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A HIGH-EFFICIENCY NEUTRON COINCIDENCE COUNTER FOR SMALL SAMPLES*

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ABSTRACT

The inventory sample coincidence counter (INVS) has been modified to enhance its performance. The new design is suitable for use with a glove box sample-well (in-line application) as well as for use in the standard at-line mode. The counter has been redesigned to count more efficiently and be less sensitive to variations in sample position. These factors lead to a higher degree of precision and accuracy in a given counting period and allow for the practical use of the INVS counter with gamma-ray isotopics to obtain a plutonium assay independent of operator declarations and time-consuming chemical analysis. A calculational study was performed using the Los Alamos transport code MCNP to optimize the design parameters.

INTRODUCTION

The inventory sample coincidence counter (INVS)[1,2] was developed to passively assay small plutonium samples by using neutron coincidence counting techniques. The original counter was recently upgraded. We redesigned the detector head and incorporated the high-performance AMPTEK preamplifier.[3] The INVS counter has been widely used by the International Atomic Energy Agency in its inspection activities at various nuclear facilities throughout the world. One facility in particular, the Plutonium Fuel Production Facility (PFPF) in Japan, has been the site of advanced use of the instrument; the INVS counter is coupled to a sample-well underneath a glove box in the analytical area. The isotopic composition of the sample is determined by gamma-ray spectroscopy in this area as well. At PFPF, the INVS counter is used with several other specialized neutron coincidence counters to provide comprehensive verification of the plutonium in the plant.

Because samples that are measured with the INVS counter are typically assumed to represent larger batches, it is important that the assay be as precise and as bias-free as possible. Experience with the original counter has shown that variations in the sample placement can lead to biases of a few percent. This sensitivity to sample position can be important because of the way samples are packaged before counting. Samples to be counted are placed in vials, which are in turn placed in larger vials. The doubly contained plutonium is finally placed in a large plastic wrap for bagging out of the glove box. This packaging reduces the ability of the instrument user to accurately locate the sample in the INVS cavity, which in turn can lead to biases in the assay.

To address this problem, we set out to redesign the counter by using Monte Carlo[4] simulation techniques to vary parameters in search of optimum performance. Goals of the calculational study included increased efficiency, decreased counter-response variation as a function of sample position, retention of portability, and the ability to couple the counter with a glove box sample-well extension to avoid bagging out the sample.

HARDWARE DESCRIPTION

Figure 1 is a schematic cross section of the INVS Mod-III counter showing materials of construction, major dimensions, and the coupling to the glove box sample-well at the sampling station at PFPF. A notable difference in the new design is the presence of graphite in the endplug regions of the counter. The addition of graphite in this position helps to improve the counter's insensitivity to sample position variations. The characteristics of the original and the redesigned INVS counters are compared in Table I. Figure 2 is a photograph of the counter with the inner graphite/polyethylene assembly removed. When the assembly is in place, the sample cavity is 5 cm in diameter by 15 cm in height, which is the standard configuration for use under the glove box. If this assembly is removed, larger diameter samples (up to ~9 cm) may be measured.

The ^{3}He detector tubes are arranged in two rings of nine tubes each. The inner ring of tubes is located at a radius of 7.2 cm and the outer tube ring is at a 10.6-cm radius. Three AMPTEK preamplifiers are used, two for the inner

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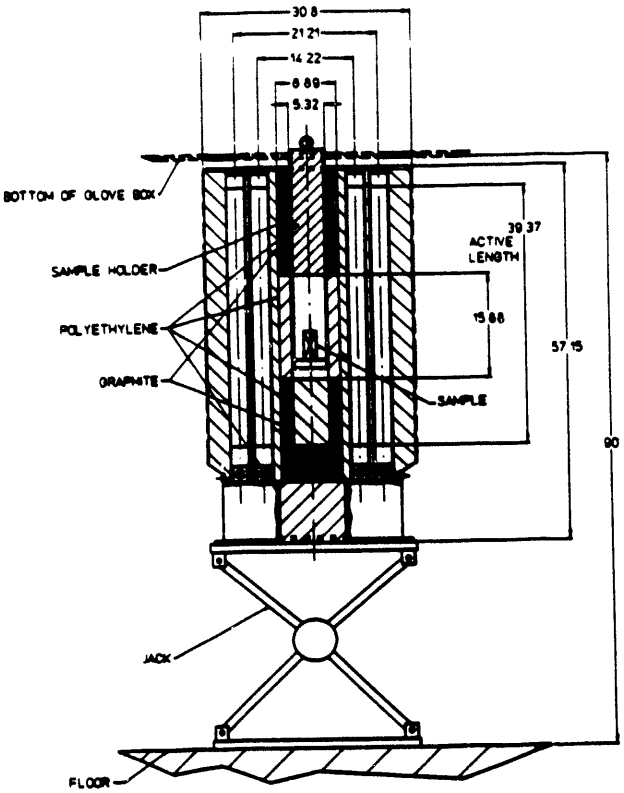

Fig. 1. Cross section of the INVS Mod-III counter showing materials of construction, major dimensions, and coupling to the sample-well from under the glove box at PFPF. The sample holder also acts as a part of the top endplug assembly because its top region is made of high-density polyethylene.

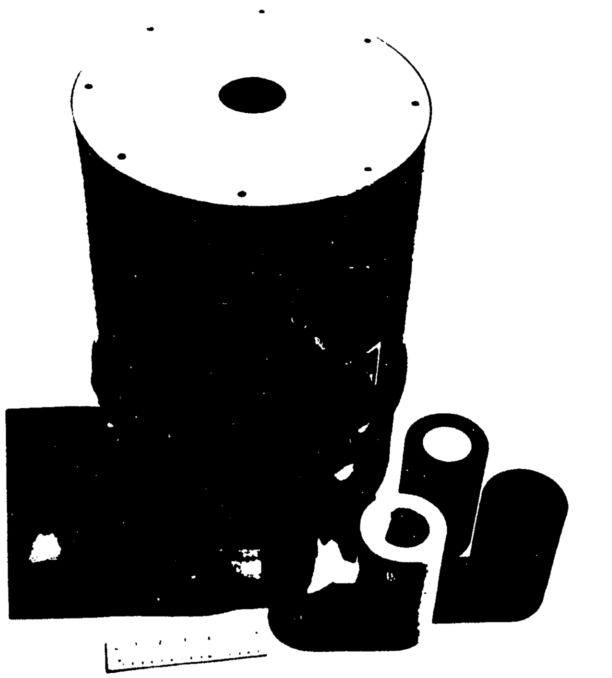

Fig. 2. Photograph of the INVS Mod-III counter with endplug assembly removed. The parts that are made of graphite are positioned at the top and bottom of the counter and help to minimize response variations as a function of sample location. When the assembly is removed, larger samples may be measured.

Table I. Comparison of Original and Redesigned INVS Counters

Item	Original INVS	Mod-III INVS
Number of tubes	16	18
Active length	30 cm	39 cm
^{3}He pressure	4 atm	6 atm
Height	48 cm	57 cm
Diameter	27 cm	30 cm
Cadmium (inner)	Yes	No
Graphite	No	Yes
Efficiency	30.4%	42.3%

tube ring and one for the outer ring of tubes. Two replaceable desiccant holders are incorporated into the junction box from the side to allow access while the counter is in position under the glove box.

MONTE CARLO DESIGN OPTIMIZATION

The Los Alamos transport code, MCNP, was used to simulate the INVS Mod-III counter to facilitate the parametric study required to optimize performance of the new counter. Design questions, which were readily answered by use of the calculational model, include (1) ^{3}He detector-tube placement, (2) location and type of material in the detector moderator assembly, and (3) overall dimensions of the counter. Figure 3 shows the input geometry for the Monte Carlo model in plan and cross-sectional view. The segmented portions of the inner assembly show areas where different materials were evaluated to obtain optimum response uniformity. Results of the Monte Carlo calculations were used to estimate totals and reals count rates.

A ^{252}Cf point source was used in the calculational model and moved to various positions axially and radially in the sample cavity. Deviations from uniformity in the response under these conditions are conservative compared to actual samples that are distributed over a finite volume. The design goal for response uniformity was less than 1% deviation in total efficiency for a minimum of 6 cm of axial

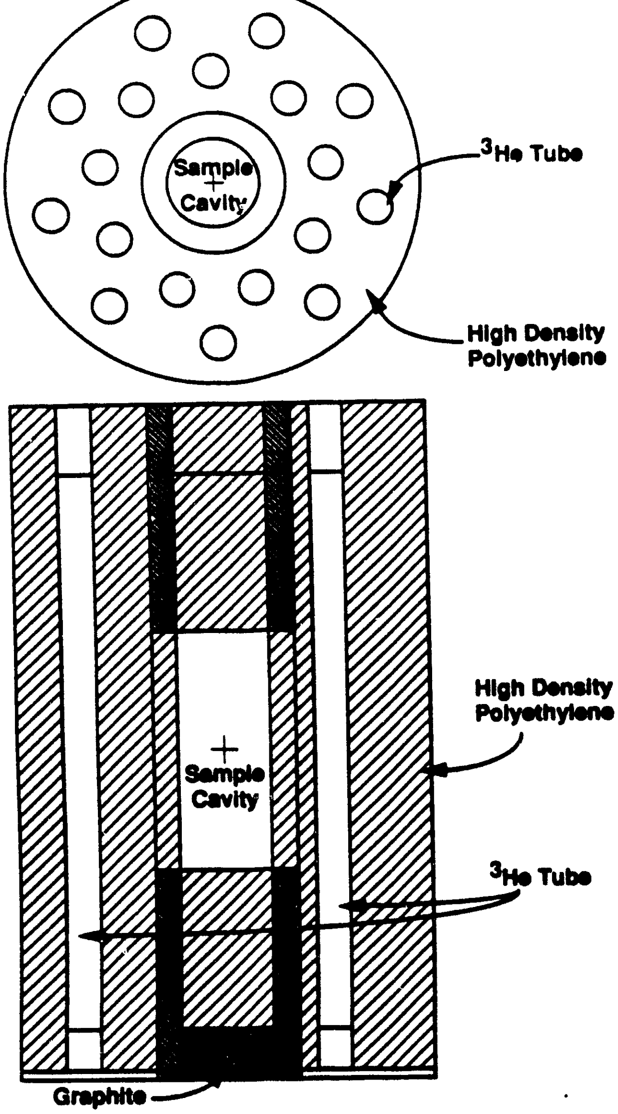

Fig. 3. Geometry used for the Monte Carlo simulation of the INVS Mod-III counter in plan and cross-sectional view. Segmented portions of the inner assembly show areas where different materials were evaluated for optimum response uniformity.

movement and 1 cm of radial movement, corresponding to the size of the standard sample container. Figure 4 presents the results of the Monte Carlo simulation for movement of a ^{252}Cf point source in the sample cavity. As can be seen from Fig. 4, the design goal for response uniformity was achieved. In addition to attaining a more uniform response, the overall efficiency was increased from 30% to 40% with the new design.

The enhanced performance of the INVS Mod-III counter is a result of (1) increasing the number and active

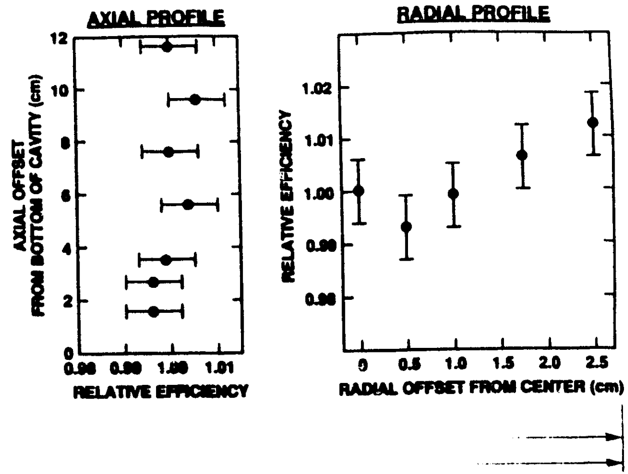

Fig. 4. Axial and radial response profiles from the Monte Carlo simulation using a ^{252}Cf point source. The design goal of less than a 1% deviation in total efficiency for at least 6 cm in the axial and 1 cm in the radial direction was achieved.

length of the ^{3}He detector tubes, (2) removing the cadmium liner from the sample cavity, (3) incorporating graphite into the endplug assembly, and (4) increasing the counter diameter. The use of Monte Carlo simulation techniques allowed for the optimal combination of these parameters within the given design constraints. This type of approach to the design of thermal-neutron coincidence counters is quite useful because the counter does not need to be modified after fabrication to meet the desired performance criteria.

PERFORMANCE CHARACTERISTICS

The performance of the INVS Mod-III was measured by using a ^{252}Cf source. We measured the counter's efficiency, die-away time, deadtime coefficients, response profiles for axial and radial source displacement, and the effect of plastic surrounding the sample. Table II summarizes these measurements.

The counting efficiency of the INVS Mod-III increased significantly over the original design: from 30.4% (with 4-atm tubes) to 42.3%. Thus the count time required to obtain a given precision for reals was reduced by a factor of 1.4. Axial and radial response profiles were made using a ^{252}Cf source. Figures 5 and 6 show the results of the profile measurements. The original and Mod-III INVS counters are compared in Fig. 5, which plots the relative coincidence response vs axial sample position. As shown in Fig. 5, the new design extends the flat-response region of the counter from ~5 cm (original INVS) to ~11 cm. The length of this uniform response region in the sample cavity is larger than the height of a typical sample vial. Axial profiles were also taken with different radial offsets. The results for radial positions of 0 cm, 1 cm, and 2 cm are presented in Fig. 6. Significant deviation in the coincidence response is not

Table II. Summary of Performance Testing	
Efficiency[a]	42.3%
Die-away time[b]	57 µs
Deadtime coefficients	a = 1.23 µs
(3 AMPTEK channels)	b = 0.615 µs²
Axial flat region[c]	11 cm
Plastic bag effect[d]	<1%

[a]Measured with source CR-5 on 900427, which gave 6268 counts/s compared to the absolute yield of 1.482 x 10^4 n/s.
[b]Average result from 32/64 µs and 64/128 µs gate settings.
[c]Response deviation of <2% for reals.
[d]Reals response deviation with up to 70 g (0.6 g/cm³) of polyethylene surrounding source.

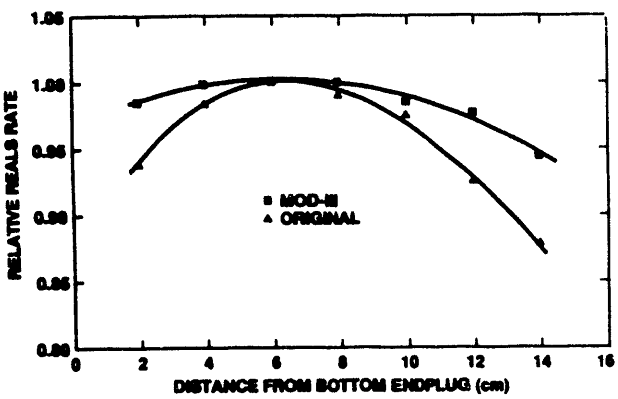

Fig. 5. Coincidence response variation as a function of axial position for the original and Mod-III INVS Counters.

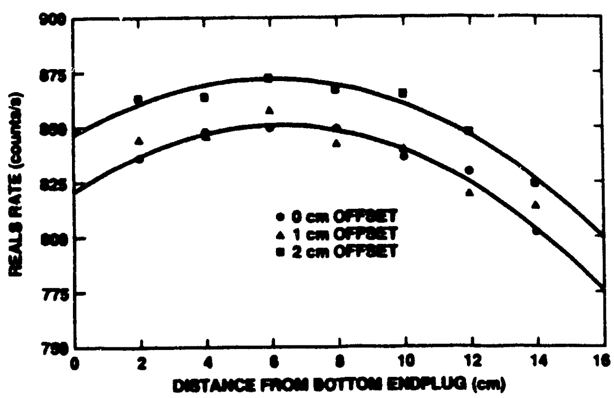

Fig. 6. INVS Mod-III coincidence response variation as a function of axial position for various radial offsets. Standard sample vials are 2 cm in diameter.

observed until the radial position is greater than 1 cm. A typical sample vial has a 2-cm diameter. The effect of varying amounts of plastic wrap were evaluated by putting the source into differing numbers of plastic bags. For plastic densities of up to 0.6 g/cm³, corresponding to 70 g of polyethylene, the reals rate varied less than 1%.

CALIBRATION

The INVS Mod-III counter was calibrated at Los Alamos before shipment to PFPF in Japan, where final calibration took place. At Los Alamos, five mixed oxide (MOX) fuel pellets containing from 17 to 90 mg of ^{240}Pu-effective were used to perform the precalibration. Each pellet was placed at the 8-cm axial sample position and counted for several cycles. Table III is a summary of the precalibration data.

Table III. Summary of precalibration data (900525)

ID	^{240}Pu-eff (mg)	$\bar{R} \pm \sigma$	R/g ^{240}Pu-eff
A1-066	16.8	1.584 ± 0.011	94.3 ± 0.7
A1-119	32.7	2.997 ± 0.029	91.7 ± 0.9
A1-089	48.9	4.409 ± 0.022	90.2 ± 0.4
A1-081	56.0	5.264 ± 0.013	94.0 ± 0.2
A1-078	88.8	8.167 ± 0.016	92.0 ± 0.2
A1-081 + A1-066	72.8	6.933 ± 0.021	95.2 ± 0.3

A Deming fit[5] of the data to the function,

$$R = aM$$

where M is the ^{240}Pu-effective mass, gives a value for the slope $a = 92.77 \pm 0.63$.

At PFPF, two samples each of MOX powder and pellets were used to perform the final calibration. The INVS counter was positioned under the glove box so that the distance from the bottom of the glove box to the top of the INVS counter was 8 cm, corresponding to the optimum position based on profile measurements. The ^{240}Pu-effective mass of the samples ranged from 250 mg to 400 mg. Samples 1 and 4 are in the form of powder, while samples 2 and 3 are pellets.

Table IV shows the data obtained by counting neutrons from the powder and pellet samples.

Fitting this data to the functional form listed above yields a slope, $a = 90.41 \pm 0.73$, which agrees with the precalibration value to better than 3%. Note that a single calibration is sufficient for the measurement of either powder or pellets. A possible source of systematic difference in the

Table IV. Summary of final calibration data (900626)			
ID	^{240}Pu-eff (mg)	$\bar{R} \pm \sigma$	R/g ^{240}Pu-eff
1	243.9	21.53 ± 0.14	88.3 ± 0.6
2	386.0	34.70 ± 0.17	89.9 ± 0.4
3	393.4	35.65 ± 0.18	90.6 ± 0.4
4	400.4	36.85 ± 0.18	92.0 ± 0.4

precalibration and final calibration results is the presence of the stainless steel sample-well at PFPF. The steel walls in the center of the detector will absorb a few percent of the thermal neutrons, but this structure was not included in the precalibration. Monte Carlo simulation of the effect of the stainless steel glove-box tube indicates a decrease in counter efficiency of ~1.7% because of the presence of the steel.

COMBINATION WITH GAMMA-RAY DETECTOR

Modification of the lower endplug assembly of the INVS Mod-III counter allows for the insertion of a high-purity germanium (HPGe) detector for simultaneously acquiring neutron (total and coincidence counts) and gamma-ray (spectral) data. Because of the small size of the INVS cavity, inserting a standard HPGe detector into the cavity significantly and unacceptably distorts the response profile and reduces the efficiency. Therefore a customized HPGe detector design was specified to minimize these effects. The outer diameter of the HPGe detector end-cap was reduced to 3.8 cm (from 6.4 cm) over a total length of 15 cm. In addition, the space behind the crystal was filled with a 3.4-cm diameter by 15-cm-long polyethylene sleeve (with a through-hole for the cold finger) to further reduce the loss of moderation caused by the penetration. Figure 7 illustrates the new HPGe detector design.

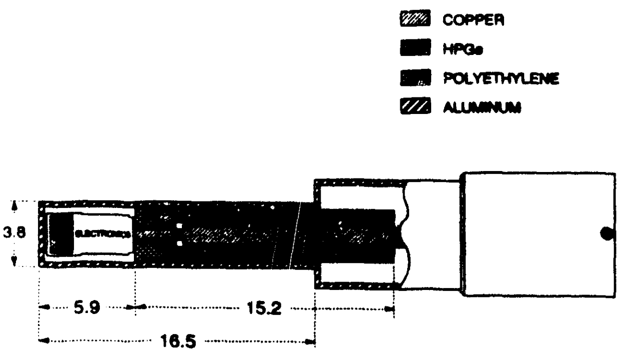

Fig. 7. Schematic drawing of the HPGe detector that can be inserted into the modified end plug assembly of the INVS Mod-III to allow simultaneous gamma-ray and neutron counting. The polyethylene sleeve behind the crystal serves to reduce the adverse effect on the neutron counter caused by insertion of the HPGe detector.

The combination of the INVS counter with an HPGe detector allows for the determination of plutonium mass in a single measurement without the need for destructive analysis. As such, it provides a mechanism for decreasing the number of samples that require destructive analysis and increasing the number of samples that can be measured in a given time. The combined neutron coincidence counter and gamma-ray detector system will undergo a facility test and evaluation in the coming year.

SUMMARY

The INVS Mod-III counter design is an improvement over the original design in both absolute counting efficiency and response uniformity. These enhancements allow the counter to perform precise and accurate assays of small plutonium samples in a timely manner. Count times can be reduced by a factor of 1.4 compared to the original INVS design and still arrive at the same precision based on counting statistics. The flat region of the sample cavity has been increased to 11 cm from the original value of 5 cm, thus allowing a much greater leeway in sample placement without causing biases in the result. The use of Monte Carlo transport methods for detector simulation has proven to be quite useful in optimizing the design of the counter. A combined INVS/gamma-ray detector system has been designed for the simultaneous measurement of neutron and gamma-ray signatures. This new system will allow the determination of a sample's plutonium content in a single measurement.

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