DP-1607

NONDESTRUCTIVE ASSAY METHODS FOR SOLIDS CONTAINING PLUTONIUM

K. W. MACMURDO
L. W. GRAY
A. GIBBS

E. I. du Pont de Nemours & Co.
Savannah River Laboratory
Aiken, SC 29808

PREPARED FOR THE U. S. DEPARTMENT OF ENERGY UNDER CONTRACT DE-AC09-76SR00001

Printed in the United States of America

Available from

National Technical Information Service
U. S. Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22161

Price: Printed Copy A03; Microfiche A01

DP-1607
Distribution Category: UC-4

NONDESTRUCTIVE ASSAY METHODS FOR SOLIDS CONTAINING PLUTONIUM

K. W. MACMURDO
L. W. GRAY
A. GIBBS

Approved by

J. W. Joseph, Jr., Department Superintendent
Equipment Engineering Department

Publication Date: June 1984

E. I. du Pont de Nemours & Co.
Savannah River Laboratory
Aiken, SC 29808

PREPARED FOR THE U. S. DEPARTMENT OF ENERGY UNDER CONTRACT DE-AC09-76SR00001

ABSTRACT

Specific nondestructive assay (NDA) methods, e.g. calorimetry, coincidence neutron counting, singles neutron counting, and gamma ray spectrometry, were studied to provide the Savannah River Plant with an NDA method to measure the plutonium content of solid scrap (slag and crucible) generated in the JB-Line plutonium metal production process. Results indicate that calorimetry can be used to measure the plutonium content to within about 3% in 4 to 6 hours by using computerized equilibrium sample power predictive models. Calorimetry results confirm that a bias exists in the present indirect measurement method used to estimate the plutonium content of slag and crucible. Singles neutron counting of slag and crucible can measure plutonium to only ±30%, but coincidence neutron counting methods improve measurement precision to better than ±10% in less than ten minutes. Only four portions of a single slag and crucible sample were assayed, and further study is recommended.

CONTENTS

NONDESTRUCTIVE ASSAY METHODS FOR SOLIDS CONTAINING PLUTONIUM

INTRODUCTION

Efforts are underway to provide the Savannah River Plant (SRP) with nondestructive assay (NDA) methods to measure the plutonium content of a variety of solid products, scrap, and waste materials. The program objective is to comply with Department of Energy special nuclear materials accountability measurement requirements at SRP.

Special NDA methods studied include calorimetry, singles neutron counting, coincidence neutron counting, and gamma ray spectrometry (plutonium isotopics). Application of these methods to the assay of slag and crucible (S&C) is described.

SUMMARY

Four portions of a single S&C sample were analyzed for plutonium content by each of three nondestructive analysis (NDA) techniques:

- Calorimetry
- Singles neutron counting
- Coincidence neutron counting

One of the four S&C samples was analyzed destructively to compare to the NDA results. The destructive analysis methods were performed after sample dissolution and include mass spectrometry and gamma spectrometry (plutonium isotopic content), gross alpha counting, coulometry, and isotope dilution mass spectrometry (plutonium content).

Calorimeter Measurements

Calorimetry was used to assay the plutonium content of four portions of an S&C sample of known isotopic content. A Mound Laboratory servo-type calorimeter was used in this study. Calorimetry, used in conjunction with gamma ray spectrometry, can provide accurate and precise analysis of plutonium content of inhomogeneous solids.

Preliminary results also indicate that the 16- to 24- hour analysis times usually required by calorimetry can be reduced to 4 to 6 hours by a computerized equilibrium power predictive technique with no measurable loss of accuracy. A computerized calorimeter system with predictive capabilities was fabricated by Mound Laboratory for SRP and installed in JB-Line in 1981.

The present study provides additional evidence for the existence of a bias in the current method used to estimate the plutonium content of JB-Line slag and crucible. Calorimetric assay should reduce the present about 40% relative uncertainty in assigning plutonium content of slag and crucible containers to about 3% (residual plutonium content of S&C is typically 5% of the plutonium charged to the reduction vessel). Use of the method can be extended to include other inhomogeneous solids, such as cabinet sweepings and some categories of offsite scrap.

Singles Neutron Counting Measurements

The singles neutron counts of four portions of an S&C sample of known isotopic content were made by using a portable well counter fabricated for the Department of Energy by the safeguards group at Brookhaven National Laboratory. The S&C samples exhibited a variable specific neutron emission rate because of the sample inhomogeneity which causes a highly variable (alpha, n) contribution. The variability in the singles neutron count rate on S&C samples on a per gram of plutonium basis was ±30%. Singles neutron counting is not a suitable method for assay of inhomogeneous solids such as S&C.

Coincidence Neutron Counting Measurements

Coincidence counts of the same S&C samples were made by using a dual range coincidence neutron well counter fabricated at Los Alamos National Laboratory. This counter operates with shift register electronics. The coincidence neutron emission rate correlated to actual plutonium content to within ±5.8%. Further development of the technique, underway at LANL, should further improve measurement precision.

Plutonium Isotopic Measurements

The isotopic content of the S&C was measured on dissolved samples by both gamma spectrometry and by mass spectrometry. These isotopic data were used to convert the specific sample power measured by calorimetry to plutonium content. A compilation is included of the plutonium isotopic content of SRP S&C for the years 1973-1978.

Offsite Plutonium Scrap Survey

A representative survey was completed on offsite plutonium-bearing scrap in JB-Line vaults in 1979 with respect to experimental variables which effect NDA measurements. These variables include container size and wall thickness, special nuclear material (SNM) content and isotopic range, and the range of container power levels. Results will be used in NDA instrument specifications.

BACKGROUND — THE JB-LINE PLUTONIUM METAL PRODUCTION PROCESS

In the SRP JB-Line plutonium metal production process, Pu^{3+} is precipitated as PuF_3, oxidized to a PuO_2-PuF_4 mixture, blended with excess calcium metal, and reduced to plutonium metal. After reduction, the button is physically separated from the S&C and cleaned in nitric acid (pickled), rinsed with water, and air dried. The button is weighed, numbered, sampled, and reweighed before being transferred to storage. The pickling process typically dissolves 0.5 to 0.6 g plutonium; this solution is recycled for plutonium recovery.

In the reduction process, a slag remains which is composed largely of CaF_2, CaO, Ca, and traces of unreduced PuO_2 and PuF_4 with small plutonium metal droplets. Average reduction yields are about 95% by weight of plutonium in the PuO_2-PuF_4 charged. Because the molten plutonium penetrates several millimeters into the wall of the MgO crucible, both the slag and crucible are crushed, stored on an interim basis, and then dissolved to recover the residual plutonium.

For process efficiency, the slag and crucible are stored for several months between generation and dissolution. The accountability number ("book value" plutonium content) applied to the stored S&C is the difference between the amount of plutonium charged to the reduction furnace and the after-pickling plutonium button weight. The final button weight is known to $\pm$0.005% relative standard deviation (RSD), but the plutonium charged to the reduction furnace is known to only $\pm$2.0% RSD. The difference between these two quantities leads to a large uncertainty in the book value plutonium content assigned to residual S&C (historically about 40%). Table 1 compares the book value plutonium content and the amount of plutonium recovered after dissolution from a series of clean out operations performed in 1979.

Clearly, more accurate knowledge of the plutonium of the S&C would give better process control in production and recovery operations and would eliminate this contribution to monthly plutonium inventory difference resulting in improved SNM accountability.

TABLE 1

Indirect Book Value Estimates of Plutonium Content of S&C Compared to Recovered Quantities

	Individual Charges			Cumulative Values		
Run	Book Value g Pu Charged	Measured g Pu Recovered	% of Charge Recovered	Book Value g Pu Charged	Measured g Pu Recovered	% of Charge Recovered
1	328	280	85.4	328	280	85.4
2	633	502	79.3	961	782	81.4
3	0	175	-	961	957	99.6
4	0	23	-	961	980	102.0
5	112	226	201.8	1073	1206	112.4
6	69	81	117.4	1142	1287	112.7
7	0	18	-	1142	1305	114.3
8	0	4	-	1142	1309	114.6
9	475	499	105.1	1617	1808	111.8
10	541	692	127.9	2158	2500	115.9
11	473	561	118.6	2631	3061	116.3
12	528	663	125.6	3159	3724	117.9
13	572	573	100.2	3731	4297	115.2
14	0	190	-	3731	4487	120.3
15	684	615	89.9	4415	5102	115.6
16	561	630	112.3	4976	5732	115.2
17	365	336	92.1	5341	6068	113.6
18	676	852	126.0	6017	6920	115.0
19	583	674	115.6	6600	7594	115.1
20	319	523	164.0	6919	8117	117.3
21	378	436	115.3	7297	8553	117.2
22	413	545	132.0	7710	9098	118.0
23	459	541	117.9	8169	9639	118.0
24	301	533	177.1	8470	10172	120.1
25	461	491	106.5	8931	10663	119.4
26	291	385	132.3	9222	11048	119.8
27	493	319	64.7	9715	11367	117.0
28	314	447	142.4	10029	11814	117.8
29	307	457	148.9	10336	12271	118.7
30	402	470	116.9	10738	12741	118.7
31	436	444	101.8	11174	13185	118.0
32	488	673	137.9	11662	13858	118.8
33	434	691	159.2	12096	14549	120.3
34	400	508	127.0	12496	15057	120.5
35	599	689	115.0	13095	15746	120.2
36	731	704	96.3	13826	16450	119.0
37	206	125	60.7	14032	16575	118.1
38	211.5	58	27.4	14244	16633	116.8
39	114	54	47.4	14358	16687	116.2
40	129	83	64.3	14487	16770	115.8

TABLE 1, Contd

Run	Individual Charges: Book Value g Pu Charged	Individual Charges: Measured g Pu Recovered	Individual Charges: % of Charge Recovered	Cumulative Values: Book Value g Pu Charged	Cumulative Values: Measured g Pu Recovered	Cumulative Values: % of Charge Recovered
41	0	33	-	14487	16803	116.0
42	1076	982	91.3	15563	17785	114.3
43	469	611	130.3	16032	18396	114.7
44	455	571	125.5	16487	18967	115.0
45	348	487	139.9	16835	19454	115.6
46	494	736	149.0	17329	20192	116.5
47	412	614	149.0	17741	20804	117.3
48	466	581	124.7	18207	21385	117.5
49	715	864	120.8	18922	22249	117.6
50	565	707	125.1	19487	22956	117.8
51	378	534	141.3	19865	23490	118.2
52	308	605	196.4	20173	24095	119.4
53	312	550	176.3	20485	24645	120.3
54	570	628	110.2	21055	25273	120.0
55	780	342	43.8	21835	25615	117.3
56	346	614	177.5	22181	26229	118.2
57	536	624	116.4	22717	26853	118.2
58	406	689	169.7	23123	27542	119.1
59	430	649	150.9	23553	28191	119.7
60	292	455	155.8	23845	28646	120.1
61	470	388	82.6	24315	29034	119.4
62	589	647	109.8	24904	29681	119.2
63	346	588	169.9	25250	30269	119.9
64	514	583	113.4	25764	30852	119.7
65	498	577	115.9	26262	31429	119.7
66	405	603	148.9	26667	32032	120.1
67	399	608	152.4	27066	32640	120.6
68	343	517	150.7	27409	33157	121.0
69	472	609	129.0	27881	33766	121.1
70	492	684	139.0	28373	34450	121.4
71	425	305	71.8	28798	34755	120.7
72	373	369	98.9	29171	35124	120.4
73	486	561	115.4	29657	35686	120.3
74	517	470	90.0	30174	36155	119.8

Mean % Recovery 121%
Cumulative % Recovery 120%
Range % Recovery 27 to 202%

EXPERIMENTAL METHODS

Nondestructive Analysis Methods

Calorimetry

A Mound Laboratory servo-type calorimeter was used repetitively to measure the sample powers of the four S&C samples. Sample power determination by servo-type calorimetry requires two measurements. First, a baseline or reference calorimeter power level is established by adjusting an electrical supply which heats the reference resistance windings in the sample and reference calorimeter cells containing no sample. This baseline power is determined accurately from voltage and current measurements across the reference windings. Next, a sample is inserted, and the servo system adjusts the power supplied to the reference windings until the new reference power, including the sample power, just equals the original reference power level. The difference represents the sample power. Equilibrium time for the S&C samples was 16 to 24 hours.

Baseline calorimeter power levels were measured before and after sample measurements. The servo-type calorimeter used in this study was designed for sample powers in the four-watt range. Since the S&C measurements involved sample powers in the $\leq$150- milliwatt range, the observed measurement precisions were adversely affected. In the present study, repetitive measurements were performed at two different baseline calorimeter powers: about 7.35 watts and 1.0 watt. The precision of the instrument at four watts is $\pm$0.1% (2σ).

Equilibrium Power Level Prediction

The approach of measured sample power to its equilibrium level is represented mathematically as a sum of exponential functions which depend on sample size, matrix and heat capacity, and on details of the calorimeter construction. DOE has funded Mound Facility and Rocky Flats Plant to develop real-time prediction of calorimetric equilibrium. Algorithms have been developed to predict calorimetric equilibrium conditions with a reduction of measurement time of a factor of 2 to 3 with no loss of measurement precision.

Predictive codes had not previously been applied to samples measured in servo-type calorimeters. In the present study, power level measurements were begun about 30 minutes after sample insertion and repetitive measurements taken manually at 2- or 3-minute intervals for 6 to 8 hours. The power level data were entered manually into the laboratory PDP 11/34 computer to test the predictive models provided by both Mound Facility and Rocky Flats Plant. The predicted equilibria were compared to those measured after 16 to 24 hours.

Singles Neutron Well Counting

The singles (total, noncoincident) neutron emission rates of the four S&C samples were measured in a portable shielded neutron counter with a cylindrical well of 4.5-inch diameter by 4.75-inch height. Six BF_3 neutron tubes are inserted into a polyethylene moderator block which surrounds the counting well. A standard scaler/timer was used to measure counting intervals and recorded neutron pulses. The correlated counts circuitry was not used in this study because the high neutron emission rate caused erratic results.

Coincidence Neutron Counting

The coincidence neutron counter used in this study is a dual range counter utilizing shift register circuitry. Dual range capability is accomplished by insertion of a cadmium liner around the 20 ^3He tubes and polyethylene moderator. The well on this instrument is 7-inch diameter and a useable depth of 16-inches. A Hewlett Packard 97S calculator is interfaced to the instrument which automatically calculates results in grams of plutonium.

Destructive Analysis Methods

Sampling and Dissolution Procedures

In this study, a single magnesium oxide reduction crucible and its contents were divided physically into three parts — the plutonium metal button, one portion containing mostly slag, and one portion containing mostly crucible. The button was cleaned (pickled) in dilute HNO_3, drill sampled, and weighed according to normal JB-Line procedures. The drilled button sample was dissolved in 6M HCl and measured for plutonium content (except Pu-242) by gamma spectrometry (see section entitled "Gamma Spectrometry"). Portions of the bottom sample were also later analyzed for isotopic content by mass spectrometry (except Pu-238) for comparison purposes. The isotopic data were used to calculate the specific sample power (watts per gram plutonium) to convert the measured sample power into grams plutonium and to calculate the specific alpha activity of the plutonium sample.

The sample composed mainly of slag and the sample composed mainly of crucible were manually crushed, and each was divided into two portions of approximately equal weight. The four samples labeled C1, C2, S1, and S2 were placed in ice cream cartons, doubly bagged in polyethylene, and analyzed nondestructively by calorimetry

and neutron counting. Following the NDA study, one of the samples (S2) was completely dissolved in 2M Al $(NO_3)_3$-2M HNO_3. Aliquots of the dissolved S2 sample were analyzed destructively by controlled potential coulometry, alpha counting-mass spectrometry, and isotope dilution mass spectrometry. Results were compared to those from calorimetry.

Plutonium Isotopic Measurements

The isotopic content of any plutonium sample must be known or measured in order to relate the sample power to total plutonium content. In the present study, both mass spectrometry and gamma ray spectrometry were used to measure plutonium isotopics.

Gamma Spectrometry

Plutonium isotopic determination by gamma spectrometry has been demonstrated to be of comparable accuracy and precision to mass spectrometry for samples of dilute solution of low burn-up plutonium.[1] With this technique, a ten-mL sample of solution is placed in a precision-made vial and measured with a small, high resolution germanium detector. Gamma ray lines in the 38-150 keV region from Pu-238, Pu-239, Pu-240, and Pu-241 are used for the analysis. Studies have indicated that small but statistically significant differences between gamma spectrometric and mass spectrometric measurement of plutonium isotopics may result in a bias in calorimetric determinations of total plutonium content of 0.15%. A bias of this magnitude is insignificant in the present study.

DATA AND MEASUREMENT RESULTS

NDA Determinations

Repetitive calorimetric analyses were performed on all samples at two calorimeter power levels. These results are shown in Table 2. Equilibrium power prediction techniques were also used on slag sample S1. A typical plot of these results is shown in Figure 1. Results of singles neutron well counting of all samples are shown in Table 3. Coincidence neutron counting results are shown in Table 4.

Destructive Analyses

Slag sample S2 was destructively analyzed for comparison to the NDA measurements. After total dissolution of S2, aliquots were given to five different technicians for both gross alpha and

TABLE 2

Calorimetric Assay Results for JB-Line Slag and Crucible

Sample	Reference Power, watts	Plutonium Contents, grams	Number of Measurements
S1	7.4	51.8	1
	1.0	55.2 ±1.5	4
S2	7.4	62.2	1
	1.0	63.4 ±1.6	2
C1	7.4	17.2	1
	1.0	19.0 ±0.3	4
C2	7.4	18.9	1
	1.0	20.8 ±0.9	3

TABLE 3

Singles Neutron Emission Rates on Slag and Crucible Samples

Sample	Relative Neutron Counts/Minute · Gram/Pu*
S1	6.4
S2	3.7
C1	7.0
C2	7.6
	6.2 ±1.7
	±28%

* Based on plutonium content measured by calorimetry (Table 2).

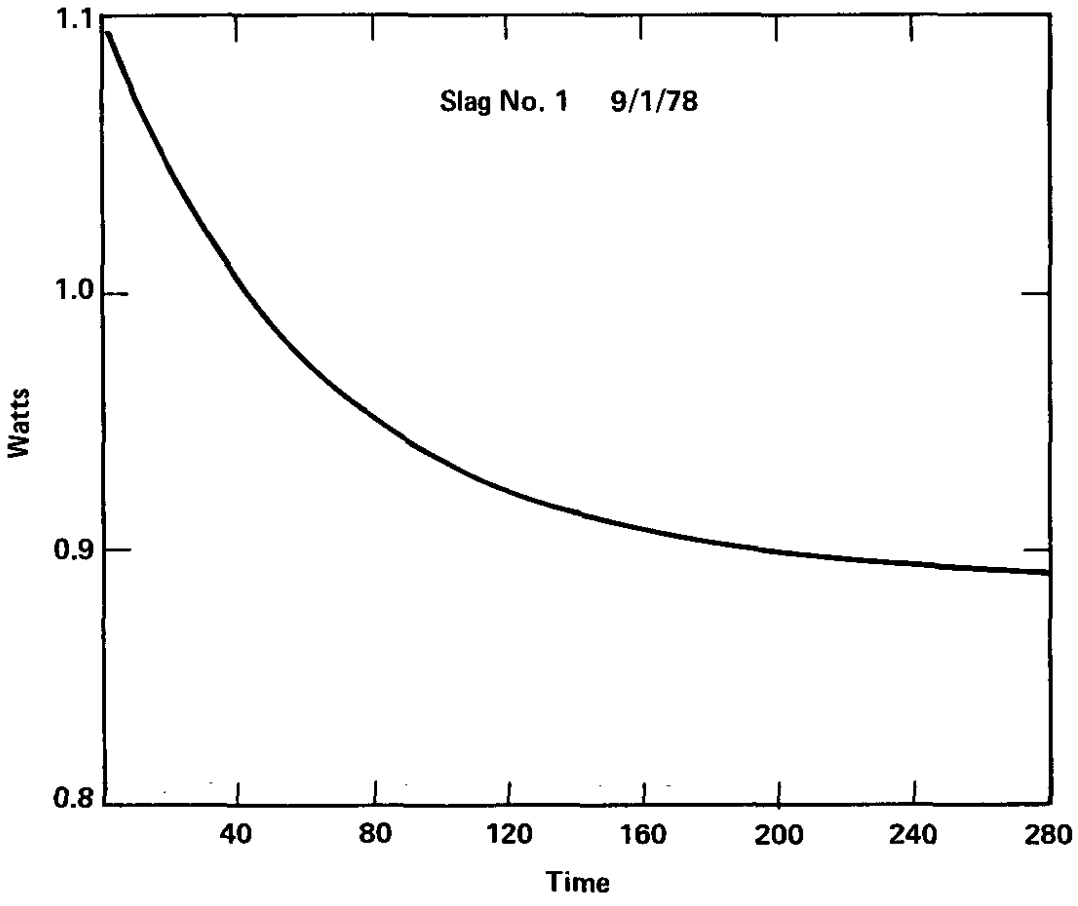

FIGURE 1. Power Level Versus Time

TABLE 4

Coincidence Neutron Counting Results on Slag and Crucible Samples

Sample	Relative Neutron Counts/Minute · Gram Pu*
S1	1.89
C1	2.04
C2	2.12
	2.02 ±0.12
	±5.8%

* Based on plutonium content measured by calorimetry (Table 2).

plutonium alpha by a thenoyltrifluoroacetone (TTA) extraction method. Gross alpha measurement data are given in Table 5. In addition, aliquots were submitted for isotope dilution mass spectrometry and for duplicate determinations by coulometry. The reduced data for the three techniques are given in Table 6. Destructive analysis results agree well with those measured by calorimetry.

Plutonium Isotopic Measurements

Results of the plutonium isotopic measurements performed on the dissolved button sample by gamma spectrometry and on the dissolved slag sample S2 are presented in Table 7. The normalized average plutonium isotopic content was used to calculate the specific power of the plutonium. A value of 0.23109 watt/gram (freshly separated from Am-241) was used to convert measured sample powers to grams of plutonium. All analytical results for calorimetry measurements were decay corrected to a common date by using the methods and half-lives recommended in ANSI N15.22-1975 and listed in Table 8.[2] The isotopic data were also used to calculate the specific alpha activity of the plutonium used in this study. The calculated value of 1.631×10^8 alpha dis/(min mg) plutonium was used to convert the measured alpha count rate of the dissolved S2 sample into grams plutonium. For reference purposes, a survey of measured isotopic content of a representative number of JB-Line plutonium product buttons was conducted (Table 9). The calculated range in specific power of JB-Line product is 0.2288 to 0.2318 watt/gram.

TABLE 5

Plutonium Determinations by Alpha Counting Techniques

Analyst	Method	10^9 dis/(min mL) Results	Average	σ
A	gross α	1.88 1.83	1.855	0.035
B	gross α	1.93 1.92	1.925	0.007
C	gross α	1.92 1.90	1.910	0.014
D	gross α	1.99 2.00	1.995	0.007
E	gross α	1.92 1.92 1.96 1.95	1.947	0.002
C	TTA/gross α	1.87 1.89	1.880	0.014
D	TTA/gross α	1.97 1.89	1.93	0.056
E	TTA/gross α	1.88 1.85	1.865	0.021
Overall Average		1.915 ±0.047 (2.4%)		

TABLE 6

Destructive Assay Results for S2 Sample

Method	No. Determinations	Plutonium Concentrations* grams/liter	Total Plutonium grams
Alpha Counting	18	11.9 ±0.3	62.4 ±1.6
Isotope Dilution Mass Spectrometry	2	11.43 ±0.15	60.0 ±0.8
Coulometry	2	13.1 ±1.0	68.7 ±2.7
Weighted Mean			61.0 ±1.2**

* Uncertainty does not include volume measurement. The volume measurement was reproducible to ±0.5%.

** 95% confidence limit in standard deviation of weighted mean.

TABLE 7

Plutonium Isotopic Measurements

Plutonium Isotope	Button	Slag		Average	Normalized
	Gamma Spec	Mass Spec	Gamma Spec		
238	0.0134	-	0.0140	0.0137	0.0137
239	93.67	93.53	93.61	93.60	93.58
240	5.68	5.74	5.72	5.71	5.71
241	0.64	0.68	0.66	0.66	0.66
242	-	0.04	-	0.04	0.04

TABLE 8

Nuclear Decay Parameters Used in This Report

Radio-nuclide	Half-Life, years	Standard Deviation, years	Specific Power, mW/g	Standard Deviation, mW/g	Decay Constant, day^{-1}
Pu-238	87.79	0.08(0.09%)	567.16	0.57(0.10%)	2.1617×10^{-5}
Pu-239	24082	46(0.19%)	1.9293	0.0053(0.27%)	7.880×10^{-8}
Pu-240	6537	10(0.2%)	7.098	0.015(0.2%)	2.903×10^{-7}
Pu-241	14.35	0.02(0.14%)	3.390	0.002(0.06%)	1.322×10^{-4}
Pu-242	379000	*	0.1146	*	5.01×10^{-9}
Am-241	434.1	0.6(0.14%)	114.23	0.16(0.14%)	4.372×10^{-6}

* None given — not significant to the analysis.

TABLE 9

Pu Isotopic Data for JB-Line Product Buttons

Year	Average Isotopic Content, wt % 238	239	240	241	242	Specific Power, mW/g
1973	0.0095(7)	93.34(10)	5.90(7)	0.72(5)	0.04(0)	2.301(5)
1974	0.01118(12)	93.27(11)	5.98(8)	0.70(6)	0.04(0)	2.318(8)
1975	0.0107(17)	93.45(15)	5.80(13)	0.70(3)	0.03(1)	2.300(14)
1976	0.0118(27)	93.20(13)	6.01(10)	0.70(4)	0.08(5)	2.319(18)
1977	0.0087(13)	93.51(8)	5.73(7)	0.72(4)	0.03(0)	2.288(7)
1978	0.0099(16)	93.53(14)	5.76(11)	0.66(4)	0.04(1)	2.296(12)

Note: values in parenthesis are the uncertainties in last digit(s) shown.

CONCLUSIONS

Calorimetric Assay

Comparison of the calorimetry assay results on slag sample S2 (63.4 ±1.6 grams plutonium) with the weighted mean results from destructive analysis techniques (61.0 ±1.2 grams plutonium) indicates that calorimetry can serve as a suitable nondestructive method for assay of inhomogeneous solids.

The total book value estimate of plutonium content of the four S&C samples was 111 grams compared with the measured value of 158 grams. This confirms the bias noted in Table 1 from data taken after dissolution of JB-Line S&C samples.

Because the specific power of the nominal 6% Pu-240 produced at SRP (see Table 9) only varies by about 1%, calorimetric assay of freshly generated S&C at acceptable precision and accuracy levels can be made without measurement of the isotopic distribution.

The calorimeter used for this study was designed for sample power level of approximately two kilograms of Pu-240 metal buttons. A systematic difference of about 6% was observed in the average measured plutonium content of each sample between measurements performed at a higher (7.4-watt) reference power level and the lower

(1.0-watt) level. This bias results from poor measurement statistics at the higher reference power level which mathematically requires measuring small differences between large power levels. Improved precisions were observed for most measurements taken at the lower power level. However, an unpredictable reference power level drift of about ±2% at the lower power level accounted for much of the difference in plutonium content observed in repetitive measurements shown in Table 2.

This drift results from using a servo-type calorimeter designed for materials with short equilibrium time constants (good heat transfer characteristics) such as metal buttons. The oxides and fluorides which are the major constituents of the S&C samples are examples of materials with poor heat transfer characteristics; therefore, equilibrium time constants are very long — 16 to 24 hours. Packaging of the material was also less than optimum for good heat transfer. Materials with poor heat transfer characteristics can best be assayed with nonservo calorimeters of the standard twin bridge design.

Equilibrium Power Level Prediction

The Mound computer program predicted an equilibrium sample power to within ±0.02% of that measured (corresponds to ±0.1 grams plutonium) after 240 minutes. The Rocky Flats computer program predicted equilibrium after 300 minutes to within ±0.31% (±1.2 grams plutonium) of that measured. Efforts are underway at both sites to improve the predictive codes. These preliminary results indicate that calorimetry can provide nondestructive assay of S&C to acceptable levels of accuracy in <6 hours.

Singles Neutron Counting Results

Results of singles neutron well counting of S&C are not consistent with the calorimetric analyses. This is due to the highly variable contribution of alpha-induced neutrons to the overall neutron emission rate. This variability results from nonuniform distributions of plutonium and light elements within the S&C samples. These data indicate that singles neutron counting cannot provide reliable assay of plutonium in inhomogeneous samples. Earlier work by Gibbs[1] does indicate, however, that singles neutron counting can be applied successfully to the assay of total plutonium in metal buttons of standard size and matrix.

Coincidence Neutron Counting

Coincidence neutron counting reduces the effect of variable single neutron emission events from (α,n) nuclear reactions on

light element impurities within the matrix. This permits multiple neutron events to be recorded from the spontaneous fission of Pu-240. Data in Table 4 indicate that significantly better precision was achieved (±5.8%) when relative coincidence neutron count rates are compared to singles count rates. Still, these results are not good enough for accountability measurements. Efforts are underway at Los Alamos National Laboratory (LANL) to improve precision by developing a means to correct for the effect of induced fission and variable sample matrix.

Survey — Onsite and Offsite Plutonium-Bearing Solids

Table 10 contains material composition and packaging data pertinent to the nondestructive assay of onsite materials. Table 10 is an incomplete list of data on some categories of oxide and metal scrap which has been received from offsite in the past. The list is considered typical of materials presently stored or anticipated to be received in the future. Each category listed is usually composed of several subgroupings of materials. Container dimensions and SNM content are considered the key variables in the design and specification of NDA instruments to measure plutonium content.

Table 11 lists data pertinent to the nondestructive assay of plutonium-bearing solids generated in the JB-Line process.

TABLE 10

Offsite Plutonium-Bearing Scrap

Material Category	Composition	SNM Weight	Container O.D, inches	Dimension Height, inches	Plutonium Isotopics 238	239	240	241	242	Specific Power, mW/g Pu	Power per Container, watts
1	Metal	2100	3.0	3.5	0.05	84	14	2	-	3.0	∿6.4
2	Oxide	600-1000	4.25	4.625	0.2	76	20	3	-	5.7	3.4-5.7
3A	Oxide	800	4.0	9.625	0.01	93.5	6.3	0.6	-	2.3	2
3B	Metal	1400	10.75	10.25	NA	NA	NA	NA	-	NA	NA
4	Oxide	370-470	4.25	4.88	0.03	94	5.6	0.25	-	2.3	0.85-1.1
5	Metal	270-2050	4.0	2.75	0.05	89	10	1	-	2.2	0.5-4.5
6	Oxide	830-1930	4.0	9.625	NA	93	6	-		2.3	2-4
7	Oxide	1-1000	4.25	7.0		88-93	5-10	0.4-1.3		2.3-3	≤3
8	Oxide	12-200	4.25	4.88			∿12			∿3	30-600

TABLE 11

Onsite Plutonium Bearing Solids

*Material**	Composition	SNM Weight, g	Outer Container Dimensions O.D.	Outer Container Dimensions Height, in.	Fill Height	Container Wall Thickness, mils	Comments
Metal Product Buttons	Pu Metal	2000-2500	4.25	4.875		14	One button per container in sealed, inner tinned steel can of dimensions 3.75 in. x 2.50-in. height of 10-mil wall thickness.
Oxide Product	PuO_2 Powder	2000-2400	4.25	7.735	NA	16	PuO_2 product powder sealed in inner, tinned steel can of dimensions 3.75 O.D. x 6-in. height and wall thickness, 16 mils.
Slag and Crucible (S&C)	Pu and PuO_2 in MgO, CaO, Ca, CaF	30-300	5.25	10.125	1/2-7-7/8	19	S&C contained in bagged** stainless steel outer container. Two S&C containers stored in vault in 5-gallon bucket.
Solid Waste	Gloves, Tools, Filters, etc.	0-500	12	13.625	0 to Full	26 gauge	Used to store S&C in vault. Also used to contain waste prior to NaI scan for disposal.
Excess Metal Turnings	Metal	0-150	4.25	4.875	0 to Full	14	Same tinned steel can as product metal. Turnings in inner plastic vials of dimensions 1.0 O.D. x 3.0-in. height.
Process Sweepings	PuF_4/PuO_2	0-300	5.25	10.125	0 to Full	19	Two containers packed per 5-gallon pail**

* Isotopics — all weapons grade at 2.3 mW/g specific power.

** Five-gallon bucket only form amenable to rapid NDA because of contamination.

REFERENCES

1. A. Gibbs. **Evaluation of a Gamma/X-ray Spectrometer System for the Nondestructive Analysis of Plutonium and Uranium.** USAEC Report DPSPU-74-11-20, E. I. du Pont de Nemours & Co., Savannah River Plant, Aiken, SC (1974).

2. **American National Standard Calibration Techniques for the Calorimetric Assay of Plutonium-Bearing Solids Applied to Nuclear Materials Control.** ANSI Report ANSI N15.22-1975, American National Standard Institute, Inc., New York, NY (1975).