes normalized to the data. It is interesting to note that the bottles appear to be in two distinct groups, the ASDA bottles (CEW, CDE, and YAE series), and the remainder. The ASDA bottles were contained in the stainless jackets but corrections had been made to account for this extra absorption. Furthermore, in all of the ASDA groups but one, CDE-8 series, at least one bottle had been removed from the container for sampling and was counted in that condition. As can be seen from the data in Table II, the adjusted intensities are in agreement with the unnormalized value from the one bottle counted without the stainless container, this indicates that a proper correction has been made for the absorption of the container. As a result of the apparent anomaly of the ASDA bottles, a redetermination was made of the total plutonium in one bottle from each ASDA series. The new values, as shown in column 14 of Table II, did not differ significantly from the previous values and the apparent existence of the two curves in Figure 7 is as yet unexplained. It indicates, however, that there are some unreported differences in the two types of bottles. The difference could be explained, for instance, if the diameter of the ASDA bottles was less than that for the other bottles. Figure 8 shows the results

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Figure 5. Plutonium concentrations vs 148-keV/267-keV intensity ratio - experimental values.

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Figure 6. Plutonium concentrations vs 148-keV/267-keV intensity ratio for YKH and MAN series bottles.

Figure 7. $I_{413\text{-keV}}/\text{gram } ^{239}\text{Pu}$ as a function of plutonium concentration, experimental values.

from the unverified individual YKH and MAN series bottles. Although the points are scattered, the grouping of the points appears to be about the curve for the "normal" bottles. Here also, the scatter in the measured values is considered to be anomalously large but no reason for the scatter is known.

Figure 9 shows the relationship between the measured intensity ratio $[I_{148}/I_{129}]$ and the ratio of relative isotopic abundances $[\%^{241}\text{Pu}/\%^{239}\text{Pu}]$. The points on the curve represent averages of all the values for a given series of bottles. A similar curve for the relationship between $[I_{152}/I_{129}]$ and $[\%^{238}\text{Pu}/\%^{239}\text{Pu}]$ is given in Figure 10. The uncertainty flags in Figure 10 are significantly larger than those in Figure 9 because of the low intensity of the 152-keV peak and the relatively large uncertainties in the ^{238}Pu determinations by mass spectrometry.

An additional analysis may be made to provide further information for bottle identification. Figure 11 shows the gamma-ray spectrum from one of the bottles, showing some of the gamma rays from the decay of ^{241}Am . Following chemical separation, the ^{241}Am builds in as a result of the decay of ^{241}Pu . Through an analysis of the data to determine the relative atom abundances of ^{241}Am and ^{241}Pu , it is possible to determine the time since chemical separation. From the data of Figure 11, the age was determined to be 1.68 ± 0.12 years. This value is consistent with the reported data of chemical separation.

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Figure 8. $I_{413}/\text{gram } ^{239}\text{Pu}$ for YKH and MAN series "unknown" bottles.

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Figure 9. Ratio of measured intensities I_{148}/I_{129} as a function of the atom ratio of $^{241}\text{Pu}/^{239}\text{Pu}$.

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Figure 10. Ratio of measured intensities I_{152}/I_{129} as a function of the atom ratio $^{238}\text{Pu}/^{239}\text{Pu}$.

Figure 11. Pulse-height spectrum of gamma rays from Plutonium Nitrate bottle, showing ^{241}Am gamma rays.

V. SUMMARY AND CONCLUSIONS

The technique appears to be suitable for the measurement of large numbers of bottles in plant environments. Information obtained from the measurements can include 1) plutonium concentration (in g/l or g/g), 2) total plutonium, 3) relative isotopic abundance for ^{238}Pu , ^{239}Pu and ^{240}Pu , and 4) date of last chemical separation. The precision with which the first three items can be measured can, with care, approach 1 to 2% except for ^{238}Pu and ^{240}Pu determinations. The accuracy of the measurements is quite dependent upon the availability of adequate standards. Additional measurements which could be made to provide further information are neutron coincidence measurements for ^{240}Pu determination and weighing of the solution during the filling operation, where the measurements are being done in a chemical processing plant, to verify total plutonium content. The technique could equally well be applied to plutonium concentration measurements of liquid streams in processing plants and could provide a continuous monitoring of the stream.

At the ARHCO facility at Hanford, the bottles are stored in steel drums. Other than the support webbing, no material is in the drum. For such containers, it is quite possible that the bottles could be assayed with precisions comparable to those above, while still in the container, slowly rotating the drum during the counting period. This would be quite meritorious in simplifying the handling of the bottles and in decreasing the likelihood of plutonium contamination of the environs.

[1] J. E. Cline, E. B. Nieschmidt, "A Technique for Assay of L-10 Bottles of Plutonium Nitrate", IN-1433 (October 1970).