THE DESIGN OF A HIGH-EFFICIENCY NEUTRON COUNTER FOR WASTE DRUMS TO PROVIDE OPTIMIZED SENSITIVITY FOR PLUTONIUM ASSAY

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ABSTRACT

An advanced passive neutron counter has been designed to improve the accuracy and sensitivity for the nondestructive assay of plutonium in scrap and waste containers. The High-Efficiency Neutron Counter (HENC) was developed under a Cooperative Research and Development Agreement between the Los Alamos National Laboratory and Canberra Industries. The primary goal of the development was to produce a passive assay system for 200-L drums that has detectability limits and multiplicity counting features that are superior to previous systems. A detectability limit figure of merit (FOM) was defined that included the detector efficiency, the neutron die-away time, and the detector's active volume and density that determine the cosmic-ray background. Monte Carlo neutron calculations were performed to determine the parameters to provide an optimum FOM. The system includes the ^{252}Cf "add-a-source" feature to improve the accuracy as well as statistical filters to reduce the cosmic-ray spallation neutron background. The final design gave an efficiency of 32% for plutonium with a detector ^{3}He tube volume that is significantly smaller than for previous high-efficiency systems for 200-L drums. Because of the high efficiency of the HENC, we have incorporated neutron multiplicity counting for matrix corrections for those cases where the plutonium is localized in nonuniform hydrogenous materials. The paper describes the design and performance testing of the advanced system.

INTRODUCTION

The measurement of plutonium content in waste containers is required prior to long-term storage and disposal. Measurement accuracy should meet the requirements of safeguards, material accountancy, criticality control, and environmental regulations. It has long been recognized that the passive neutron coincidence assay of typical waste containers is intrinsically more accurate than active neutron techniques because of the excellent penetrability of the passive spontaneous fission neutrons. However, the signal level of the spontaneous fission neutrons is low, which limits the sensitivity, and it is necessary to know the plutonium isotopics to convert the measured ^{240}Pu-effective to total plutonium.

We have developed a passive neutron counter with advanced design features to improve the detectability limit and to make the system accurate for a large range of matrix materials. To improve the detectability limit, the efficiency was increased, the cosmic-ray background was decreased, and external shielding was increased to reduce the room background neutron rate.

The dependence on the waste matrix was reduced by incorporating the ^{252}Cf add-a-source correction and multiplicity counting to make corrections for localized shielding.

Joint development of the advanced passive neutron assay system was accomplished under a cooperative research and development agreement (CRADA) between Canberra Industries and the Department of Energy. Table I lists design goals for successful completion of this CRADA. The relative weighting and prioritization of these goals was determined by Canberra, based on market considerations. The parameters that can be used to meet these goals include sample cavity size, active detector volume, detector efficiency, die-away time, shielding (both internal and external), moderator materials, electronic background rejection, and add-a-source.

Table II lists some of the design parameters that can be used to optimize the system design. The cavity size for the system was set to be the same as the Model WM3100 to take advantage of the existing mechanical system. Smaller cavity dimensions would give higher efficiencies and less expensive fabrication costs. However, the requirement to accommodate samples somewhat larger than 200-L drums dictated the WM3100 cavity size.

The design goals that are based on counting statistics, such as precision, can be met by higher efficiency and a lower die-away time as well as by smart software that terminates a measurement based on the statistical error rather than a preset run time.

In general, multiplicity counting will require higher efficiency than simple doubles counting, and calculations have been performed to provide the statistical error in multiplicity counting.[1]

Competitive cost criteria determine many of the design parameters (e.g., number of He tubes, number of detector banks, and shielding). A modular design is the key in meeting competitive pricing factors, and the WM3100 provides a good design platform to allow a modular approach.

TABLE I. Design Goals for the Waste Drum Counter.
1. Low detectability limit (good sensitivity at low mass)
2. Ability to meet Performance Demonstration Program (PDP) requirements a. High (α,n) backgrounds b. Variable Pu distribution c. Accuracy requirements
3. Matrix independence a. Pu distribution independence b. Accurate matrix corrections
4. Modular detector design a. high/low efficiency b. high/low shielding c. with/without add-a-source d. with/without multiplicity e. flexible software

TABLE II. Design Parameters for the Waste Drum Assay System	
1.	Cavity size
2.	Active detector volume
3.	Detector efficiency
4.	Detector die-away time
5.	Internal shielding
6.	External shielding
7.	Moderator materials
8.	Statistical background rejection
9.	Multiplicity counting
10.	Add-a-source

DETECTABILITY LIMIT

Optimization of the detectability limit was one of our primary design goals. To obtain a low detectability limit, we need a high counting efficiency as well as a small active detector volume, a large coincidence gate fraction, and a small neutron background rate from the room.

Two of these design parameters work in opposition to each other. That is, the higher-efficiency designs require a larger active volume for the detector. The problem with the large active detector volume is that the cosmic-ray spallation background increases as detector volume and density increase. A detector with three rings or layers of ^{3}He tubes will have a larger detectability limit than a two-ring or one-ring detector because of the increased detector volume for three rings. Also, a large detector volume displaces the external neutron shielding, resulting in an increased measured background from room source neutrons (e.g., drums stored near the detector).

The detectability limit can be obtained from totals counting or coincidence counting. In general, the totals-based limit is lower than the coincidence-based limit because of the high totals counting efficiency. However, variable room background rates and unknown sample (α,n) rates make the totals results difficult to interpret so we will use the coincidence rate for our detectability limit calculation. The limit based on totals neutrons is still a useful screening tool to pass uncontaminated samples and to set an upper limit on the plutonium contamination. The totals neutrons also provide a good measurement of any alpha decay in the waste because of the (α,n) reactions in the waste materials.

The detectability limit *d* (in grams of ^{240}Pu) at 3 standard deviations above background can be calculated for the counter using the equation

$$d = (3/a)\cdot\left(\frac{B+ad}{t}\right)^{1/2} , \qquad (1)$$

where

a	=	response of counter in counts/(s • g ^{240}Pu)
B	=	room background rate (based on a counting time much greater than *t*), and
t	=	counting time.

Equation (1) is an approximation based on a long counting time for the cosmic-ray background and a negligible accidental background from the room totals rate. The detectability limit is a function of the neutron coincidence background; we can reduce the background by eliminating the cosmic-ray spallation events with high multiplicity by using a statistical filtering technique.[2]

For coincidence counting, both the calibration constant *a* and the background *B* are coincidence rates that depend on the efficiency squared. Thus, from Eq. (1) we get

$$d \sim \frac{\sqrt{\varepsilon^2}}{\varepsilon^2} = \frac{1}{\varepsilon} \quad . \tag{2}$$

However, the background term is complex and it contains two primary components—the cosmic-ray spallation rate and the accidental coincidence rate from the room source totals rate.

The cosmic-ray spallation neutrons increase with the active volume and density of the ^{3}He tube area moderator. Thus, if we double our detector volume or density, we will approximately double our cosmic-ray spallation background. The additional cosmic-ray background from spallation reactions in the sample drum is negligible for hydrogenous drums because the drum absorbs as many neutrons as it creates. Of course, if the drum contains high-density materials or metals, it becomes a background source of cosmic-ray coincidence neutrons. Figure 1 shows the cosmic-ray coincidence background as a function of drum loading.[2] We see that most hydrogenous (combustibles) loadings and concrete rubble have the same background as the empty case. Pure polyethelene reduces the cosmic-ray background because the CH_2 matrix absorbs more neutrons than it creates by spallation reactions.

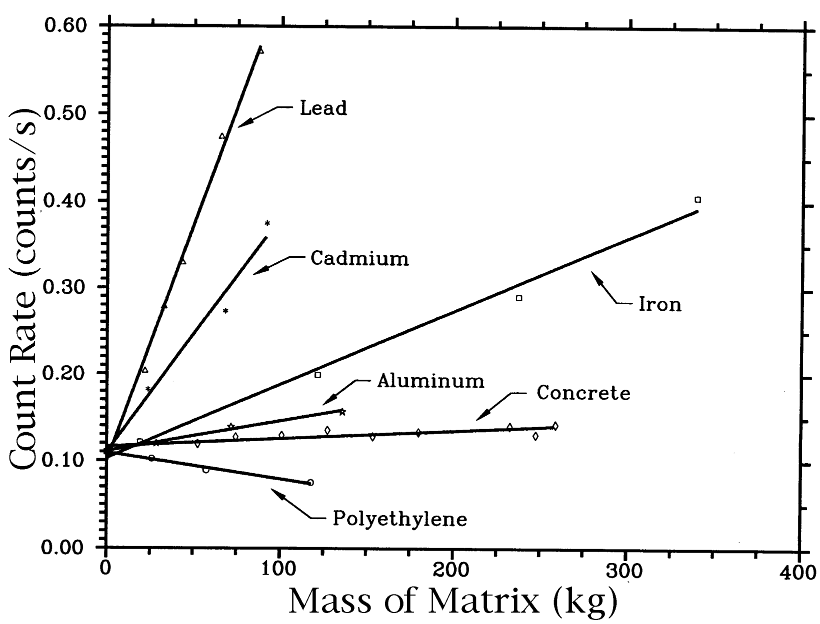


Fig. 1. Cosmic-ray coincidence backgrounds as a function of matrix materials in the shielded JCC-21. Most combustibles give no change in the background rate.

The accidental coincidence rate from room source totals neutrons can be calculated from

$$A = T^2 G \quad , \tag{3}$$

where

A = accidental coincidence rate,
T = totals background rate, and
G = gate width(s).

The coincidence background can be written as

$B = B$ (cosmic ray) + B (room accidentals)

and

B (cosmic ray) ~ ε^2 • volume • density.

As a first step in the design optimization, we removed all of the cadmium and heavy metal in the detector to reduce the cosmic-ray spallation rate. The room source neutrons are relatively easy to remove with external shielding; however, the cosmic-ray background is several orders of magnitude harder to reduce.

The detector's active volume can be estimated by the ^{3}He tube and moderator thickness as shown in Fig. 2. The back edge (away from the sample) boundary merges with the CH_2 used for the external shielding. The distance from the edge of the back tube to the shielding boundary was defined as one diffusion length (2.73 cm) of a thermal neutron in CH_2.

Figure of Merit

To aid in the design optimization based on the detectability limit, we defined a figure-of-merit (FOM) as follows:

$$FOM \sim \frac{1}{d} \sim \varepsilon^2 \left[\frac{f_g}{\varepsilon^2 \bullet t \bullet \rho} \right]^{1/2} \text{, or} \qquad (4)$$

$$FOM = \varepsilon \left(\frac{f_g}{t \bullet \rho} \right)^{1/2} , \qquad (5)$$

where

ε = totals counting efficiency (%),
ρ = moderator density,
t = moderator thickness (cm), and
f_g = the fraction in the gate for a 128-μs gate length.

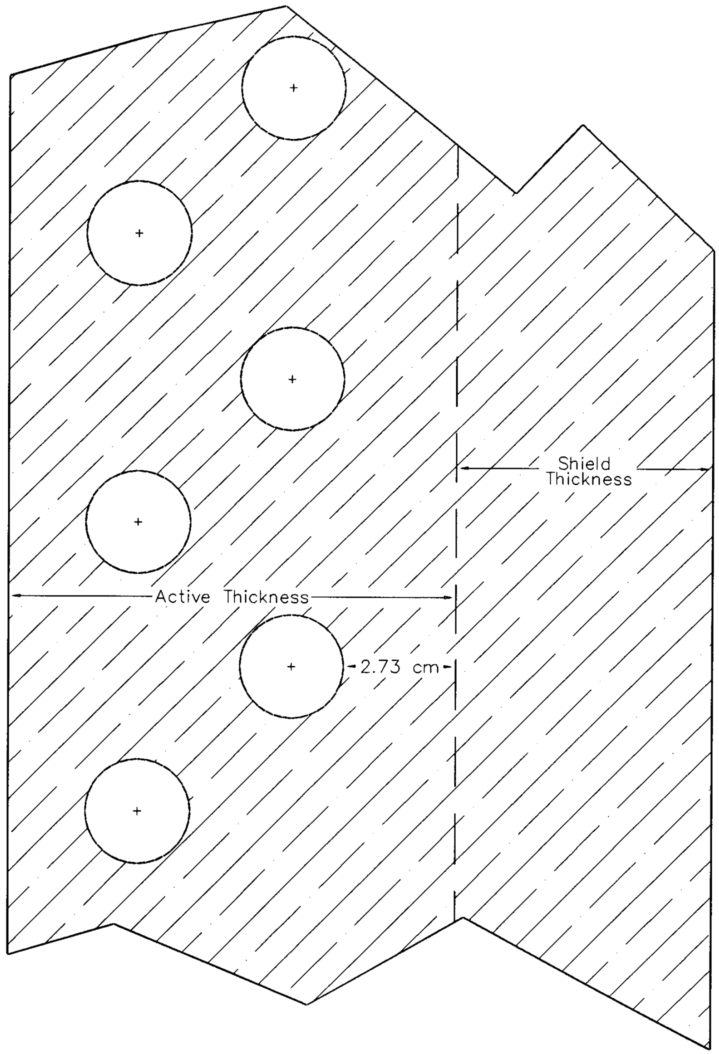


Fig. 2. A two-row tube detector geometry used to calculate the FOM.

The reals coincidence counting efficiency is a function of the number of time correlated neutrons that fall within the coincidence time gate. The fraction in the gate for a 128-μs gate length is defined as

$$f_g = \frac{\sum_0^{128\mu s} T}{\sum_0^{\infty} T} \quad , \tag{6}$$

where

T = the totals counts.

The ε was determined from the unnormalized results of Monte Carlo N-Particle Code (MCNP)[3] calculations. The moderator thickness was determined to be the distance from the inner face of the detector cavity to 1.0 diffusion length (2.73 cm in poly and longer for low-hydrogen plastics) beyond the edge of the back tube row as shown in Fig. 2. The fraction in the gate can be determined from the unnormalized output tallies produced by MCNP.

The FOM equation given above does not include the effect of the moderator thickness on the room source neutrons. Thus, this source of background neutrons must be negligible or there will be additional thickness penalties. The largest values for the FOM give the smallest detectability limit. Thus, the counting efficiency is only one of the parameters that go into the optimization of the detectability limit. A detector design with two rows of ^{3}He tubes or 5-cm-diam tubes is inherently less desirable for the detectability limit (or FOM) optimization because of the added detector thickness caused by the second row of tubes. In effect, the second row of tubes can increase ε by a factor of ~1.5 but it also increases *t* by ~1.7 so the FOM improves only by $1.5/\sqrt{1.7}$, or ~1.15, and we doubled the number of tubes by adding the second row. The optimized design will have a high number of ^{3}He tubes located near the sample cavity to obtain a high efficiency, a small detector volume, and a small neutron die-away time.

SHIELDING AND BACKGROUND REDUCTION

We have investigated several methods to reduce neutron backgrounds for passive counting. The backgrounds have several components including neutrons from

1. external-area radiation sources (for example, waste-drum storage areas),
2. external-area cosmic-ray sources (for example, spallation reactions in the room),
3. spallation reactions between cosmic rays in the sample or the detector body, and
4. radioactive alpha or beta decay in the walls of the detector tubes.

Items 1, 2, and 4 produce only single neutrons, whereas item 3 produces both single and coincidence neutrons.

The detectability limit of a passive system varies approximately as the square root of the background rate (B). Thus, a factor-of-10 decrease in B results in about a factor-of-3 decrease in d. Different methods can be used to reduce the background for the different source terms listed above. These background reduction methods include the following:

1. External shielding such as CH_2 and concrete
   a. 10 cm of CH_2 gives a factor-of-10 reduction for items (1) and (2),
   b. 100 cm of concrete gives a factor-of-4 reduction for item (3), and
   c. the reduction in atmospheric shielding in going from an altitude of 2200 m to 300 m gives a decrease in items (2) and (3) by a factor of ~5.

2. Underground shielding locations
   70 m of dirt gives ~10^3 reduction for items (2) and (3).

3. Removal of high-mass-number elements in the detector and shield
   a. cadmium liners on both sides of the waste-drum detector banks increase the coincidence background by a factor of ~2 because of item (3), and
   b. the exterior cadmium liners on the ^{3}He detector banks decrease items (1) and (2) by only ~15%.

4. Statistical filter for the rejection of cosmic-ray spallations. The statistical outlier test can reduce item (3) by a factor of ~1.8 for coincidence counting and ~1.1 for singles counting in a well-shielded location. Short runs of 10 s each are used to add sensitivity to the statistical test.

5. Choice of the tube wall and an energy window on the thermal-neutron peak of the ^{3}He detector.
   a. After large reductions in the true neutron background in the detector, there is a residual background of counts from the radioactive alpha and beta decay products from the interior wall of the ^{3}He tube. We have measured these background levels to be

      1. 2.8×10^{-4} counts/s • inch for a 1-in.-diam aluminum tube and
      2. 4.3×10^{-5} counts/s • inch for a 1-in.-diam stainless steel tube.

      A 200-L-drum assay system might contain 100 tubes (36 in. long) giving 0.16 counts/s of this type of background (1.0 counts/s for aluminum tubes). For 2-in.-diam tubes, these backgrounds would approximately double.

6. Active cosmic-ray veto counters. Plastic scintillator "paddles" can be placed over the ^{3}He detector system to operate in the anticoincidence mode. Background item (3) can be potentially reduced using this technique; however, for a large drum counter, the deadtime would be excessive.

The combination of all of these background-reduction techniques can reduce the measured background rate by orders of magnitude.

DESIGN OPTIONS

The design options focused on an HDPE moderator and on a composite moderator (H, C, O, and F). For both options, the number of tubes, gas pressure, and tube placement can be varied to increase the FOM at the expense of tube and fabrication costs. In general, there is a performance saturation effect so that the initial cost increments produce more gain than the final cost increments as we approach saturation. Parametric studies of the variables are needed to make the final parameter selections.

Moderator Materials

Two sets of MCNP design calculations were performed, the first for a moderator that was 100% HPDE and the second for a composite moderator containing layers of HPDE and other plastics. These plastics contain less hydrogen and more C, O, and F than does CH_2.

In general, as the neutrons penetrate deeper into the moderator, the role of hydrogen for thermalizing the neutrons is less essential than at the surface. In the moderator, the thermalized neutrons are absorbed by hydrogen before they reach the ^{3}He tubes so it is desirable to replace H with C, F, and O in moderator locations away from the front surface and near the ^{3}He tubes. The primary benefit of this substitution is in the second row of ^{3}He tubes where the neutrons are well moderated. The drawback of this substitution is that the detector active volume gets larger and the average density gets larger. Both of these factors hurt the FOM and can wipe out the efficiency gain.

Modular Design

The WM3100 platform and the HPDE moderator make it possible to add or subtract performance capability with only minor changes in the electro-mechanical design. The number of tubes in the detector bank can be decreased to save tube costs and the shielding thickness can be decreased to save CH_2 costs. The MCNP design study was be used to select the desired FOM and efficiency.

This modular design allows the fabrication of a system with reduced cost and reduced performance with very minor mechanical changes.

Multiplicity Counting

We have assumed that the dominant consideration in the design optimization was the detectability limit. A high efficiency is also needed for multiplicity counting. The multiplicity counting uses the singles, doubles, and triples rates and high efficiency is required to get reasonable counting statistics for the triples counts. The ratios of the triples : doubles : singles varies as $\varepsilon^3 : \varepsilon^2 : \varepsilon$ so changes in the efficiency can be detected by changes in the ratios. The efficiency measured by multiplicity counting has the advantage that it directly tracks the actual efficiency for each neutron escaping the drum. Thus, localized shielding and nonuniform plutonium distributions are not a problem. The primary problem is that low plutonium mass samples have poor counting statistics.

DESIGN RESULTS

HDPE Moderator Design

The design work included the normal HDPE moderator as well as the more complex composite moderators made of HDPE combined with different types of plastic and Teflon. The added efficiency from the composite moderators needs to be balanced against the increased material and fabrication costs. The efficiency can be increased by using a composite moderator or by adding additional ^{3}He tubes. Both of these activities increase the cost and the best approach is an economic decision.

HPDE Design Results

The MCNP calculations were performed for the HDPE case using the as-built JCC-21[4] as the benchmark case. Figure 3 shows the geometry used in the calculations. The JCC-21 has 10 tubes on a side, giving a total of 60 tubes. In the study, we evaluated the tube pitch, depth in CH_2, gas pressure, and number of rows. Spontaneous fission source spectra for ^{252}Cf and ^{240}Pu were modeled. The ^{240}Pu neutron spectrum was used for the final FOM analyses.

The key parameters that were used to estimate the performance include the efficiency, the moderator thickness, and the gate fraction (128-μs gate). The efficiency as a function of gas pressure and tube depth in the HDPE is shown in Fig. 4. We see that the optimum depth is about

35 mm from the inside face of the moderator to the center of the ^{3}He tubes. This distance reduces to ~31 mm for the 10-tube case.

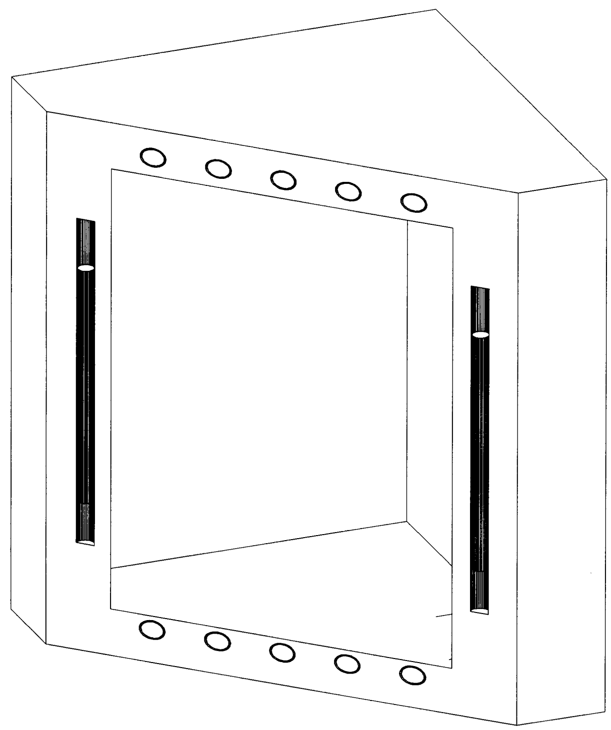


Fig. 3. Diagram of the detector geometry that was used for the MCNP calculations.

Figure 5 shows the data for the FOM; we see that the two-row design gives a low FOM. The selection of the number of tubes is a tradeoff between the desired FOM and the cost. Figure 6 shows the calculated ε vs the number of tubes for different gas pressures. The cases with and without cadmium are shown and a two-row (staggered) configuration is shown. For one row of tubes and no corner leakage, the ε tends to saturate at 37% for 20 tubes on a side. However, if the tubes are staggered to make two rows as shown in Fig. 2, the ε is increased to ~46%. The second row of tubes significantly increases our room background (from shielding displacement) and our cosmic-ray background (from volume increase).

5-cm-Diameter ^{3}He Tubes

For large detector systems of the type described in this report, higher counting efficiencies per detector can be obtained with 5-cm-diameter tubes compared with 2.54-cm tubes. However, the FOM for the larger tubes is worse than for the smaller tubes because the detector body thickness

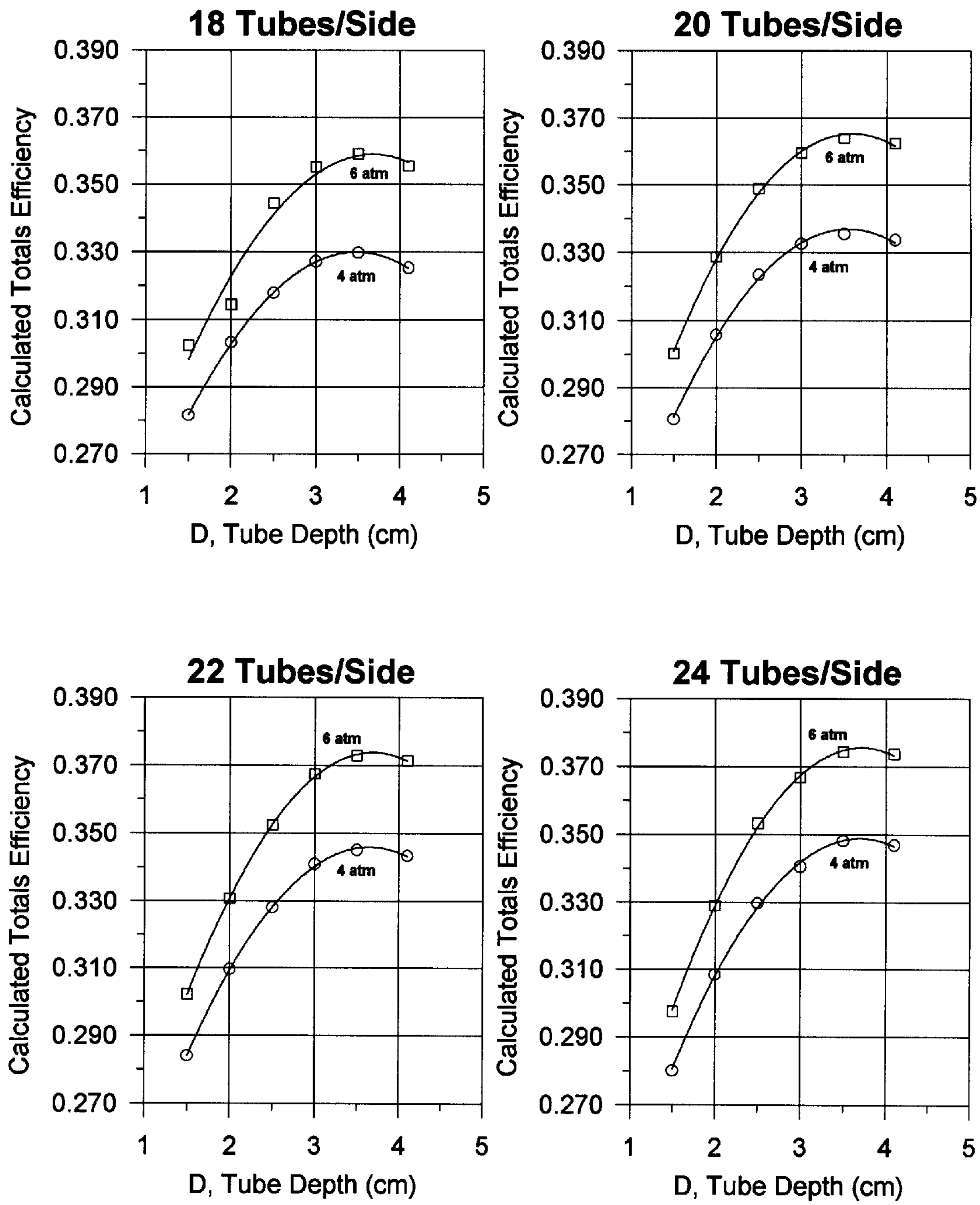


Fig. 4. Calculated efficiency for ^{240}Pu neutrons as a function of ^{3}He tube depth in the CH_2 and as the number of tubes per side.

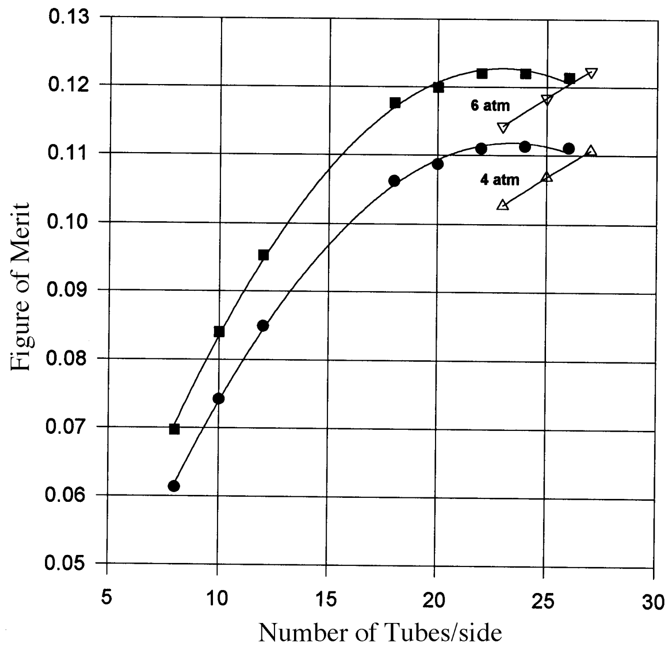


Fig. 5. Calculated FOM as a function of the number of tubes per side (triangle symbols represent a two-row design).

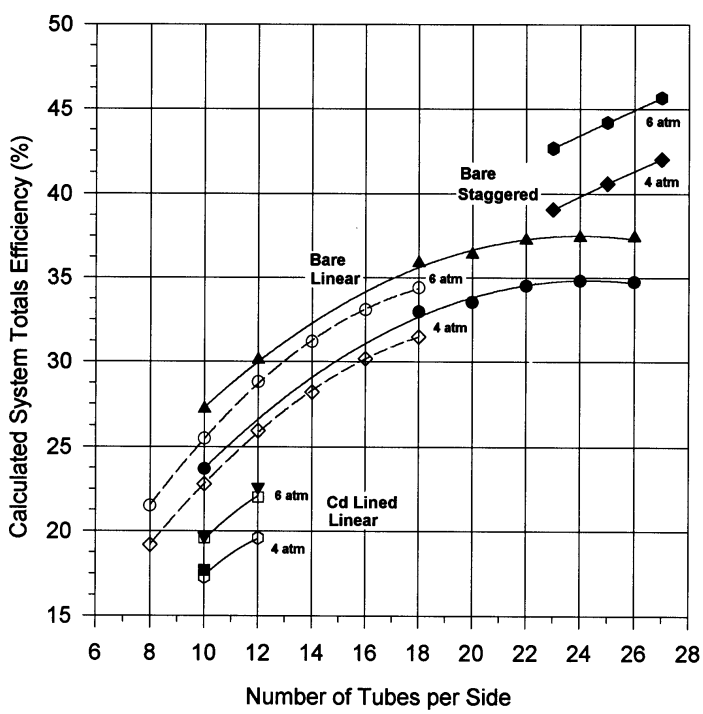


Fig. 6. Calculated efficiency for ^{240}Pu neutrons as a function of the number of tubes per side and the ^{3}He gas pressure. The highest efficiencies are for the staggered (two-row) design.

increases for the larger tubes. The larger-diameter tubes also have the problem that the AMPTEK amplifiers have: time constants that are too short for the 5-cm-diameter tubes. The use of amplifiers with long time constants makes the 5-cm tube system much more sensitive to gamma-ray backgrounds. The smaller-diameter tubes with the AMPTEK amplifiers are roughly an order of magnitude less sensitive to gamma-ray pileup than the 5-cm-diameter tubes.

For high-efficiency systems, the 2.54-cm tubes would use 6 atm pressure and the 5-cm tubes would use 3-atm ^{3}He pressure. The larger tube has an efficiency that is ~1.7 larger than the smaller tube; however, the cost of the larger tube is ~1.6 times larger so the economics are similar for the two options.

Composite Moderator Results

Composite moderators were evaluated using the MCNP calculations. Plastics other than HDPE were chosen to get more scattering atoms such as C, O, N, and F in place of the H in the CH_2 or moderator. The composite moderators gave results with a higher efficiency per ^{3}He tube than the pure HDPE moderator; however, the FOM for the composite materials was no better than the HPDE moderator. A separate paper[5] in this conference will give the detailed composite moderator results.

System Performance

The High-Efficiency Neutron Counter (HENC) was fabricated using design guidance from the MCNP calculations. The HDPE option was used to obtain a good FOM and to use standard fabrication methods. A single row of ^{3}He tubes was used with a pitch of 3.95 cm. The performance specifications are listed in Table III. The design was optimized for a moderated ^{240}Pu neutron spontaneous fission energy spectrum.

The 30-cm-thick HDPE neutron shield on the outside of the HENC reduces the room source neutron rate by 3 orders of magnitude; however, it gives only a small reduction (<10%) in the cosmic-ray coincidence neutrons. To reduce the cosmic-ray background, a large quantity of overhead shielding, such as concrete, is required.

TABLE III. Performance Specification for the HENC	
Parameter	Value
Neutron efficiency for ^{240}Pu	32%
Neutron die-away-time	50 µs
Deadtime (a)	0.50 µs
(b x 10^{-6})	0.161 µs
Multiplicity deadtime	171 µs
correction coefficient c	0.147 µs
correction coefficient d	0.147 µs
Coincidence gate	128 µs
Predelay	3.0 µs
Doubles calibration coefficient	53.8 counts/s•g ^{240}Pu
Multiplication constant (ρ_0)	0.178

The minimum mass detectability limit for the HENC depends on the location of the system and whether totals or reals are used for the assay. At sea level, the detectability limit (3 σ) is ~1.7 mg ^{240}Pu-eff. for doubles assay and 0.48 mg ^{240}Pu-eff. for singles neutron counting. At the high elevation of Los Alamos (2200 m), the cosmic-ray background is 4 to 5 times higher than at sea level and the detectability limit approximately doubles.

The HENC was calibrated using plutonium standards including MOX pellets and small PuO_2 powder samples. Figure 7 shows the linear doubles calibration fit to the standards. The neutron multiplication was negligible for the calibration standards.

Figure 8 shows the triples calibration line for the HENC with a slope of 5.83 counts/s•g ^{240}Pu. This calibration demonstrates that the triples rate can be used down to sub-gram quantities of ^{240}Pu-eff. In general, the triples rate will be used to make matrix corrections.

The HENC was used in the second round of the PDP drum measurement test and the results of this test should be available in early 1997.

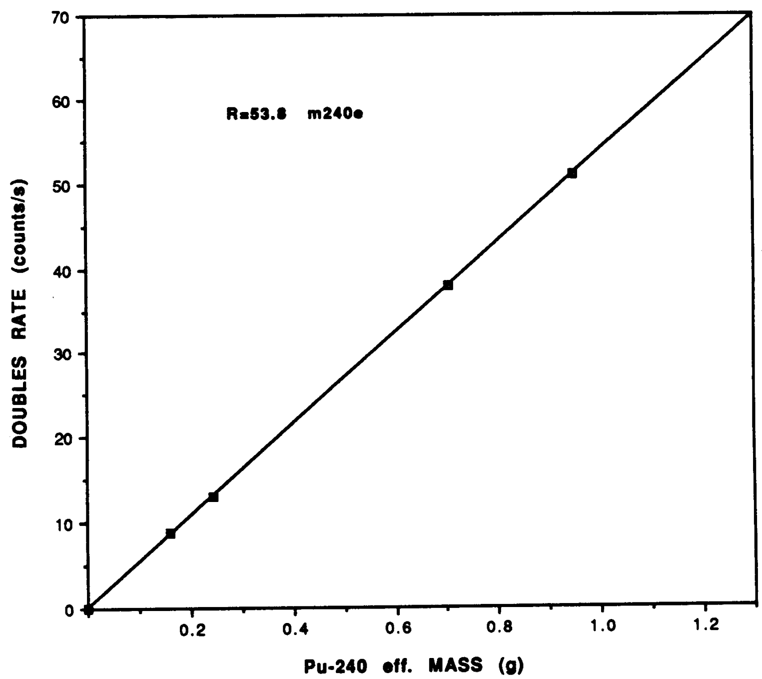
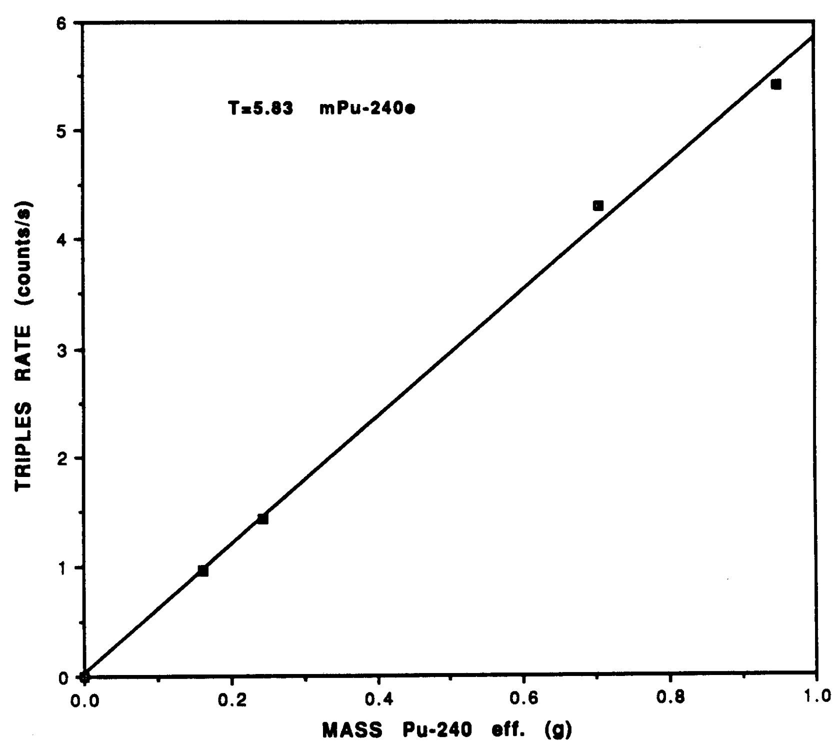


Fig. 7. The linear doubles calibration curve for the HENC.

Fig. 8. The triples rate calibration curve for the HENC.



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