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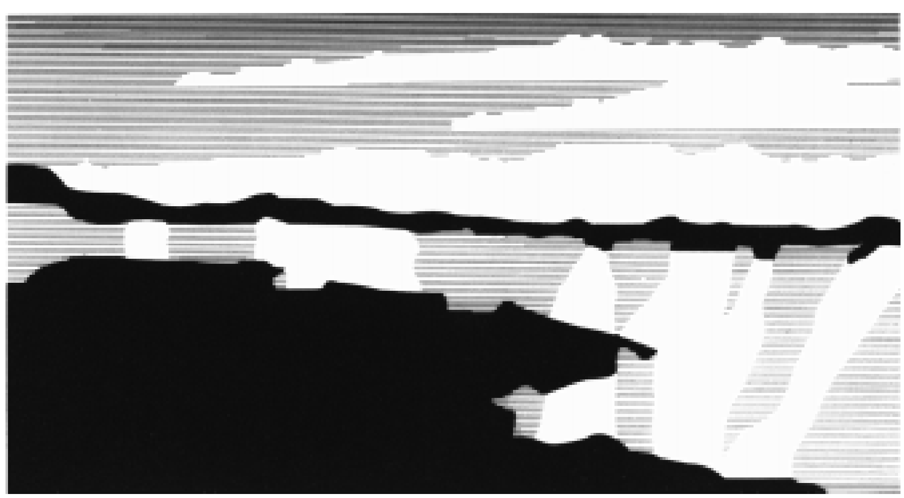

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PERFORMANCE OF THE PLUTONIUM SCRAP MULTIPLICITY COUNTER (PSMC) FOR VERIFICATION OF EXCESS SCRAP PLUTONIUM OXIDE FROM NUCLEAR WEAPONS PRODUCTION

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ABSTRACT

Neutron multiplicity counting has become the method of choice for International Atomic Energy Agency (IAEA) verification of impure plutonium residues at the Hanford reservation and the Rocky Flats Environmental Technology Site. This report describes the performance of the plutonium scrap multiplicity counter (PSMC) at the Hanford Plutonium Finishing Plant (PFP) for measurements of reference materials, sampled and analyzed chemically by the IAEA. The PSMC performance provides confidence in application for partial defects measurements of pure and impure plutonium items in the Hanford PFP inventory.

INTRODUCTION

The US Nonproliferation and Export Control Policy, announced by President Clinton before the United Nations General Assembly on September 27, 1993, commits the US to place under International Atomic Energy Agency (IAEA) Safeguards excess nuclear materials no longer needed for the US nuclear deterrent.

As of June 1, 1997, the IAEA had completed the initial physical inventory verification (PIV) at three US sites: a storage vault at the Oak Ridge National Laboratory Y-12 plant containing highly enriched uranium (HEU) metal, a second storage vault at the Hanford Plutonium Finishing Plant (PFP), and a third storage vault at Rocky Flats Environmental Technology Site (RFETS). The Hanford PFP and RFETS vaults house containers of plutonium oxide and plutonium residue powders.

Conventional neutron coincidence counting (NCC) is one of the routinely applied nondestructive assay (NDA) methods for IAEA verification of pure uranium and plutonium product materials. The excellent results from implementing the active-well coincidence counter

(AWCC) to verify the HEU material at Y-12 is described in reference 1. For most of the plutonium materials at Hanford PFP and all plutonium materials at RFETS, conventional NCC could not be used because of impurities that give rise to unknown intensities of neutrons from (α,,n) reactions. In both of these cases, neutron multiplicity counting (NMC) was successfully applied by the IAEA to verify impure materials. Reference 1 includes a comparison of three NCC and one NMC assay methods for 21 of the Hanford items. Each item was packaged in three nested metal cans, with the outer can typically 4 in. in diameter and 6 in. tall. Reference 1 shows that, compared with the NCC methods, the NMC assay method provides the best results for impure plutonium residues, but requires longer counting times. These data were taken with the 3-ring multiplicity counter (3RMC). Reference 2 describes the results from using the 3RMC for NMC assay of 69 items drawn from the Hanford inventory. Sixty-seven of these items passed acceptance criteria and two items were sampled for assay by destructive analysis (DA). For the 67 items, the average percent difference between the declared and NMC assay values was +0.02 ± 0.83%. The present report describes the NMC assay performance of the plutonium scrap multiplicity counter (PSMC)[3] as applied to IAEA standards taken from the population of Hanford materials. The PSMC was designed originally as a multiplicity counter, and therefore has a higher efficiency (55% vs 45%) than the 3RMC. The higher efficiency results in shorter counting times for the same assay precision. The 12 IAEA standards were weighed and sampled for DA. These weights and DA analyses are compared directly with the PSMC NMC assay values in this report. A separate report describes the results from applying a much larger instrument for the Rocky Flats Environmental Technology Site material containers.

THE MULTIPLICITY ASSAY SYSTEM

The PSMC[3] evolved from multiplicity neutron detectors[4,5] developed at Los Alamos National Laboratory for assay of impure plutonium items. Compared with earlier designs, the PSMC was designed to be smaller, lighter, and to use fewer ^{3}He proportional counters to obtain high neutron counting efficiency. The PSMC was designed using Monte Carlo simulations of neutron transport. The design goals for the PSMC were

1. high efficiency (primary importance),
2. uniform efficiency vs sample height,
3. small die-away time,
4. uniform efficiency vs neutron energy,
5. minimum number of ^{3}He counters, and
6. minimum overall size and weight.

The PSMC has 80 ^{3}He tubes embedded in a polyethylene moderator surrounding the sample cavity that is 20 cm in diameter and 40 cm tall. The measured efficiency of the counter is 55% and the die-away time is 47 µs. Other detector performance parameters and characteristics are given in reference 3 and are equal to or superior to those of the 3RMC. The PSMC used for the Hanford verification measurements was manufactured by Aquila. In addition to the detector head, the assay system consisted of an Aquila PSR-B multiplicity electronics package, an IBM-compatible desktop computer with monitor, and a Hewlett-Packard 4L printer. The PSR-B contains the multiplicity counting electronics and provides the +1680 V for the ^{3}He tubes along with the +5 V for the Amptek preamplifiers. The software used for data collection and analysis was the general-purpose NCC code, which is a Windows program written to cover a wide variety of passive and active NCC applications, including NMC assay.

MEASUREMENTS

The PSMC was calibrated at Los Alamos National Laboratory before the IAEA PIV at Hanford in August 1996. Detector parameters needed for NMC assay are the electronic dead time, the neutron detection efficiency, the doubles gate fraction, and the triples gate fraction. The first three of these parameters were obtained from measurements of reference ^{252}Cf sources, whose absolute neutron yields are known. The value for the triples gate fraction was obtained from measurements of plutonium items whose masses and isotopic compositions are known.

Before the PIV at the Hanford PFP in August 1996, 12 of the 17 IAEA standards were measured in the PSMC system. For each item, the NMC analysis determined the effective ^{240}Pu mass from the measured singles, doubles and triples count rates. The NMC analysis also uses the known parameters for the electronic dead time, neutron detection efficiency, doubles gate fraction, and triples gate fraction. Then the plutonium mass was calculated from the effective ^{240}Pu mass using the IAEA measured DA isotopic composition.

The measurement procedure was as follows. The items were placed in the PSMC sample cavity, radially centered on top of an empty can (about 10 cm tall) to position the plutonium in the uniform-efficiency counting region. Each sample was measured for 15–60 minutes, depending on the level of impurities. The NMC assay algorithm yields the parameter α, the ratio of the (α, n) to spontaneous fission neutron production in the sample. Higher values of α require longer measurement times for multiplicity assay because of reduced signal-to-background ratio.

The IAEA reference values, based on weighing and DA, for plutonium mass, effective ^{240}Pu mass, and plutonium/americium isotopic distribution are shown in Table I. The first seven items are pure plutonium oxide powder standards and the last five are impure plutonium residue standards.

Table I. IAEA reference values for powder and scrap standards

Item Id.	Std. Id.	Pu (g)	^{238}Pu (% Pu)	^{239}Pu (% Pu)	^{240}Pu (% Pu)	^{241}Pu (% Pu)	^{241}Pu (% Pu)	^{241}Am (ppm Pu)	$^{240}Pu_{eff}$ (g)
LAOC10260	STDPD1	264.8	0.051	82.407	16.43	0.776	0.336	6482	45.3
LAOC10240	STDPD2	470.6	0.052	82.459	13.371	0.771	0.347	6099	80.4
LAOC08255	STDPD3	863.6	0.055	82.462	16.377	0.762	0.345	6372	147.6
LAOC06249	STDPD4	865.4	0.053	82.467	16.385	0.751	0.343	6638	147.9
LAOC03244	STDPD5	859.3	0.051	82.792	16.103	0.727	0.326	6312	144.2
PBO45123811	STDPD6	1374.7	0.098	85.51	12.767	1.376	0.247	7462	184.6
PBO45113220	STDPD7	1593.1	0.101	84.792	13.481	1.357	0.268	7389	226
41-86-03-240	STDSC1	268.8	0.025	93.809	5.773	0.332	0.061	2498	16
64-85-10-1584	STDSC2	385	0.012	93.817	5.95	0.185	0.036	1233	23.3
64-85-10-1543	STDSC3	467.8	0.015	93.577	6.162	0.201	0.044	1886	29.3
BO46-04-001	STDSC5	1660.9	0.016	93.858	5.895	0.189	0.042	993	99.8
ARF10286230	STDSC10	917	0.009	94.007	5.837	0.122	0.025	818	54.1

RESULTS

Results of the PSMC measurements of the 12 IAEA standards described in Table I are given in Table II. The % differences between references and assay value are given in the colunm labeled (R-A)/R(%).

II. PSMC assay results for IAEA powder and scrap standards

Item Id.	Std. Id.	Ref. Pu (g)	Pu Assay (g)	Assay error (g)	(R-A)/R (%)	Assay error (%)	Assay error (sigmas)
LAOC10260	STDPD1	264.8	263.7	1.67	0.4	0.6	0.7
LAOC10240	STDPD2	470.6	469	2.47	0.3	0.5	0.6
LAOC08255	STDPD3	863.6	856.2	6.01	0.9	0.7	1.2
LAOC06249	STDPD4	865.4	861.4	3.98	0.5	0.5	1.0
LAOC03244	STDPD5	859.3	877.5	4.06	-2.1	0.5	-4.5
PBO45123811	STDPD6	1374.7	1371	14.5	0.3	1.1	0.3
PBO45113220	STDPD7	1593.1	1624.6	14.5	-2.0	0.9	-2.2
41-86-03-240	STDSC1	268.8	283.6	21.4	-5.5	8.0	-0.7
64-85-10-1584	STDSC2	385	381.1	8.6	1.0	2.2	0.5
64-85-10-1543	STDSC3	467.8	469.4	50.5	-0.3	10.8	0.0
BO46-04-001	STDSC5	1660.9	1677.5	16.1	-1.0	1.0	-1.0
ARF10286230	STDSC10	917	890.6	23.4	2.9	2.6	1.1

The PSMC assay results are in excellent agreement with the reference values. The largest relative difference observed is -5.5% for STDSC1, which is only -0.7 σ. This sample had an α of 7.6 and was measured for 15 minutes. Standard STDSC3 was measured for 1 hour and had an α value of 12. The PSMC assay result was only -0.3% different from the reference value for STDSC3. Standard STDPD5 was measured for 30 minutes and had an α value of 0.5. While the PSMC assay differed from the reference value by only -2.1%, this represented -4.5 σ for STDPD5. All the other PSMC assay results were within 1.2 σ of the reference values, except for STDPD7, which differed from the reference value by -2.2 sigmas (-2.0%).

The PSMC assay results are also shown in Figure 1. Plotted is the relative assay error (reference value -assay)/reference value or (R-A)/R, in percent vs reference plutonium mass.

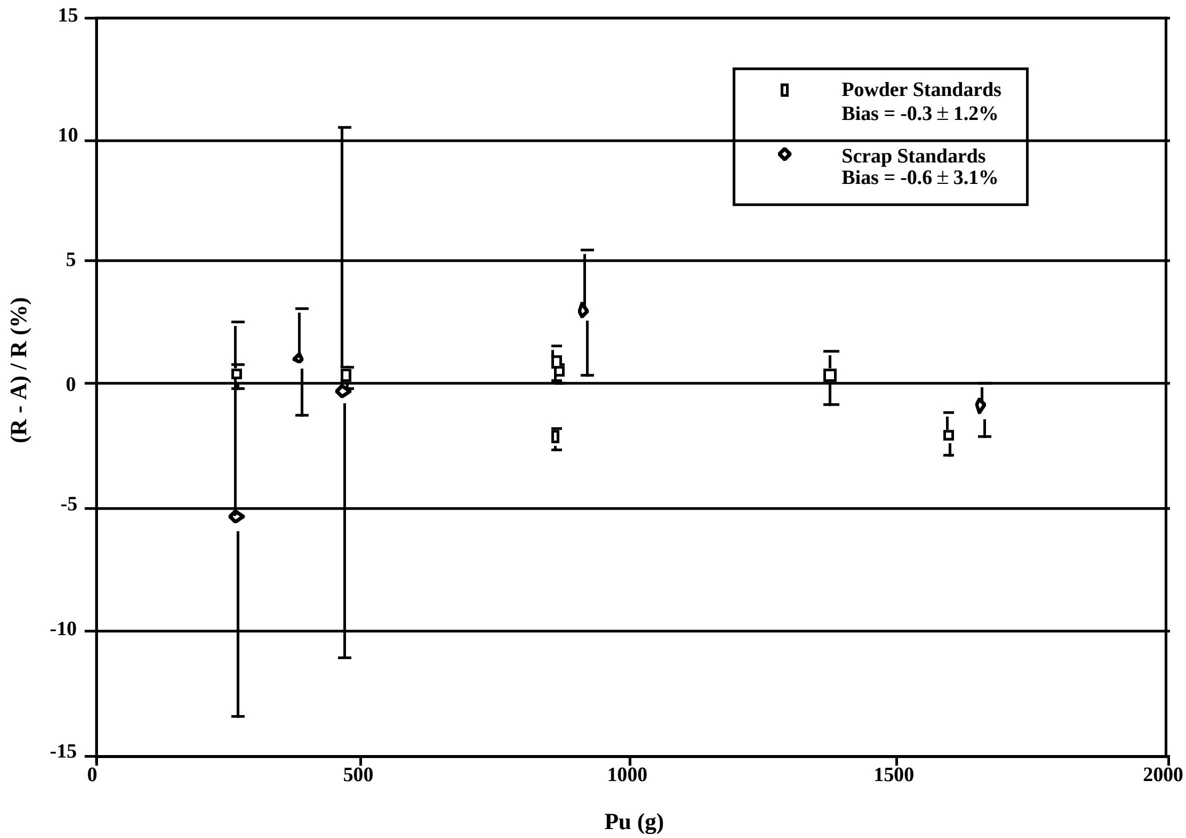

Figure 1. **PSMC assay results compared with reference IAEA values for pure PuO_2 powder and impure plutonium residue standards.**

The average difference between the PSMC assay values for the seven powder standards is -0.3 ± 1.2% and for the five scrap standards is -0.6 ± 3.1%. The overall difference when grouping all 12 standards together is -0.4 ± 2.1%.

CONCLUSIONS

The IAEA is using the PSMC for partial defects measurements of the excess pure plutonium powder and impure plutonium residue items stored in the vault at the Hanford PFP. For partial defects measurements, the expected measurement uncertainty is 6% (1 σ). All the results reported in this paper are within the 6% criterion, and eight of the measurements are within the 1% criterion (bias defects). The PSMC may then be used with confidence for partial defects measurements on the inventory of pure and impure items. For very highly impure items, e.g., for α values above 10, the PSMC may become precision limited for 15–60 minutes count time. These items should be measured in a calorimeter. The combination of the PSMC and calorimetry (both combined with high-resolution gamma spectrometry) provides quite effective and efficient plutonium mass verification for plutonium residue items at Hanford, and minimize the need for sampling and DA. Rocky Flats results are discussed in a separate report.

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