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TITLE **PROTOTYPE FAST NEUTRON COUNTER FOR THE ASSAY OF IMPURE PLUTONIUM**

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PROTOTYPE FAST NEUTRON COUNTER FOR THE ASSAY OF IMPURE PLUTONIUM

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ABSTRACT

A fast coincident neutron counter using liquid scintillators and gamma-ray/neutron pulse-shape discrimination has been constructed for the analysis of plutonium samples with unknown self-multiplication and (α,n) production. The counter was used to measure plutonium-bearing materials that cover a range of masses and (α,n) reaction rates of importance to the safeguards community. Measured values of the ^{240}Pu effective mass differed, on average, from their declared values by 0.4% for plutonium oxides and by -2.2% for metal and MgO-loaded samples. Poorer results were obtained for materials with large (α,n) reaction rates and low self-multiplication such as plutonium ash and plutonium fluoride.

I. INTRODUCTION

For plutonium-bearing materials that undergo (α,n) interactions, such as oxides, metal with oxide coatings, salts, and residues, the present generation of thermal neutron coincidence counters often gives inaccurate assays. This inaccuracy occurs because thermal counters now measure only two quantities, corresponding to coincident and total neutrons, and cannot uniquely specify the plutonium mass for samples with unknown self-multiplication and (α,n) reaction rates. Fast neutron coincidence counters using liquid or plastic scintillators have the capability of counting more than two quantities and can potentially assay these materials. But fast counters have seen limited application because of the sensitivity of the scintillators to gamma rays. Currently a fast neutron counter using liquid scintillators and gamma-ray/neutron pulse shape discrimination is being developed and evaluated for the non-destructive assay of impure plutonium materials. The counter can detect triply coincident neutron events in addition to doubles and singles, but is not limited by the gamma-ray response of the scintillators. This type of counter may provide a direct solution to the measurement problems associated with self-multiplying, impure plutonium samples.

In this paper, the prototype fast neutron counter and the data analysis employed for the measurements will first be described. Then the types of plutonium-bearing materials that have been measured with this counter and the results of these measurements will be discussed. Lastly, some concluding remarks on these results and recommendations for future counter design will be given.

II. MEASUREMENT APPARATUS

The prototype fast neutron counter (Fig. 1) consists of four 10.2-cm-diam by 10.2-cm-deep NE-213 or BC-501 liquid scintillator cells surrounding a 12.7-cm-square sample chamber. In one configuration, all four cells are at the same height and the chamber is 23.0 cm high. In a second configuration, two of the cells are positioned from 10.2 cm to 20.3 cm higher than the

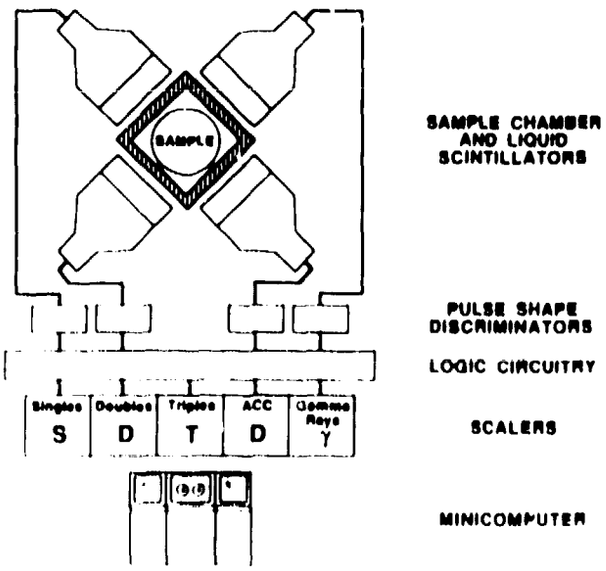

Fig. 1. Sample chamber and prototype fast neutron counter for plutonium analysis.

other two to desensitize the detector to variations in sample fill height. The chamber height then ranges from 38.2 to 48.3 cm. Lead lining of 2.5-cm thickness surrounds the chamber to attenuate the gamma-ray flux from the samples. The top and bottom of the chamber are lined with 2.5 cm of nickel.

High-gain phototubes process signals from the cells and direct them to pulse-shape discriminators[1] that distinguish between neutrons and gamma rays. The discriminators are currently adjusted so that the absolute efficiency of the counter is 4% to 8% for fission neutrons, depending on the chamber configuration. The absolute efficiency for counting 1-MeV gamma rays is about 0.8% to 1.6%. We measured gamma-ray breakthrough, that is, the number of pulses from a strictly gamma-ray source that are counted as neutrons, to be 1% at 20 000 gamma rays/s for these discriminators. The threshold level in the discriminators is adjusted so that pulses with energies less than 60-keV gamma rays are rejected. Total discriminator processing time for a single pulse is about 500 ns. The coincidence logic circuitry consists of conventional fast timing modules and has a resolving time of 30 ns. Single, double, and triple coincidence events are measured and a minicomputer analysis of these data is used to obtain sample mass, self-multiplication, and the ratio of (α,n) to spontaneous-fission neutrons. The number of gamma rays and accidental doubles are also measured but the gamma-ray information is not used in the analysis.

III. DATA ANALYSIS PROCEDURE

The coincidence events must be corrected for a number of effects before the sample characteristics can be determined. Table I summarizes these corrections and their magnitudes. For most samples, the corrections are small, on the order of several percent. For large samples, the subtraction of accidental coincidences is not negligible, but the ratio of accidental to real coincidences is still 3 orders of magnitude less than for thermal neutron coincidence counters. The sample fill-height correction and the normalization correction for instrument drift could be reduced in future fast counters by using more detector cells and stabilizing the photomultiplier tube gain.

The multiple scattering correction is important for samples with very high (α,n) yields because the single neutron emission rate is high compared to the doubles rate. Multiple scattering of neutrons from one detector to another can then cause an appreciable enhancement in the doubles rate. Corrections to the count rates were computed from formulas based on an equation given by Murakami.[2] For this prototype detector, the multiple scattering fraction is roughly 1% to 3% depending on the neutron energy.

TABLE I

FAST NEUTRON COUNTER DATA CORRECTIONS AND THEIR MAGNITUDES

Data Correction	Range of Magnitude (%)
Background subtraction	0.1 to 1
Accidental coincidence subtraction	1 to 50
Deadtime correction	1 to 2
Sample fill-height correction	1 to 15
Normalization correction	1 to 10
Multiple scattering correction	1 to 50
Pileup correction	1 to 50

The pileup correction is large for highly multiplying metal buttons where the neutron fission chains are long. Twenty to thirty prompt and induced fission neutrons and gamma rays may be emitted within a few nanoseconds in these chains. If two or more neutrons or gamma rays enter one scintillator, the pileup circuitry in the pulse shape discriminator will reject the event. This is an example of a count-rate correction that is worse for a fast counter than for a thermal counter because the fission neutrons and gamma rays are so close together in time. The correction factor involves a summation over all orders of fission chains.

After applying corrections to the data, the three corrected rates, S (singles), D (doubles), and T (triples), are used to obtain the three unknown parameters: effective ^{240}Pu mass, m; the ratio of (α,n) to spontaneous-fission neutrons, α; and self-multiplication, M. This procedure requires an iterative solution of the following three equations:

$$m = D/KM^2\,[1 + 2.11\,(M - 1)(1 + \alpha)]\ , \quad (1)$$

$$(1 + \alpha) = r_0\,S/KMm\ , \quad (2)$$

and

$$M^3 + [-1.822 + 0.4739/(1 + \alpha)]M^2 + [0.822 - 0.1215t - 0.4154/(1 + \alpha)]M + [0.1215t - 0.0585t/(1 + \alpha)] - 0\ , \quad (3)$$

where K = linear calibration constant,
$r_0 = D_0\,(1 + \alpha_0)/S_0$,
$t = T/Dt_0$, and
$t_0 = T_0/D_0$.

Use of a nonmultiplying sample provides the reference values α_0, S_0, D_0, and T_0. Equation (1) for m and Eq. (3) for M are derived from K. Böhnel's equations for the reduced moments of the neutron multiplicity distribution.[3]

IV. PLUTONIUM SAMPLES

Plutonium-bearing materials that represent a range of (α,n) yields and plutonium mass of interest to the safeguards establishment were selected for measurement. Samples of plutonium metal, oxide, fluoride, ash, and impure oxide with high-MgO content were measured in quantities ranging from 1 to 150 g of ^{240}Pu. The material was contained in 0.16-cm-thick steel cans, which were positioned on a pedestal 6.0 to 10.0 cm above the chamber floor depending on the configuration. Table II lists the types and quantities of materials measured.

TABLE II

MATERIALS USED IN THE FAST NEUTRON COUNTER ANALYSIS

Material	^{240}Pu Mass (g)	Total Pu (g)
Pu oxide	2 - 150	20 - 880
Pu oxide with MgO (450-600 g of MgO)	7 - 15	120 - 240
PuF_4	3 - 18	50 - 300
Pu metal	34 - 116	585 - 2000
Pu ash	7 - 29	--

V. MEASUREMENT RESULTS

Values for the ^{240}Pu effective mass were determined using a single-point, straight-line calibration and 1 000-s counting times. Table III shows the results of the data analysis for plutonium oxide samples along with previously ascertained values of α, M, and m (determined by calorimetry and Monte Carlo methods) and the percent difference between the measured and declared mass values. Table IV presents these same quantities for the measurements of impure oxides (containing MgO), metal, ash, and PuF_4 samples. The percent difference between the measured and previously determined mass values averaged 0.4% ± 3.1% for the oxide samples and -2% ± 10% for the impure oxide and metal samples.

TABLE III

FAST NEUTRON COUNTER MEASUREMENT (MEAS) RESULTS AND DECLARED (DECL) VALUES OF α, M, AND m FOR PLUTONIUM OXIDE SAMPLES
(The percent difference between the measured and declared mass values is given in the last column.)

Plutonium Type	Declared α	Declared M	Declared m (g)	Meas α	Meas M	Meas m (g)	$\frac{^{240}m_{MEAS} - {}^{240}m_{DECL}}{^{240}m_{DECL}}$
Oxide	0.60		2.0	0.84	0.992	1.9	-3.0
	0.60		7.4	0.79	0.994	7.1	-3.9
	0.60		14.8	0.77	1.009	15.5	+4.4
	0.60		29.7	0.76	1.022	30.5	+2.8
	0.61	1.046	43.4	0.77	1.039	42.7	-1.5
	0.59	1.056	65.0	0.73	1.034	65.3	+0.4
	0.60	1.061	81.4	0.73	1.043	79.9	-1.8
	0.60	1.061	81.4	0.69	1.058	83.6	+2.7
	0.40	1.018	10.1	0.39	0.993	9.9	-1.5
	0.40	1.018	10.1	0.38	0.992	10.5	+4.4
	0.40	1.036	29.3	0.39	1.004	29.7	+1.3
	0.40	1.054	54.4	0.42	1.019	53.1	-2.3
	0.40	1.054	54.4	0.42	1.017	53.5	-1.6
	0.40	1.059	65.2	0.40	1.026	64.5	-1.0
	0.41	1.071	92.2	0.46	1.036	87.5	-5.1
	0.41	1.071	92.2	0.40	1.042	96.0	+4.1
	0.40	1.075	104.6	0.43	1.042	102.7	-1.8
	0.40	1.088	144.5	0.40	1.048	142.8	-1.2
	0.40	1.090	149.2	0.38	1.068	157.5	+5.6

TABLE IV

FAST NEUTRON COUNTER RESULTS AND DECLARED VALUES OF α, M, AND m FOR IMPURE OXIDE, METAL, ASH, AND PuF_4 SAMPLES
(The percent difference between the measured and declared mass values is given in the last column.)

Plutonium Type	Declared α	Declared M	Declared m (g)	Meas α	Meas M	Meas m (g)	$\frac{^{240}m_{MEAS} - {}^{240}m_{DECL}}{^{240}m_{DECL}}$
Impure	4.75	1.019	7.7	5.11	0.994	7.2	-6.5
Oxide	2.78	1.034	15.3	2.93	1.002	16.0	+4.3
	2.78	1.034	15.3	2.98	1.010	16.3	+6.3
Metal	>0	-	345	0.73	1.217	27.4	-20.6
	0	1.480	57.7	-0.01	1.517	58.5	+1.6
	0	1.960	115.6	0.14	2.000	117.4	+1.6
Ash	-	-	10.8	3.6	0.999	13.6	+26
	-	-	20.3	7.4	1.018	22.1	+8.9
	-	-	16.9	5.02	1.023	27.2	+61
	-	-	18.2	7.72	1.019	23.0	+26
PuF_4	~100	-	3.0	302	1.011	3.0[a]	-
	~100	-	18.0	23	1.008	66.6	+370

[a]Mass was assumed to be 3.0 g for instrument calibration.

Measured values for the self-multiplication and (α,n) ratio were not determined as accurately as sample mass. The values for M - 1, which is related to the fission probability, typically differed by 50% from values obtained by Monte Carlo calculations. Measured values for 1 + α, the total sample neutron yield, generally differed by 10% from known values. Not included in the averages above are measurements of the ash and PuF_4 samples, which did not give correct assays. The multiple scattering correction is not yet sufficiently understood to assay samples with such high (α,n) yields.

The accuracy of the measurements is depicted in Fig. 2, which graphs the percent mass difference as a function of ^{240}Pu mass for oxide, metal, and impure oxide samples. Values for the ash and PuF_4 samples are not shown because they do not conform to the scale of the graph. The graph indicates that more than 80% of these measured mass values fall within ±5% of their calorimetric values. For samples with less than 15 g of ^{240}Pu, there is a trend toward slightly larger percent differences.

Measurement precision is shown in Fig. 3 as a function of ^{240}Pu effective mass with samples categorized according to their α ratio. The figure indicates the loss of precision that occurs with increases in α and also reveals the decreased measurement reproducibility for low-mass samples because of their reduced count rates. Reduced reproducibility also accounts for the slightly higher percent mass difference ratios for the low-mass samples seen in Fig. 2. The relative standard deviation (RSD) for all samples with masses less than 15 g was 8.2%; while for samples with mass greater than 15 g, the RSD was 1.6%. Figure 3 also indicates that the RSD increases for large metal samples, (α = 0) after a minimum at about 70 g. This may be due to pulse pileup at the higher counting rates for these samples. The results can be compared to those found by Krick and Swansen[4,5] who have developed a novel thermal neutron counter using multiple ^3He detectors and shift-register coincidence circuitry. The comparison indicates that, for samples with small amounts of ^{240}Pu, the thermal counter produces more precise measurements than the fast counter described here. This occurs because of the greater absolute efficiency of the thermal counter. However, for larger mass values the thermal counter has poorer reproducibility for some materials because of uncertainties in unfolding data from the many ^3He tubes at higher count rates.

VI. CONCLUSIONS

The goal of a fast neutron counter is to correctly determine the plutonium content of any sample without prior knowledge of its self multiplication or (α,n) rate. The present prototype meets this goal to within 5% for many in-plant materials containing more than 15 g of ^{240}Pu. For materials with high (α,n) yields and

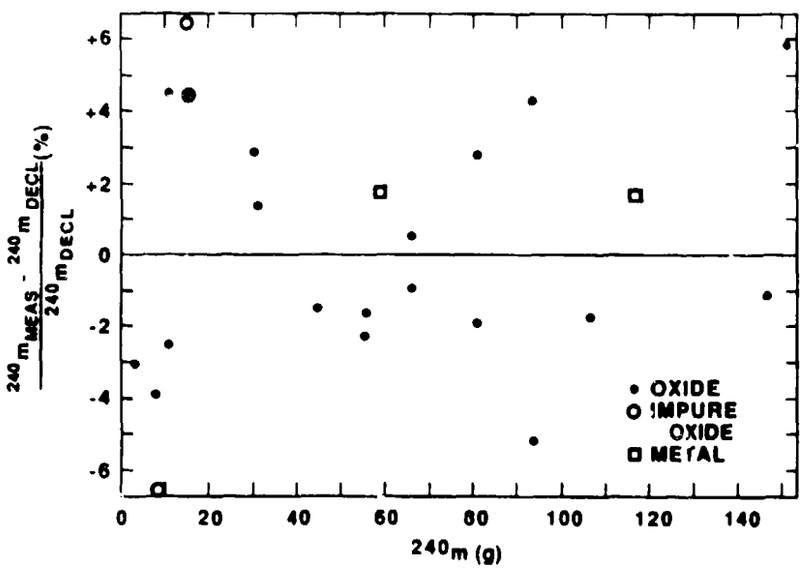

Fig. 2. Percent mass differences for plutonium oxide, impure oxide, and metal samples graphed as a function of ^{240}Pu effective mass.

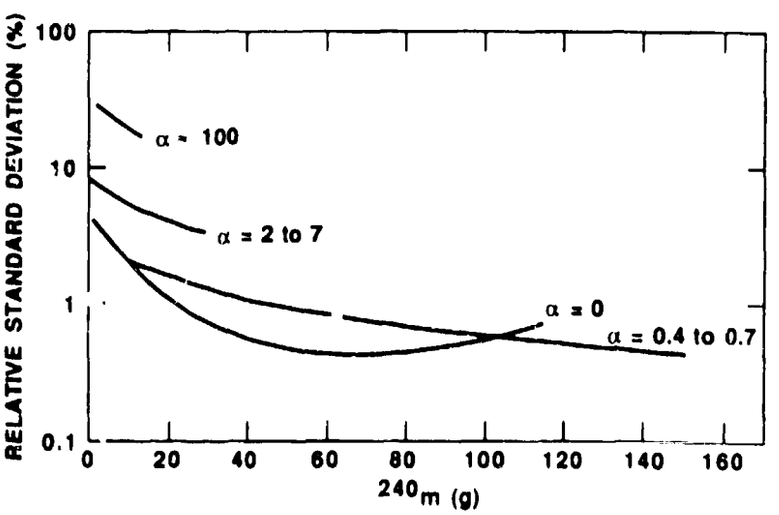

Fig. 3. Relative standard deviation (1σ) based on 1 000-s measurements shown for plutonium samples ranging from 2 to 150 g.

low self-multiplication such as plutonium-bearing ash or PuF_4, however, the sample masses measured to date are in error by 10% to 50% or more. This is probably due to the large and incompletely understood pileup and multiple scattering corrections.

To increase the accuracy of the fast neutron counter for in plant materials, improvements are needed in several areas. The counter design can be improved by adding more detectors to flatten the vertical efficiency profile and reduce the pileup correction. Measurements with a variety of (α,n) sources are needed to better determine the multiple scattering fraction as a function of neutron energy. The detector design itself should attempt to make the neutron detection efficiency and multiple scattering fraction independent of neutron energy. Gain stabilization circuitry is needed to provide long-term stability and to bring the assay accuracy for pure samples close to 1%.

We are encouraged by the results obtained with the prototype fast neutron counter, but the above-mentioned improvements are needed to develop a second, better prototype. Alternative techniques for neutron detection and/or multiplicity analysis should also be pursued so that the ultimate goal of an accurate, matrix-insensitive safeguards instrument can be realized.

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