Performance Characteristics of a High Efficiency Passive Neutron Assay System using Alternative Neutron Detectors to Helium-3

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***Abstract*– Passive neutron non-destructive assay systems demand high sensitivity in order to be capable of detecting milligram levels of plutonium for safeguards and waste characterization applications. Chamber efficiencies greater than 30% are required for neutron coincidence and multiplicity counting systems. Existing systems are based on ^{3}He proportional counters and require hundreds of liters of this gas. The severe ^{3}He shortage has created a driver to evaluate alternative detectors in this application. An alternate design must meet the following performance requirements: 1) high absolute detection efficiency, 2) low gamma ray sensitivity and 3) short die-away times. In addition, practical aspects of the detectors must be considered including: 1) system lifetime costs, 2) backward compatibility with existing protocols, 3) long term stability in industrial environments, 4) low maintenance, 5) technology maturity, 6) production scalability and 7) materials toxicity. Several alternative sensors are currently available that could potentially meet the above requirements. These include Boron trifluoride (BF_3) filled detectors, ^{10}B lined proportional counters (including novel designs that increase surface area) and various scintillators based on ^{6}Li. For each category the most promising candidate has been identified and evaluated for use as a direct replacement for ^{3}He in a typical High Efficiency Passive Neutron Assay System designed for transuranic / low-level waste sentencing with a lower limit of detection of 3700 Bq/g (alpha activity concentration). Performance of the alternative designs has been modeled and compared to the ^{3}He baseline with any potential technical improvements also being considered.**

I. INTRODUCTION

Passive neutron Non-Destructive Assay (NDA) systems demand high sensitivity in order to achieve low (milligram level) detection limits for plutonium measurement. Closed chambers with efficiency greater than 30% are typically required for Passive Neutron Coincidence Counting (PNCC) and multiplicity-based techniques.

Existing systems based on ^{3}He require hundreds of liters of gas. The ^{3}He shortage has created a driver to evaluate alternative detectors in this application. There is competing ^{3}He supply demand from the oil and gas industry for well logging, medical imaging applications, low-temperature physics, portal monitoring, health physics, and research projects in nuclear and condensed matter physics.

Table I illustrates the volume of ^{3}He required to furnish a typical range of passive neutron assay system units. Building future systems with exclusive reliance on ^{3}He would put significant strain on this limited resource. Development of alternative neutron detectors would not only ease the shortage of supply issue, but could also provide the potential to improve the technical capabilities in this field.

TABLE I. ^{3}HE GAS VOLUME REQUIRED FOR PASSIVE NDA SYSTEMS

System	^{3}He Volume Required (liters at 1 atmosphere)
Slab Counter	50 - 100
Canister Assay	75 - 150
Drum Assay	300 - 600
Crate Assay	1000 - 2000

II. TECHNICAL REQUIREMENTS

PNCC assay systems are normally configured into a chamber design with arrays of detectors on all sides (roof, walls and floor) effectively producing a 4π geometry. There are three basic requirements for neutron detectors relevant to such systems, primarily relating to detection and measurement of spontaneous fission neutron emitting nuclides such as ^{240}Pu: 1) high absolute detection efficiency, 2) low gamma ray sensitivity, 3) short chamber die-away times.

In addition, practical aspects of the detectors must be considered including: 1) system lifetime costs, 2) backward compatibility with existing protocols, 3) long term stability in industrial environments, 4) low maintenance, 5) technology maturity, 6) production scalability and 7) materials toxicity.

In order to evaluate the performance of alternatives for use in typical high efficiency passive neutron assay system (for the measurement of standard 200 L drums of waste) the following typical PNCC measurement scenario has been considered. International standards required waste sorting at a threshold of < 3700 Bq/g (alpha activity concentration). This is normally the threshold between classification of Intermediate Level Waste (ILW) and Low Level Waste (LLW) that dictates the path of ultimate disposal between shallow burial and geological disposal. This low level of detection translates to milligram levels of plutonium in a 200 L drum.

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Challenges that system designers face include high neutron background rates, the presence of (alpha,n) emissions that interfere with the spontaneous fission signal and high gamma backgrounds (for example from ^{239}Pu, ^{241}Am and co-mingled fission products such as ^{137}Cs).

III. DETECTOR ALTERNATIVES

There are three primary neutron reactions that are exploited in neutron detectors. Examining the thermal neutron cross sections given in Table II it is clear why ^{3}He is the preferred detector for PNCC applications where high closed chamber efficiency is required.

Table II. Neutron Detector Reactions

Reaction	Thermal neutron cross-section (barns)
^{3}He (n,p)	5330
^{6}Li (n,α)	940
^{10}B (n,α)	3840

In all cases moderating materials are required to slow down the fast neutrons emitted by plutonium as the neutron absorption cross has an inverse-velocity relationship over the energy range of interest.

^{3}He detectors also offer good discrimination of the gamma signal as well as inflammability, non-toxicity, physical robustness and long operational lifetime.

A. Boron trifluoride (BF_3)

BF_3 filled proportional counters are similar to ^{3}He but with lower efficiency. These are direct replacements and were commonly found in NDA applications prior to the 1990s. They provide good neutron/gamma separation and high count rate capabilities. The problem, however, is the hazardous nature of the gas which has led to limited usage of this technology. It should also be noted that the lower cross section of ^{10}B and the safety-related limitation on high pressure tubes results in a reduced efficiency (by approximately a factor of 2-5) compared to ^{3}He detectors.

B. ^{10}B lined detectors

^{10}B lined proportional counters are also a direct physical replacement for ^{3}He tubes. Geometrically they are identical to ^{3}He detectors, so they can simply replace the existing tubes with minimal re-design. This detector is therefore a very practical short-term replacement in terms of market availability and like-for-like electronics compatibility. However the limiting factor in PNCC applications is that they are a factor of approximately seven times less efficient than ^{3}He and so must be used in far greater numbers to achieve equivalent performance. Increasing the packing density of detectors produces diminishing returns as moderator materials are sacrificed to make room for the detectors. This fundamentally limits the efficiency of a ‘4π’ chamber to approximately 5 - 10% with ^{10}B lined detectors (and with a significantly increased expense as large numbers of detectors are required). Therefore ^{10}B lined detectors are better suited to smaller instruments with lower efficiency requirements. With excellent gamma discrimination these detectors could be ideal alternatives to ^{3}He in applications with simultaneous high neutron and gamma flux.

C. ^{10}B high surface area detectors

The efficiency of the traditional ^{10}B-lined tubes is limited by the tube’s inner surface area. Therefore configurations with increased surface area can yield increased efficiency. This can be achieved with use of either multiple ‘tubelets’ [1] or interior ‘baffles’. Another approach is to pack multiple coated “straw” cathodes [2] into a larger outer tube. The disadvantage of this arrangement is that charged particles are lost through absorption in the boron coating and inner layers are ‘self-shielded’ from the outer layers. The increased complexity in design also drives up the price and creates large scale manufacturing concerns.

D. ^{10}B doped plastic scintillators

Plastic scintillator material may be embedded with particles of boron-containing compounds. This results in a detector with high efficiency, which is economical to mass produce and has a very fast pulse decay time (to provide higher counting rate capability). High gamma sensitivity is the major issue that needs addressing for this design. Although generally limited for use in detecting fast neutrons, in some cases ^{10}B plastic scintillators are suitable for detection of thermal neutrons and their fast response time provides potential advantages in pulsed neutron active neutron applications.

E. Glass fibers

^{6}Li-loaded glass fibers have the advantage of comparable sensitivity to ^{3}He and a short decay time making them suitable for high count rate applications. However they suffer from a relatively poor neutron/gamma separation capability. The technology can be mass-produced but the costs for a large area detector system could be high. Non-scintillating fibers can be coated with a scintillator material, such as ZnS, and a coating of ^{10}B or ^{6}Li for neutron capture. This provides good neutron sensitivity and low gamma sensitivity (the scintillator is thin). This detector has not been manufactured in large area applications and its current cost is relatively high.

F. Cesium-lithium-yttrium-chloride (CLYC)

The elpasolite, Cs_2LiYCl_6(Ce) offers the advantage that neutron and gamma rays produce light with different time profiles offering the potential for electronic pulse shape discrimination [3]. Combined with their high light yield and high gamma energy equivalent of the neutron peak (>3MeV) this technology is very promising for PNCC applications. For thermal neutrons, CLYC has over two times the cross-section of ^{3}He for samples with enriched ^{6}Li. Furthermore the scintillator achieves good gamma-ray energy resolution (3.9% FWHM at 662 keV) offering the intriguing possibility of combined medium resolution gamma spectroscopy and neutron detection in the same detector package.

G. *^{6}LiF/ZnS screen with wavelength-shifting light-guide*

An excellent thermal neutron detector can be constructed from a ^{6}LiF/ZnS(Ag) screen with a bulk, wavelength-shifting light-guide with a layer of moderator in front. The ^{6}Li plus ZnS(Ag) coating serves as neutron absorber and phosphor. Thermal neutrons interact via the ^{6}Li (n,α) ^{3}H reaction, and the resultant charged particles produce light in the zinc sulfide. This detector offers a very high neutron detection efficiency, can be easily manufactured in large scale and can be optimized for excellent gamma rejection using a digital discrimination method. In a recent study performed by PNNL [4], the detector was identified as one of the leading candidate sensors that is currently of sufficient technological maturity to provide a direct replacement for ^{3}He in the near term for security applications. The PNNL unit contained four ^{6}LiF/ZnS paddles (a 1000 cm^{2} detector) which produced a detection efficiency that exceeded the ^{3}He standard by more than 60%. Simultaneous gamma and neutron acquisition is also feasible, although the gamma data would only provide limited spectroscopic information.

One concern for scintillator based detectors is backward compatibility of existing electronics designs with the output of the photomultiplier tube. However this can be readily solved by adding conversion electronics that process the signals to provide the neutron count rate in whatever format is required. Electronics can also be adapted to provide a gamma ray count rate as a separate data channel.

IV. PERFORMANCE METRICS

Absolute efficiency is the most important performance metric for passive neutron counting. A typical system requires an efficiency of 30 - 55% (comprising six ‘walls’ surrounding the item of interest). In order to investigate suitability of the various alternatives, a set of performance metrics have been defined.

The efficiency of the chamber shall be at least 30% measured using a ^{252}Cf source in the center of an empty chamber. Furthermore, the efficiency profile must be flat across the detector’s surface (no more than +/- 20% variation in efficiency).

In order to achieve low detection limit less than 0.01 g Pu typical of a waste assay application, the slab must achieve very low counts in a typical background field. Differences in background sensitivity compared to baseline detection technology (^{3}He) require evaluation for candidate detectors (and their associated electronics) together with the various means for elimination of background effects (e.g. addition of polyethylene shielding). One effect of particular concern is the sensitivity to cosmic ray spallation effects, which can be a limiting factor in the detection limit, particularly at high elevation.

The detector and its electronics must be capable of supporting coincidence and multiplicity counting modes i.e. with appropriate timing characteristics (e.g. pulse shape and die-away time).

The system must be capable of measurement of a neutron source emitting up to 500,000 n/s (equivalent to 500 grams of commercial grade PuO_2) and must also provide accurate measurement of alpha contaminated waste in the co-presence of the following gamma radiation fields: (i) unshielded gamma-rays produced from 500 grams of commercial grade PuO_2 (including associated gamma emissions from ^{241}Am) placed at the chamber center, (ii) a dose rate field of 2 mSv/hr produced from ^{137}Cs.

In a 0.1 mSv/hr field, the full-scale intrinsic gamma ray efficiency (gamma ray rejection) must be less than 10^{-6}. In the case of scintillator based detectors, this can often be a challenge requiring complex signal differentiation methods. By contrast, ^{3}He detectors can readily obtain gamma ray rejection of 10^{-8} or less [5].

Shielding may be used to reduce the impact of the gamma radiation, assuming that the neutron performance requirements are still achievable.

Other concerns include that the detector must be suitable for use with matrix correction techniques (e.g. add-a-source [6] & ring ratio method). Practical considerations must be taken into account including large scale production, safety, durability, transportation, testing, environmental stability (in particular, variation in efficiency due to temperature is a concern for field applications) and susceptibility to electromagnetic interference.

V. MONTE CARLO SIMULATION

A Monte Carlo model (using MCNP5) was developed in order to test the performance aspects of alternative neutron detectors used in a typical PNCC application. Fig. 1 illustrates the layout of the model. This chamber uses six walls of neutron counters. Each wall comprises two rows of detectors. For clarity, Fig. 1 shows the chamber with the top and bottom detectors removed so that the 200 L drum at the center of the chamber may be seen. Thick polyethylene shielding was included around the system in order to simulate plant conditions i.e. for background reduction.

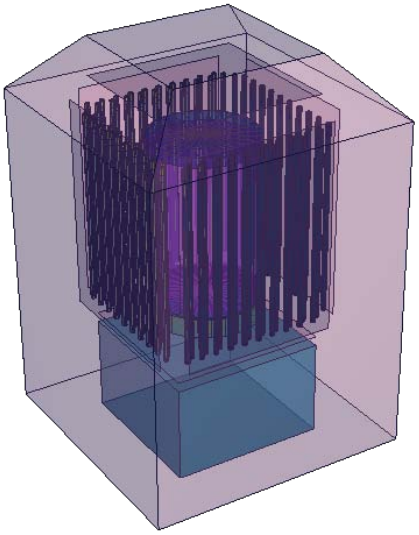

Fig. 1. Monte Carlo model of PNCC system.

Two types of detector were modeled in this initial stage of the work. Firstly in order to define a baseline, ^{3}He detectors were modeled at various fill pressures (2 - 8 atmosphere). Secondly, BF_3 detectors were simulated (with 1 atmosphere fill pressure) and 96% enriched ^{10}B. In both cases the packing density of detectors was varied using both 2.54 cm and 5.08 cm diameter stainless steel tubes. The detectors were arranged using a standard 'repeated structure' module in MCNP with five such modules used in each wall.

For each configuration, an initial phase of optimization was performed (using a neutron source centered in an empty chamber) in order to determine ideal location of the rows of tubes within the moderator.

For the performance study, a uniform distribution of source material inside the drum was used and the source was modeled with the Watt Fission energy spectrum. The matrix contents of the drum were varied to simulate the effect of debris waste (with a wide range of density and composition) as well as soil and concrete. The types of matrix that were analyzed are provided in Table III.

Table III. MCNP Matrix Materials

Matrix	Bulk Density (g/cm^3)	Hydrogen Density (g/cm^3)
Air	0.0	0.0000
Soil	1.3	0.0008
Metals	0.8	0.0070
Combustibles	0.1	0.0076
Concrete	2.2	0.0122
Combustibles	0.2	0.0153
Combustibles	0.4	0.0305
Mix	0.8	0.0344
Combustibles	0.8	0.0610

VI. RESULTS

In order to compare performance for PNCC applications, a Figure of Merit (1) (FOM) has been developed based upon the product of total neutron counting efficiency, ε_T , and efficiency to neutrons detected within 128 μs of source origination, ε_{128}.

$$FOM = \varepsilon_T \varepsilon_{128} \qquad (1)$$

This FOM takes into account the fact that the standard coincidence gate length is set to a width of 128 microseconds. In order to contribute to the "Reals-plus-Accidentals" signal, one neutron must be detected and then a second neutron from the same spontaneous fission must be detected within the gate length.

Another useful parameter to consider is the die-away time, τ, for the chamber (2). In the MCNP model, this was determined from the efficiency at 64 and 128 microseconds after particle generation.

$$\tau = \frac{-64}{\ln\left(\frac{\varepsilon_{128}}{\varepsilon_{64}} - 1\right)} \qquad (2)$$

The die-away characteristics for two different detector types are illustrated in Fig. 2.

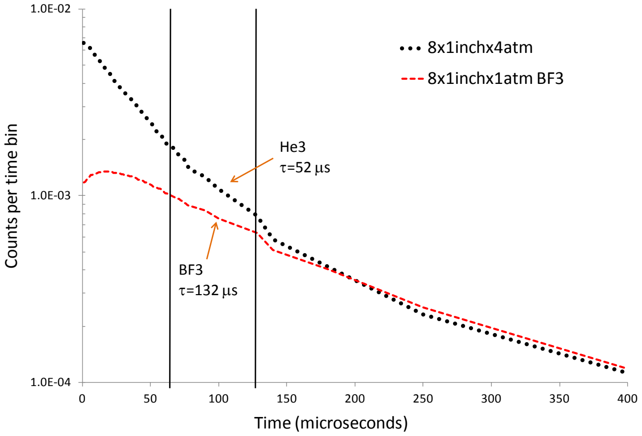

Fig. 2. Chamber Die-Away.

The results of the MCNP simulation are summarized in Table IV for a source at the center of an empty chamber. In addition to comparing the relative performance of the two detector types, these results also allow the optimum usage to be determined when normalizing to relative gas volume within each category.

Table IV. MCNP Predicted Efficiencies and Die-Away Times (Empty Chamber)

Gas	Units per module	Diam (cm.)	Pressure (bar)	Totals Efficiency	Die-Away Time (μs)	FOM
^{3}He	4	2.5	8	33.6%	52.4	0.81
^{3}He	6	2.5	8	39.0%	40.9	1.18
^{3}He	6	2.5	4	37.3%	61.0	1.01
^{3}He	4	2.5	4	29.5%	65.8	0.59
^{3}He	3	5.1	2	34.1%	79.9	0.81
^{3}He (ref)	**8**	**2.5**	**4**	**40.9%**	**52.5**	**1.27**
BF_3	3	5.1	1	24.6%	182.9	0.34
BF_3 (ref)	**8**	**2.5**	**1**	**22.5%**	**132.0**	**0.28**

The effect of addition of matrix to the 200 L drum is illustrated in Fig. 3. FOM is plotted for the two reference cases that are highlighted in Table IV.

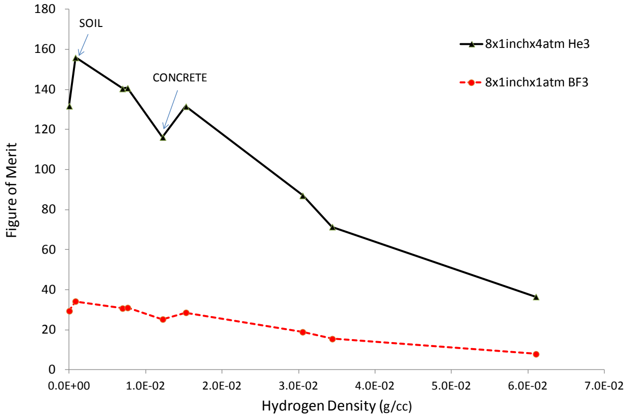

Fig. 3. Figure of Merit as a function of Hydrogen Density.

VII. CONCLUSIONS

Alternative detectors have been studied for use in a PNCC chamber. Important parameters such efficiency, die-away and gamma-rejection must be considered when selecting an appropriate alternative to ^{3}He.

An MCNP model was developed based upon a generic 200 L drum counter. Various performance parameters were evaluated with for ^{3}He and BF_3 proportional counters. The optimum size, pressure and arrangement have been determined in each case by reference to a standard "Figure of Merit".

The reference case for ^{3}He was calculated to be a modular arrangement with 8 detectors per module in a double row arrangement using 2.54 cm diameter tubes with 4 atmosphere fill pressure. It was found that the best possible BF_3 arrangement could only achieve 26% Figure of Merit of the ^{3}He reference case.

Usage of BF_3 appears to be fundamentally limited by its long die-away and poor efficiency characteristics resulting from the relatively low thermal neutron absorption cross-section and fill pressure limitation (the latter of which arises due to safety concerns). The next phase of the study will address other alternative designs such as the next generation of advanced ^{10}B lined / high surface area tubes, ^{6}LiF/ZnS(Ag) screens and CLYC scintillators.

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