*Passive Neutron Design Study for
200-L Waste Drums*

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CONTENTS

ABSTRACT.....	1
I. INTRODUCTION.....	2
II. DETECTABILITY LIMIT.....	2
Figure of Merit.....	6
III. SHIELDING AND BACKGROUND REDUCTION.....	7
A. Intrinsic ^3He Tube Background.....	9
B. Active Cosmic-Ray Veto Methods	9
C. External Shielding.....	9
D. Cosmic-Ray Shielding.....	11
E. Statistical Filter for Cosmic-Ray Rejection.....	12
IV. DESIGN OPTIONS.....	13
A. Moderator Materials.....	13
B. Modular Design.....	13
C. Amptek Preamplifiers.....	13
D. Multiplicity Counting	14
V. DESIGN RESULTS	14
A. HDPE Moderator Design.....	14
B. HDPE Design Results.	14
C. 5-cm Diameter ^3He Tubes	15
D. Composite Moderator Results	19
VI. SYSTEM PERFORMANCE.....	22
REFERENCES	23

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by

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ABSTRACT

We have developed a passive neutron counter for the measurement of plutonium in 200-L drums of scrap and waste. The counter incorporates high efficiency for the multiplicity counting in addition to the traditional coincidence counting. The ^{252}Cf add-a-source feature is used to provide an accurate assay over a wide range of waste matrix materials. The room background neutron rate is reduced by using 30 cm of external polyethylene shielding and the cosmic-ray background is reduced by statistical filtering techniques. Monte Carlo Code calculations were used to determine the optimum detector design, including the gas pressure, size, number, and placement of the ^3He tubes in the moderator. Various moderators, including polyethylene, plastics, teflon, and graphite, were evaluated to obtain the maximum efficiency and minimum detectable mass of plutonium.

I. INTRODUCTION

The measurement of the plutonium content in waste containers is required prior to long-term storage and disposal. The accuracy of the measurement 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, limiting 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 correction¹ and multiplicity counting² to make corrections for localized shielding in the drums.

The joint development of the advanced passive neutron assay system was accomplished under a cooperative research and development agreement (CRADA) between Canberra Industries and Department of Energy (DOE). For the drum counter to be developed under the CRADA, the design goals that require optimization are listed in Table I. The relative weighting and prioritization of these goals will be 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, such as precision, that are based on counting statistics 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.³

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

II. DETECTABILITY LIMIT

We have used the optimization of the detectability limit to be 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 with the detector volume and density. A detector with three rings or layers of ^3He 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, the large detector volume displaces the external neutron shielding resulting in an increase of the measured background from the 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.

TABLE I. Design Goals for the Waste Drum Counter.

1. Low detectability limit
(good sensitivity at low Pu mass)
2. Ability to meet 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

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

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

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, and we can reduce our background by eliminating the cosmic-ray spallation events with high multiplicity by using a statistical filtering technique.⁴

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

$$d \sim \frac{\sqrt{\epsilon^2}}{\epsilon^2} = \frac{1}{\epsilon}.$$

However, the background coincidence rate 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 ^3He tube area moderator. Thus, if we double our detector volume or density, we will more than 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 the drum loading.⁴ We see that most hydrogenous loadings have a slightly lower background than the empty case.

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

$$A = T^2 G,$$

where

- A = accidental coincidence rate,
- T = totals background rate, and
- G = gate width.

The coincidence background can be written as

$$B = B(\text{cosmic ray}) + B(\text{room accidentals})$$

and

$$B(\text{cosmic ray}) \sim \epsilon^2 \cdot \text{volume} \cdot \text{density}.$$

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 ^3He 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 active 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 .

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

Figure of Merit

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

$$FOM \sim \frac{1}{d} \sim \varepsilon^2 \left[\frac{f_g}{\varepsilon^2 \cdot t \cdot \rho} \right]^{1/2}$$

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

where

ε = totals counting efficiency (%),

ρ = moderator density,
 t = moderator thickness (cm), and
 f_g = the fraction in the gate for a 128- μ s gate length.

The real 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- μ s gate length is defined as

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

where

T = the totals counts.

The ϵ was determined from the unnormalized results of Monte Carlo neutron-proton (MCNP) code⁶ calculations. The moderator thickness was determined to be the distance from the inner face of the detector cavity to the 1.0 diffusion length (2.73 cm in poly and longer for low hydrogen plastics) beyond the edge of the 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 ^3He tubes or 5-cm diameter 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. The optimized design will have a high number of ^3He tubes to obtain a high efficiency, a small detector volume, and a small neutron die-away time. However, the detector tubes must be placed in the moderator so that the detection efficiency is insensitive to minor changes in the matrix loadings.

III. 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 give only single neutrons, whereas item 3 gives 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 factor of ~ 5 decrease in items (2) and (3).
2. Underground shielding locations. 70 m of dirt gives $\sim 10^3$ 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 ^3He detector banks decrease items (1) and (2) by only $\sim 15\%$.
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. For locations with poor cosmic-ray shielding, these reduction factors are significantly less.
5. Choice of the tube wall and an energy window on the thermal-neutron peak of the ^3He detector. 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 ^3He tube. We have measured these background levels to be
 1. 2.8×10^{-4} counts/s $\cdot$ inch for a 1-in. diameter aluminum tube, and
 2. 4.3×10^{-5} counts/s $\cdot$ inch for a 1-in. diameter 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. diameter tubes, these backgrounds would approximately double.
6. Active cosmic-ray veto counters. Plastic scintillator “paddles” can be placed over the ^3He detector system to operate in the anticoincidence mode. Background item (3) potentially can be reduced using this technique; however, for a large drum counter, the dead time would be excessive.

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

A. Intrinsic ^3He Tube Background

The background from radioactive alpha-particle decay in the walls of the ^3He tubes has been measured⁵ to be $\sim 4.3 \times 10^{-5}$ counts/s per in. for a 1-in. diameter stainless steel tube. Our design basis detector might contain ~ 18 tubes per side with 6 sides, and a typical tube is 91 cm long. This results in 3888 in. of tubing yielding ~ 0.117 counts/s. These are random events that only contribute to the accidental coincidence rate. Because the room-totals rate is typically larger than 0.117 counts/s, we have not attempted to suppress this intrinsic background.

However, aluminum tube walls and some stainless steel tubing have radioactive decay rates that are much higher than the above rates. These tubes containing low-level radioactivity must be avoided in detectors used for low-level counting.

B. Active Cosmic-Ray Veto Methods

The cosmic-ray background can be decreased by electronic anticoincidence shields. A thin plastic scintillator can be placed over the ^3He detector to sense a cosmic-ray event and to gate off the electronics for the ^3He detector. Tests on a small inventory sample (INVS)-type⁷ ^3He detector showed that the background could be reduced by a factor of ~ 2 using this approach. However, the ^3He systems must be gated off for a period that is longer than the coincidence gate (128 μs). The resulting dead time for the small counter was $\sim 15\%$ and extrapolating the much larger geometry of the WM3100 gives a dead time that would be approaching 100%. Thus, the electronic anticoincidence method to reject cosmic-rays is not recommended for the large drum counters using ^3He tubes because of their long coincidence gates.

C. External Shielding

The neutron background from external sources in the area can be reduced with exterior CH_2 shielding for room neutrons and overhead concrete for cosmic-rays. The neutron shielding effectiveness was calculated using the MCNP code. To estimate the neutron slowing down spectrum for room source neutrons, we modeled a ^{240}Pu spontaneous fission source distributed on the concrete walls surrounding the detector as shown in Fig. 3. The room walls were 30-cm-thick concrete. The results of the calculations are shown in Fig. 4, where a thickness of 10 cm of high-density polyethylene (HDPE) ($\rho = 0.943 \text{ g/cm}^3$) gives a dose reduction of a factor of 10. If we remove the room-scattered neutrons, the spectrum is harder and it requires 13 cm of HDPE to obtain a factor of 10 reduction in dose.

If we are using totals neutrons for the assay, then the detectability limit improves as the square root of the background reduction. Thus, 10 cm of shielding yields a factor of 10 reduction in the background and a factor ~ 3 reduction in d if the neutrons originate in the room. However, as the cosmic rays become the dominant source of totals neutrons, the external CH_2 shielding provides little or no benefit.

For most applications, we would use the coincidence rate (R) for the assay and the background totals rate impacts R by the accidental coincidence counts. The accidental rate is given by $A = T^2 G$ and it is desirable to use enough CH_2 shielding to keep the accidental coincidence background small compared to the cosmic-ray background. For a well-shielded system, the cosmic-ray background might be ~ 0.2 counts/s (reals). This background is always positive and it can be measured using a long overnight count.

Fig. 3. Room geometry used to simulate scattered background neutrons from the MCNP shielding calculations.

Fig. 4. Normalized room background counting rates from the MCNP calculations for direct and scattered neutrons.

The totals rate that gives us an accidental rate of 0.1 counts/s is

$$T = \sqrt{A/G} = \sqrt{\frac{0.1}{128 \cdot 10^{-6}}}, \text{ or}$$

$$T = 28 \text{ counts/s}.$$

Thus, we need to provide enough shielding to keep the room background rate below 28 counts/s so that the accidental rate does not degrade our detectability limit.

In typical field applications with the WM3100 system (10-cm CH_2 shield), the room background is in the range of 5–200 counts/s depending on the proximity of waste drums and neutron sources. A 30-cm-thick shield would add 20 cm of CH_2 to the existing system and further reduce the room background rate by 2 orders of magnitude. This amount of shielding would make the accidental coincidence background negligible and the counter would be very sensitive in the totals mode of assay. However, the CH_2 shielding provides a negligible reduction in the cosmic-ray neutron background.

D. Cosmic-Ray Shielding

The cosmic-ray origin neutrons are much more difficult to shield because of the penetrability of the high-energy cosmic rays prior to the spallation reactions that produce the neutrons. Figure 5 shows a comparison of the shielding differences between room neutrons and cosmic rays. It requires ~160 cm of concrete to obtain a factor of 10 reduction in the neutron dose for cosmic rays.

Fig. 5. Neutron shielding effectiveness for room-source neutrons and cosmic-ray source neutrons.

In general, high-density shielding should be used for cosmic rays, but it is necessary to keep the shielding far enough above the neutron detector to prevent the shield from being a spallation source feeding coincidence neutrons into the detector. For example, a layer of iron or lead shielding on top of the neutron detector will actually increase the neutron background.

E. Statistical Filter For Cosmic-Ray Rejection

The detectability limit is a function of the neutron time-correlation background from cosmic rays, and we have reduced the background by a factor of ~ 1.8 in well-shielded locations by eliminating the cosmic-ray spallation events that produce high-multiplicity events by using a statistical filter. The cosmic-ray events can be counted as prompt charged-particle reactions in the detector tubes or as spallation-source neutrons that extend in time over the slowing-down time of the detector body. The predelay ($3 \mu\text{s}$) eliminates the first category because they are short lived and the predelay⁸ vetoes them from the coincidence gate. The spallation neutrons fall within the coincidence gate but often with high multiplicity. We use the data collection software to isolate the high-multiplicity events and to eliminate them from the data averages. We are currently using statistical techniques to accomplish this.

Our statistical filter for background reduction consists of a $2.4\text{--}3.0 \sigma$ rejection threshold from the average of multiple, short data intervals. A typical counting time for a drum is 600 s, and we divide this into 60 intervals of 10 s each. If any interval is more than $2.4\text{--}3.0 \sigma$ out of the average, we reject that interval from the average. This type of filter does not interfere with the data

collection for drums with high or low plutonium content. Approximately 1%–10% of the useful data points are eliminated by this statistical filter.

IV. DESIGN OPTIONS

The design options focused on a 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.

A. 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 CH₂.

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 ³He tubes so it is desirable to replace H with C, F, and O in moderator locations away from the front surface and near the ³He tubes. The primary benefit of this substitution is in the second row of ³He 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.

B. 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 CH₂ costs. The MCNP design study was used to select the desired FOM and efficiency. Also, the shielding curve in Fig. 4 can be used to select the required shielding thickness. This modular design allows the fabrication of a system with reduced cost and reduced performance with very minor mechanical changes.

C. Amptek Preamplifiers

The High Efficiency Neutron Counter (HENC) uses 3 AMPTEK preamps on each side to service ~16 tubes (~36 in. long). The input capacitance into the preamp from the multiple tubes is more a function of the number of tubes than the tube length. A single preamp can service 6–9 tubes so an 18-tube bank could be serviced by 2 to 3 AMPTEK preamps.

In many cases, multiple preamps are used to reduce dead time, but for waste counters, the rates are low and dead time is not a problem. From the standpoint of cost and reliability, the number of preamps should be kept to a minimum unless drums with high gamma-ray backgrounds are expected.

D. 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 $\epsilon^3 : \epsilon^2 : \epsilon$, 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.

V. DESIGN RESULTS

A. 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 ^3He tubes. Both of these activities increase the cost and the best approach is an economic decision.

The key design parameter is the FOM and the resulting cost. However, the cost is a more complex issue involving the ^3He tubes, the moderator cost, the fabrication, and shielding costs. A convenient modular design is required to give flexibility in the cost to meet variable competitive conditions.

B. HPDE Design Results

The MCNP calculations were performed for the HDPE case using the as-built JCC-21¹ as the benchmark case. Figure 6 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. 7. We see that the optimum depth is about 35 mm from the inside face of the moderator to the center of the ^3He and this distance reduces to ~31 mm for the 10-tube case.

Figure 8 shows the same type of data for the FOM, and we see that the optimum depth is less than for the maximum efficiency. The selection of the number of tubes is a tradeoff between the desired FOM and the cost. Figure 9 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 6-atm 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%. However, the second row of tubes significantly increases our room background (from shielding displacement) and our cosmic-ray background (from volume

Fig. 6. Diagram of the detector geometry used for the MCNP calculations.

increase). Thus, the FOM gets worse with the two-row design as shown in Fig. 10. We see that the FOM curve saturates at ~20 tubes for the single-row design; however, the FOM continues to improve for the two-row design and if we were willing to pay for a large number of tubes (≥ 28 per side) the two-row design provides a higher FOM. This can be improved using composite moderators.

When tube costs are factored into the decision, we probably would choose an 18-tube (6 atm) configuration to yield an FOM of 0.117 and an efficiency of 36% for ^{240}Pu with full geometry coverage. Our 10-tube benchmark design has an FOM of 0.074 and an efficiency of ~23% for ^{240}Pu . This efficiency was measured to be ~19% for a ^{252}Cf source in the center of the cavity so it is necessary to reduce the MCNP results to correspond to the measurements. Normalizing our MCNP result to the measured JCC-21 efficiency, gives a projected efficiency of 29.8% for 18 tubes (6 atm) for a ^{252}Cf source.

The effect of the gas pressure and the cadmium liner is shown. The efficiency for counting ^{240}Pu is about 3.5% higher than ^{252}Cf because of the lower neutron energy for ^{240}Pu compared with ^{252}Cf . For a two-row design, this increase for ^{240}Pu is reduced.

C. 5-cm Diameter ^3He 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 increases for the larger tubes. The larger diameter tubes also have the problem that the AMPTEK

Fig. 7. Calculated efficiency for ^{240}Pu neutrons as a function of ^3He tube depth in the CH_3 and as the number of tubes per side.

1 inch tubes, Pu-240 spectrum

Fig. 8. FOM vs tube depth for different number of tubes per side.

Fig. 9. Calculated efficiency for ^{240}Pu neutrons as a function of the number of tubes per side and the ^3He gas pressure. The highest efficiencies are for the staggered (two-row) design.

Fig. 10. Calculated FOM as a function of the number of tubes per side (triangle symbols represent a two-row design).

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 ^3He 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 greater so the economics are similar for the two options.

D. 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. Table III lists the commercial plastics that were evaluated. The higher-density materials have the problem that the cosmic-ray background increases and that reduces the FOM.

Figure 11 shows a conceptual diagram of a composite moderator using polypropylene and polystyrene. After the neutrons have been thermalized by the hydrogen on the front CH_2 face of the moderator, the atoms that scatter neutrons without the absorption of hydrogen tend to increase the probability of getting the neutrons into the ^3He tubes.

The composite moderator gives most of its potential benefit for a two-row design because the neutrons that are deeper into the moderator are well thermalized so the longer scattering paths take the neutrons to the back row of ^3He tubes.

The FOM is inversely proportional to the square root of the effective detector thickness t . If the moderator shown in Fig. 11 had as many tubes on the back row as the front row, the effective thickness would be 10.1 cm. However, because the back row has only half as many tubes as the front row, the back row thickness increment was cut in half for the study. The front row alone has an effective thickness 6.55 cm and the addition of the back row increases the effective thickness to 8.32 cm. If the increased diffusion length for the plastics was considered, this effective thickness would increase to over 9 cm.

The relative efficiency vs the number of tubes on a side are shown in Fig. 12 for a two-row design. We see that we get more efficiency per tube than for the single-row HPDE design. Part of the gain is from the double row of tubes and part is from the composite moderator. The FOM is reduced because of the larger t and ρ as well as the smaller fraction in the gate fg ; however, the higher efficiency from the composite moderator compensates these losses.

The comparison of the different plastic materials is shown in Fig. 13. Polypropylene and polystyrene were selected because of their low densities and reasonable costs.

The composite moderator study showed several interesting improvements compared to an HDPE moderator. The composite provides a higher efficiency per tube than the HPDE moderator and a two-row design is needed to obtain most of the benefit. The back row of tubes gave a higher efficiency if a small air gap (~ 3 mm wide) was placed around each tube. The HDPE layers were always required on the front and back of the composite plastics.

TABLE III. Specifications for Plastics Evaluated for Composite Moderator

Material Type	Trade Name	Manufacturer	Density	Hydrogen %	Carbon %	Nitrogen %	Oxygen %	Fluorine %	Other %	H Density
Polyethylene	Ultra ethylux	Westlake	0.96	0.667	0.333	0	0	0		0.137
Polypropylene	Propylux	Westlake	0.988	0.625	0.375	0	0	0		0.120
Nylon	Zytel 42/Nylon 6	Hyde	1.14	0.578	0.315	0.0526	0.0526	0		0.111
Composite Nylon/Kevlar	Hydlar-Z	Hyde	1.16	0.5584	0.3354	0.0531	0.0531	0		0.105
Acetal Homopolymer	Delrin	Hyde	1.42	0.5	0.25	0	0.25	0		0.095
Acetal Copolymer	Ultra-Form Acetal	Westlake	1.4	0.5	0.25	0	0.25	0		0.093
Polystyrene	HIPS	Westlake	1.05	0.515	0.485	0	0	0		0.085
Phenolic Paper	Phenolic	Westinghouse	1.45	0.452	0.354	0	0.192	0		0.084
Polystyrene	Styraclear	Westlake	1.05	0.5	0.5	0	0	0		0.081
Terephthalate	4101	Hyde	1.31	0.428	0.428	0	0.142	0		0.071
Polycarbonate	ZELUX/LEXAN	Westlake	1.2	0.424	0.484	0	0.0909	0		0.066
PVDF Fluoropolymer	Kynar/Homo 740	Westlake	1.78	0.333	0.333	0	0	0.33		0.056
PVDF Fluoropolymer Composite	Kynar/Copolymer 2800	Westlake	1.78	0.307	0.333	0	0	0.36		0.056
Polyetherimide	ULTEM-1000	Westlake	1.27	0.350	0.534	0.0287	0.086	0		0.052
Polysulfone	Thermalux/UDEL	Westlake	1.24	0.3333	0.5	0	0.125	0	0.04 sulfur	0.050
Fluorinated ethylenepropylene	Teflon/FEP	E. I. DuPont	2.16	0	0.357	0	0	0.642		0

Fig. 11. Moderator geometry used for the composite moderator MCNP design study.

Fig. 12. Normalized efficiency vs number of tubes for the composite moderator with the two-row design.

Fig. 13. Relative efficiency and FOM for the different composite moderators with 15 tubes per side.

VI. 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 ^3He tubes was used with a pitch of 3.95 cm. The design was optimized for a moderated ^{240}Pu neutron spontaneous-fission energy spectrum.

The post-fabrication testing of the HENC showed that the optimization study should have included the matrix in the waste drum as well as the detector moderator. A key design goal is to make the detector relatively insensitive to low-density hydrogen changes in the matrix. This can be accomplished by making the tube moderator thickness slightly under-moderated so that the neutron absorption in the drum is effectively compensated by the increase in efficiency for the scattered neutrons that escape from the drum.

This final optimization for the drum matrix was done experimentally on the as-built HENC and it is documented in the follow-up report "HENC Performance Evaluation and Plutonium Calibration."

REFERENCES

1. H. O. Menlove, J. Baca, W. Harker, K. E. Kroncke, et al., "WDAS Operation Manual Including the Add-A-Source Function," Los Alamos National Laboratory report LA-12292-M (April 1992).
2. M. S. Krick and J. E. Swansen, "Neutron Multiplicity and Multiplication Measurements," *Nucl. Instrum. Methods* **219**, 384-393 (1984).
3. N. Ensslin, M. S. Krick, and H. O. Menlove, "Expected Precision of Neutron Multiplicity Measurements of Waste Drums," *Nucl. Mater Manage.* **XXIV** (Proc. Issue) 1117-1124 (1995).
4. D. H. Beddingfield and H. O. Menlove, "Statistical Data Filtration in Neutron Coincidence Counting," Los Alamos National Laboratory report LA-12451-MS (November 1992).
5. H. O. Menlove and G. W. Eccleston, "High Sensitivity Measurements for Low-Level TRU Wastes Using Advanced Passive Neutron Techniques," presented at the Transuranic Waste Characterization Conference, Idaho State University, Pocatello, ID, August 10-13, 1992, Los Alamos National Laboratory document LA-UR-92-2563.
6. J. F. Briesmeister (ed.), "MCNP - A General Monte Carlo Code for Neutron and Photon Transport," Los Alamos National Laboratory report LA-7396-M, Rev. 2 (1991).
7. H. O. Menlove, O. R. Holbrooks, and A. Ramalho, "Inventory Sample Coincidence Counter Manual," Los Alamos National Laboratory report LA-9544-M (ISPO-181) (November 1982).
8. J. E. Swansen, P. R. Collinsworth, and M. S. Krick, "Shift-Register Coincidence Electronics System for Thermal Neutron Counters," Los Alamos Scientific Laboratory report LA-8319-MS (April 1980). Also in *Nucl. Instrum. Methods* **176**(3) 555-565 (1980).