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Preparation of Pure Plutonium Metal Standards for Nondestructive Assay

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ABSTRACT

To calibrate neutron coincidence and neutron multiplicity counters for passive assay of plutonium, certain detector parameters must be determined. When one is using small plutonium metal samples, biases can be introduced from non-zero multiplication and impurities. This paper describes preparing small, pure plutonium metal standards with well-known geometries to enable accurate multiplication corrections and with acceptably low levels of impurities. To minimize multiplication, these standards are designed as 2-cm-diameter foils with varying thicknesses and masses of 1.4, 3.6, and 7.2 g plutonium. These standards will significantly improve characterization and calibration of neutron coincidence and multiplicity counters. They can also be equally useful for gamma-ray spectrometry and calorimetry. Five sets will be made: four for other United States Department of Energy plutonium facilities, and one set to remain at Los Alamos. We will also describe other nondestructive assay standards that are planned for the next few years.

I. INTRODUCTION

Neutron time-correlation counting has been used extensively during the past 20 years for the nondestructive assay (NDA) of nuclear material. The usefulness of the technique is due primarily to the good penetrability of neutrons and the uniqueness of time-correlated neutrons to nuclear material content. Passive neutron coincidence and multiplicity counting are useful in the independent verification or NDA of plutonium mass, in particular the ^{240}Pu effective mass, of an item.

To characterize neutron coincidence and multiplicity counters, certain detector parameters such as efficiency, doubles gate fraction, and triples gate fraction, must be determined. To determine these detector parameters, the ideal plutonium standards would be nonmultiplying and have no significant sources of neutrons other than from spontaneous fission of the ^{240}Pu effective mass. In practice, ^{252}Cf sources are used and empirical corrections are necessary to obtain parameters for ^{240}Pu. Small plutonium metal and oxide items are also used, but biases are introduced because of unknown multiplication and impurity concentrations.

This paper describes the specifying and preparing several sets of small, pure plutonium metal standards with well-known geometries and acceptable low levels of impurities. These standards are designed specifically for the calibration and characterization of neutron coincidence and multiplicity counters. With these standards, neutron coincidence and multiplicity counters can be characterized much better than

before. Calibration of these counters will be improved, and bias of assays with these counters will be reduced. The need for this set of standards was identified in a previous review report on NDA standards.[1]

II. STANDARDS SPECIFICATIONS

A. Impurity Limits

Impurities in plutonium can contribute to neutron emission in two ways: (1) very small concentrations of ^{244}Cm and ^{252}Cf can yield spontaneous fission neutrons in addition to ^{240}Pu effective, and (2) low-Z elements can contribute to total neutron yield of plutonium metal through (α,n) reactions.

Source neutrons from pure plutonium metal arise solely from spontaneous fission, primarily from the even isotopes of ^{238}Pu, ^{240}Pu, and ^{242}Pu. Very small concentrations of the impurities of ^{244}Cm and ^{252}Cf can also yield spontaneous fission neutrons that bias assays of the even plutonium isotopes. Limits on the allowable concentrations of ^{244}Cm and ^{252}Cf were calculated from basic nuclear yield data, based on the assumption that the impurity isotope contributes 1% of the spontaneous fission neutron specific yield for pure weapons-grade plutonium. These limits are given in Table I. The mass limits are listed as parts μg/g of plutonium.

Table I. Impurity Limits for Spontaneously Fissioning Isotopes

Isotope	μg/g mass Limit
^{244}Cm	5.63E-02
^{252}Cf	2.60E-7

Low-Z elements can also contribute to total neutron yield of plutonium metal through (α,n) reactions. Limits on the allowable concentrations of 11 impurity elements were calculated by using basic nuclear and atomic data with a simple stopping power model.[2] Again, it was assumed that the impurity element contributes 1% of the spontaneous fission neutron specific yield for pure weapons-grade plutonium. These limits are given in Table II.

Table II. Impurity Limits for Elements Producing (α,n) Neutrons	
Element	μg/g mass limit
Be	1
B	4
F	13
Li	57
Na	71
Mg	88
Al	200
Si	1100
Cl	1300
C	800
O	1200

B. Multiplication

For an ideal neutron standard, no neutron multiplication is preferred but there is always some multiplication, even in the smallest items. One then must know the sample multiplication well. Neutron multiplication is a complicated function of the sample composition (including impurities), density, and geometry. The best standard, therefore, is one with as small a multiplication as possible so as to have the minimum effect on the neutron emission rate. The least multiplying geometry is a thin disc. Multiplication effects of thin metal discs can be estimated well using Monte Carlo simulations. An alternative procedure for using these standards is given in section VII. This procedure is based on extrapolation to zero mass of ^{240}Pu effective.

The approximate diameter and thicknesses of the standards are listed in Table III.

Table III. Radii and Thicknesses of the Plutonium Metal Standards		
Mass (g)	Radius (cm)	Thickness (cm)
1.4	1.0	0.0321
3.6	1.0	0.0802
7.2	1.0	0.1604

III. STANDARDS FABRICATION

A. Purification

To achieve the impurity levels described previously, the plutonium metal must be purified. Electrorefining is a powerful method of purifying crude plutonium metal.[3] A typical electrorefining cell used in plutonium processing operations is shown in Fig. 1. The crude batch of plutonium alloy is placed in the anode compartment. The cell system is heated, and an electric current is passed between the anode and cathode to produce electrolysis. At the conclusion of the process, the purified plutonium is recovered as an annular metal ring.

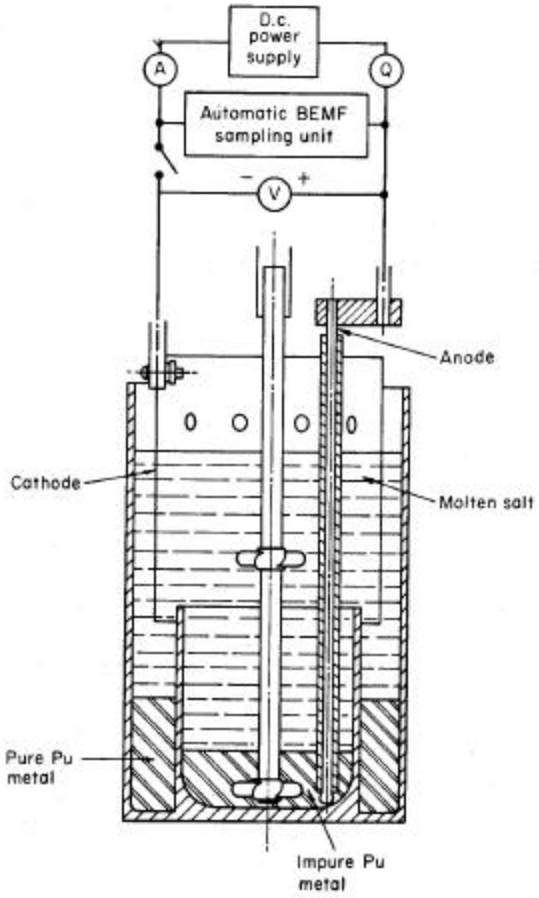

Fig. 1. Schematic of electrorefining cell showing major features.

Plutonium, as a metal of the actinide group, has six allotropes. Of the six allotropic phases, alpha is the hardest and least ductile, and delta is the softest and most ductile. The metal resulting from electrorefining is alpha-phase plutonium. In order to fabricate the plutonium metal into thin discs as required for this project, the plutonium metal needs to be in the softest, or delta phase, to minimize the risk of breakage during fabrication. Alpha-phase plutonium metal is converted into delta phase by means of vacuum distillation and addition of gallium; the distillation process further reduces the impurities in the plutonium metal. The distillation is repeated three times to maximize the purification. The plutonium is then cast into an ingot by using a furnace as shown in Fig. 2.

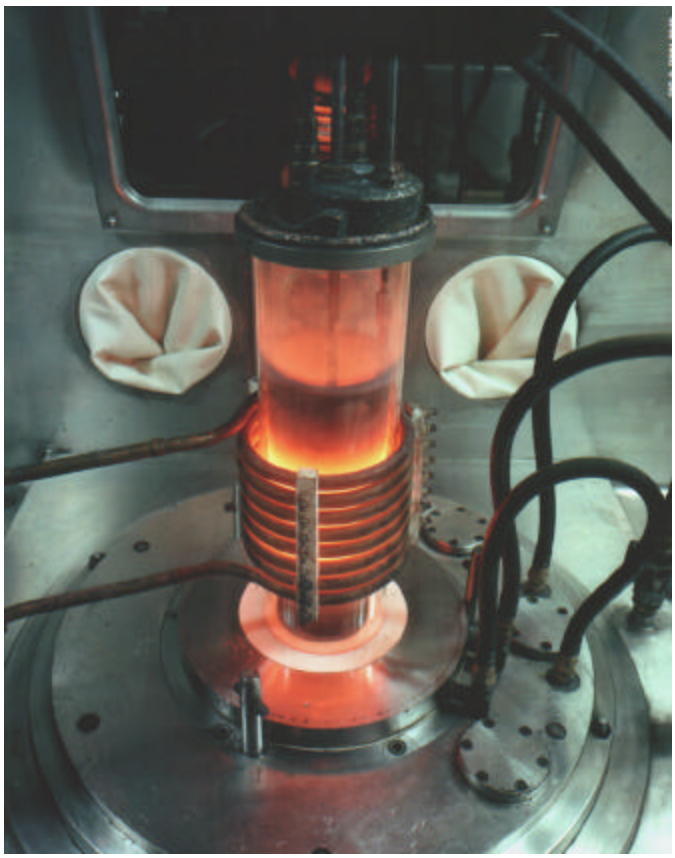

Fig. 2. Furnace to cast plutonium metal into ingot.

B. Fabrication of Foils

The purified plutonium metal ingot is sliced into three pieces; each is rolled into a disc of approximate thickness (shown in Table III). Figure 3 shows the feeding of the disc to the rolling machine and Fig. 4 shows catching the disc during the rolling operation. This process is repeated until the desired thickness is achieved; the disc thickness is checked with a micrometer, as shown in Fig. 5.

Figs. 3 and 4. Feeding and catching the disc during the rolling operation.

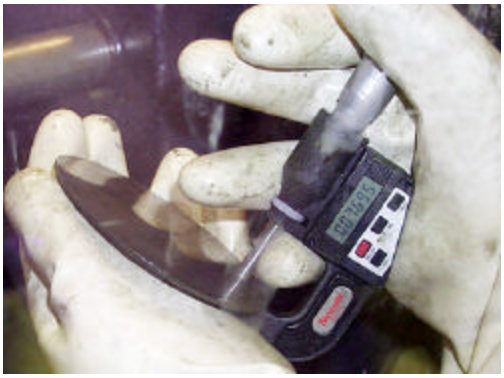

Fig. 5. Monitor the thickness of the disc during the rolling to decide whether further rolling is necessary.

A stamping machine is used to punch out 2-cm-diameter plutonium foils; the punching operation is shown in Fig. 6. Five foils of each thickness are punched. The plutonium foils are stored in glass tubes (Fig. 7) to minimize oxidation.

Fig. 6. Plutonium foil being punched out of the disc with the stamping machine.

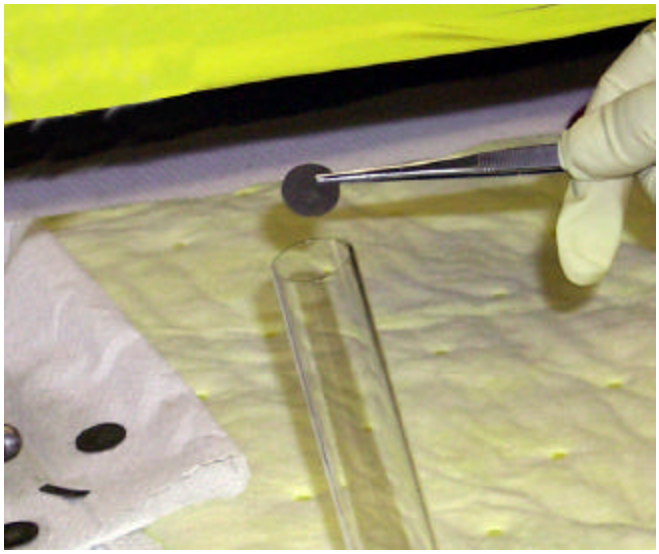

Fig. 7. Plutonium foils are stored in glass vial to reduce oxidation and contamination. The glass vial is transferred to an inert glove box for encapsulation.

IV. SAMPLING AND CHEMICAL ANALYSES

From each foil thickness, two samples are taken from different parts of the foil (after the discs are punched) for destructive chemical analysis. A total of six samples will be analyzed to allow for determining uniformity. Each of the samples will be electropolished and sealed in a glass tube. Archive samples will also be taken.

The chemical analysis for each sample includes:

1. g Pu/g sample

2. Mass spectrometry to determine plutonium isotopic distribution
   - ^{241}Am determination by gamma counting or alpha spectroscopy
   - ^{238}Pu determination by alpha counting

3. Impurity analyses
   - The impurity analyses include all the elements listed in Table II: Be, B, F, Li, Na, Mg, Al, Si, Cl, C, and O.

4. Spontaneous fissioning isotopes listed in Table I.
   - ^{244}Cm and ^{252}Cf

A. Isotopic Distribution

The chemical analysis results are summarized in Table IV. Table IV shows the plutonium isotopic of the six samples submitted for analysis. All the plutonium isotopic distribution and Am^{241} have been decay corrected to January 1, 2000. The ^{238}Pu was determined by alpha counting, and Am^{241} was determined by gamma counting. The Am^{241} was reported as ppm of sample; this number was divided by the g Pu/g sample (on 1/1/2000) to obtain Am^{241} relative to plutonium mass, which is reported in this table.

Table IV. Plutonium isotopic distribution as of 1/1/2000

Sample ID 1/1/00	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am
WH0321-C1	0.0119	93.9410	5.8519	0.1511	0.0441	0.0548
WH0321-C2	0.0122	93.9405	5.8526	0.1508	0.0439	0.0556
WH1604-C1	0.0117	93.9423	5.8519	0.1501	0.0440	0.0556
WH1604-C2	0.0117	93.9407	5.8526	0.1509	0.0441	0.0547
WH0802-C1	0.0123	93.9410	5.8519	0.1508	0.0440	0.0549
WH0802-C2	0.0120	93.9404	5.8529	0.1506	0.0441	0.0548
Average	0.0120	93.9410	5.8523	0.1507	0.0440	0.0551

B. Impurity Determination

^{244}Cm and ^{252}Cf impurity levels were determined by means of alpha spectroscopy, and in the case of ^{252}Cf, extremely long counts (three days). These impurity levels are given in Table V.

Table V. Impurity Limits for Spontaneously Fissioning Isotopes

Isotope	μg/g upper Limit (1 %)	μg/g measured Limit
^{244}Cm	5.63E-02	<0.04
^{252}Cf	2.60E-7	<8.4E-5

Impurity limits for the elements listed in Table II are given in Table VI:

Table VI. Impurity Limits for Elements Producing (α,n) Neutrons

Element	μg/g upper limit (1%)	μg/g measured limit	Method
Be	1	0.9	ICP*
B	4	<4	ICP
F	13	9.0	Ion Chromatography
Li	57	<0.9	ICP
Na	71	<40	ICP
Mg	88	<4	ICP
Al	200	<9	ICP
Si	1100	<40	ICP
Cl	1300	10.3	Ion Chromatography
C	800	61.7	MS**
O	1200	48.3	Combustion

*Inductive coupled plasma spectroscopy
**Mass spectroscopy-carbon

C. Plutonium concentration

The plutonium concentration of the samples submitted for analysis was determined by coulometry. The results are shown in Table VII.

Table VII. Plutonium concentration

Sample ID 1/1/00	% Pu
WH0321-C1	99.261
WH0321-C2	99.311
WH1604-C1	99.291
WH1604-C2	99.411
WH0802-C1	99.351
WH0802-C2	99.301
Average	99.321
St Deviation	0.053

D. Traceability

These analysis methods are based on the New Brunswick Laboratory certified standards with the exception of the impurity analysis. Assays of certified standards are part of the routine procedure.

V. CONTAINERS AND ENCAPSULATION

The cans for the standards are stainless steel and will be American National Standards Institute (ANSI) certified. A schematic drawing of the cans is shown in Fig. 8. A picture of the inner and outer cans is shown in Fig. 9. Welding is performed by an ANSI-certified welder in a helium-atmosphere glove box.

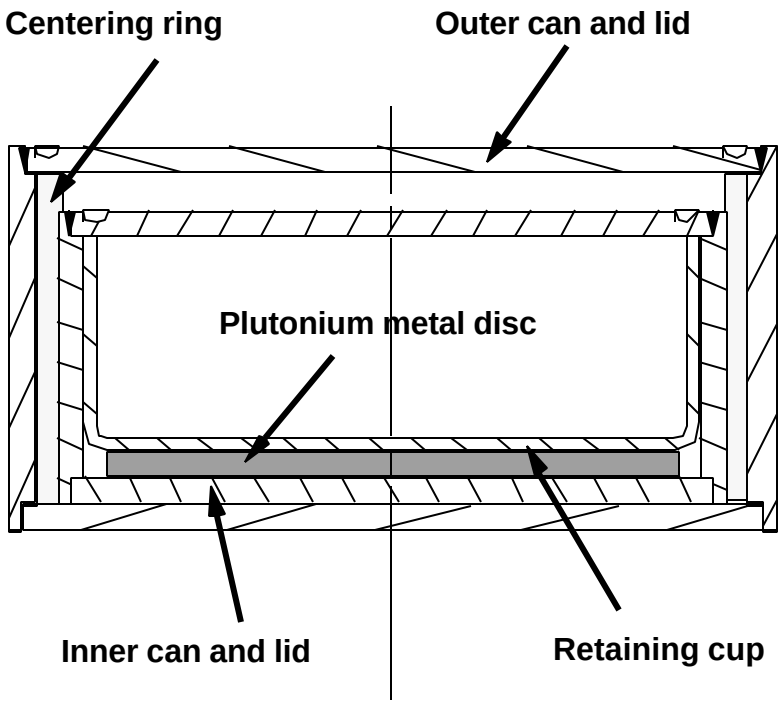

Fig. 8. Schematic drawing of the cans. All the parts, except for the plutonium disc, are made of stainless steel.

Fig. 9. Picture of stainless steel cans and parts. The right-hand side of the picture shows the inner can, lid, and retaining cup. The left-hand side shows the outer can, lid, and the centering ring.

VI. MASS CALCULATION

The plutonium foils were weighed by using scales accurate to 0.0001 g. The scales were checked with National Institute of Science and Technology certified weights within the range of 1 to 10 g. These weights are listed in the second column of Table VI. The plutonium mass of the standards is obtained by multiplying the foil mass by the average plutonium concentration listed in Table V.

Table VIII. Calculation Plutonium Mass of Foil

STD ID	Plutonium Foil Mass (g)	Plutonium Mass (g)	Relative error (%)	Absolute error (g)
A1	1.7775	1.765	0.077	0.0014
A2	4.0702	4.043	0.058	0.0024
A3	7.7402	7.688	0.055	0.0042
B1	1.833	1.821	0.076	0.0014
B2	3.9538	3.927	0.059	0.0023
B3	8.0085	7.954	0.054	0.0044
C1	1.8351	1.823	0.076	0.0014
C2	4.057	4.029	0.058	0.0024
C3	7.9672	7.913	0.054	0.0043
D1	1.6997	1.688	0.079	0.0013
D2	3.7651	3.740	0.059	0.0022
D3	7.4829	7.432	0.055	0.0041
E1	1.7529	1.741	0.078	0.0014
E2	3.4216	3.398	0.061	0.0021
E3	7.811	7.758	0.055	0.0043

VII. PROCEDURE FOR USE OF STANDARDS

The primary purpose of the plutonium metal standards is for better determination of certain neutron detector parameters used in neutron coincidence and neutron multiplicity assays of plutonium-bearing materials.

In order to define procedures for uses of the standards, some of the basic concepts of neutron coincidence and multiplicity counting must be defined. For an introduction to neutron coincidence counting, see Ref. 4 and the references therein. For an introduction to neutron multiplicity counting, see the application guide (Ref. 5) and the references therein.

There are two measured multiplicity distributions of neutron pulses (0s, 1s, 2s, 3s, 4s, 5s, etc.) measured: the multiple counts in the "real-plus-accidental" (R+A) gate and the "accidental" (A) gate. For a purely random (in time) pulse stream, the two distributions are the same within statistical errors. For a time-correlated pulse stream with fission neutron pulses, the R+A distribution has more higher order multiplicity events than the A distribution. The two distributions are analyzed by the INCC[6] code to obtain the number of time-correlated double, triple, and quadruple pulses, etc. The term "doubles" (D) means the number of correlated pulse pairs in the pulse stream; "triples" (T) means the number of correlated triplets; "singles" (S) means the total number of neutron counts. In practice, triple events are usually the highest correlations that can be obtained with reasonable statistical precision. A standard shift register circuit determines the number of singles and doubles but can't determine the number of triples. A multiplicity shift register determines S, D, and T.

Neutron source multiplicity distributions are the probability distributions for the emission of source distributions per event; for plutonium metal, an event can be, for example, a spontaneous fission followed by an induced fission. Passive neutron multiplicity assay (PNMA) analysis uses factorial moments of the spontaneous and induced distributions.

For a plutonium metal sample with a very small mass, the background-corrected rates S, D, and T are given by

$$S = Fen_{s1} \quad , \tag{1}$$

$$D = \frac{1}{2} Fe^2 f_d n_{s2} \quad , \text{ and} \tag{2}$$

$$T = \frac{1}{6} Fe^3 f_t n_{s3} \quad , \tag{3}$$

where

$F =$ the rate of source events,

$e =$ detector efficiency,

f_d, $f_t =$ double and triple gate fractions, and

$n_{s1}, n_{s1}, n_{s3} =$ 1st, 2nd, and 3rd factorial moments of the neutron source distributions.

The gate fractions account for the fact that the R+A gate is not open long enough to count all the correlated neutrons.

Ideally, an assay based on the doubles counts should provide a unique signature for plutonium and should also determine the actual grams of ^{240}Pu-effective in the sample, where this quantity is defined as that mass

of ^{240}Pu which would give the same D response as that obtained from all the even isotopes of ^{240}Pu-effective in the sample:

$$^{240}Pu_{eff} = 2.52^{238}Pu + ^{240}Pu + 1.68^{242}Pu \quad . \tag{4}$$

The total plutonium mass is then obtained from the plutonium isotopic composition:

$$^{Total}Pu = ^{240}Pu_{eff} / (2.52c_{238} + c_{240} + 1.68c_{242}) \quad , \tag{5}$$

where c_{238}, c_{240}, and c_{242} are the fractions of the plutonium isotopes present in the sample.

The parameters that are obtained by using the small plutonium metal standards are, in order, e, f_d, and f_t, using equations (1) – (4),

where

$$F = ^{240}Pu_{eff}\ 473.5 \quad , \tag{6}$$

$$e = S/(F/n_{s1}) \quad , \tag{7}$$

$$f_d = 2n_{s1}D/(en_{s2}S) \quad \text{, and} \tag{8}$$

$$f_t = 3f_d n_{s2}T/(en_{s3}D) \quad . \tag{9}$$

The parameters n_{s1}, n_{s2}, and n_{s3}, respectively, are 2.154, 3.789, and 5.211.

The parameters in Eqs. (7) – (9) are obtained for all three standards in a given set. The results are extrapolated back to zero mass to obtain the final values.

The quantities e, f_d, and f_t should be determined with a precision from counting statistics of not more than 0.5%. This will require a minimum of 3-hour counts per standard in a detector with e = 50%.

ACKNOWLEDGMENT

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