ORNL/TM-2015/310

^{3}He Alternatives Summary Report

Robert D. McElroy, Jr.

November 2015

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Nuclear Security and Isotope Technology Division

^{3}HE ALTERNATIVES SUMMARY REPORT

Robert D. McElroy, Jr.

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ABBREVIATED TERMS

AWCC	active well coincidence counter
COTS	commercial off-the-shelf
ENMC	Epi-Thermal Neutron Multiplicity Counter
Euratom	European Atomic Energy Community
FAAS	Fuel Assembly Assay System
FCAS	Fast Carton Assay System
FOM	figure of merit
FPAS	Fuel Pin Assay System
HDPE	High density polyethylene
HLNCC	High Level Neutron Coincidence Counter
IAEA	International Atomic Energy Agency
JRC	Joint Research Centre
LEU	low-enriched uranium
MCNPX	Monte Carlo N-Particle eXtended software package
MOX	mixed oxide
MTBF	mean time between failures
NDA	nondestructive assay
ORNL	Oak Ridge National Laboratory
PEL	Permissible Exposure Limit
PMT	photomultiplier tube
PSD	pulse shape discrimination
PCAS	Plutonium Container Assay System
PNCL	Passive Neutron Coincidence Collar
PSMC	Plutonium Scrap Multiplicity Counter
PTI	Proportional Technologies, Inc.
TLV	Threshold Limit Value
UNCL	Uranium Neutron Collar
WCAS	Waste Crate Assay System
WDAS	Waste Drum Assay System

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1. INTRODUCTION

The ongoing renewal of the nuclear fuel cycle infrastructure and the consolidation of facilities in weapons programs have resulted in the construction and the planned construction of numerous new nuclear material processing facilities, each requiring implementation of a comprehensive safeguards program. These high profile safeguards programs and frontline safeguards instruments have relied on ^{3}He-based neutron coincidence detection mechanisms that are no longer readily available. Without a viable alternative neutron detection technique tailored to the unique needs of the safeguards environment, the viability of these safeguards projects becomes problematic.

The current worldwide shortage of ^{3}He has caused a resurgence in research and development programs dedicated to alternate neutron detection technologies. Several of these programs have produced neutron detection systems targeted to specific application areas such as Homeland Security portal monitoring. Unfortunately, the neutron detection performance requirements for these more commercially lucrative applications are generally less challenging and less diverse than the requirements encountered in international safeguards. A replacement technology specific to the needs of the various safeguards applications is required.

Most of the ^{3}He consumed by neutron-based safeguards assay systems is used in neutron coincidence and neutron multiplicity counting systems. Detection methods for the limited number of total neutron counting applications are not considered in this report, but it is believed that the boron-lined tubes [1, 2] already developed for portal monitoring applications will serve as a suitable replacement for those measurements. In this report, we discuss the potential replacement technologies that are most suitable for use in safeguards applications.

2. CLASSES OF SAFEGUARDS NEUTRON SYSTEMS

Safeguards neutron detection systems can be divided into three broad functional categories, each representing approximately one-third of the safeguards usage of ^{3}He. The general categories are attended assay, unattended/remote monitored assay, and waste assay. The attended monitoring systems can be subdivided by performance requirement into standard coincidence and multiplicity counting systems, while the unattended systems can be divided by the physical arrangement of the measurement system. These general categories are characterized by plutonium mass range, analysis method, and physical form factor. However, each of these broad categories encompasses diverse material forms, interferences, container volumes, and operational constraints that have required many variants of the basic neutron detection system to provide optimal configurations for the specific measurement conditions. Table 1 provides a summary of the typical basic parameter ranges accommodated by the existing ^{3}He-based detection systems. The table illustrates the broad range of materials and configurations encountered. There are exceptions to almost all the parameter ranges listed in the table that exceed the range (e.g., systems to accommodate gamma-ray exposures of up to 10 Sv/h have been fielded).

One should also consider that the systems fielded to date have been bounded by the properties of the ^{3}He proportional tube and supporting electronics. That is, the target performance values and operational ranges have been defined by the ^{3}He detector, and perhaps the performance targets would be different if ^{3}He had not been the detector of choice for the last 30 years.

Table 1. Typical neutron detection parameter ranges for passive neutron coincidence-based plutonium assay by basic application area

	Waste	Attended		Unattended	
		Coincidence	Multiplicity	Well Counters	Slab Counters
Pu mass	0–200 g	0–3 kg	0–3 kg	up to 10 kg	up to 20 kg
Pu-240 effective	5–40%	5–40%	5–40%	5–40%	5–40%
^{241}Am relative content	0.1–4%	0.1–4%	0.1–4%	0.1–4%	0.1–4%
Alpha value	0.7 to >100	0.7–1.1	0.7–10	0.7–1.1	0.7–10
Container sizes (liters)	2–12,000	0.005–200	1–200	2–40	Small cans to fuel assemblies
Container net weight (kg)	0.01 to >1000	<200	<200	<200	1 to >1000
Neutron emission rate	0 to 1E6 n/s	0 to 2E6 n/s	0 to 1E7 n/s	0 to 1E7 n/s	0 to 1E7 n/s
Typical max gamma-ray exposure rate	10 mSv/h	5 mSv/h	5 mSv/h	100 mSv/h	100 mSv/h
^{3}He per system (liters)	10–3000	10–500	10–1500	20–100	20–300
Typical ^{3}He per system (liters)	100–450	10–50	10–300	20–50	20–250
Target performance values [3]					
Random error	10%	1–5%	1–5%	1–5%	10%
Systematic error	5%	0.5–3%	0.5–3%	0.5–3%	5–10%
Assay times (minutes)	5–60	5–10	5–10	1–10	1–10

2.1 HISTORIC AND FUTURE ^{3}HE USAGE IN SAFEGUARDS-RELATED APPLICATIONS

To determine future needs for ^{3}He detectors or a suitable replacement technology for safeguards applications, it is informative to examine both the current and past consumption of ^{3}He detectors. However, this analysis is complicated by a number of factors, chief among them are market visibility, market definition, and the changing market.

Market visibility. Market visibility is the knowledge of the customer base, their procurement plans, and the state of the competition. A lack of market visibility haunts both vendors and national laboratories alike. In the case of ^{3}He proportional detectors and systems, no one entity possesses a complete knowledge of what detectors or systems have been fabricated and installed for safeguards-related applications. During the period from the 1980s through the early part of the new century, the supply of ^{3}He was relatively unrestricted, and the gas cost far less than it does today. While there were only two significant sources (US and Russian government programs), there were many ^{3}He proportional tube manufacturers. These detector vendors did not always know the ultimate use of their products, nor were they necessarily aware of the extent of their competitors' sales. At the system fabricator level, the end use application was understood, but their visibility of the market was also not complete as systems would sometimes be constructed by other vendors, national laboratories, the regulatory body, or the production facilities themselves.

Market definition. Market definition is simply the customer and application space addressed by the ^{3}He proportional tubes. In this case the questions are "what is a safeguards system?" and "should domestic as well as international programs be considered?" For example, a waste assay system provides quantitative mass results for less desirable materials typically of only a few grams per container, so some argue that waste systems are not essential for safeguards. However, because a facility may generate hundreds of waste containers per year, the impact on the material balance can be dramatic. Therefore, we must consider that these waste assay systems are firmly within the safeguards arena. Domestic material control and accountability programs (in the United States and abroad) are often not considered safeguards although they serve the same ultimate function—protecting special nuclear material.

Changing market. Recently, significant impacts to the market have created a lull in the demand for ^{3}He or equivalent detectors. In the 1990s, ^{3}He cost as little as $100/L and generally formed an insignificant portion of the cost of a typical safeguards neutron assay system. As the cost for non-allocated ^{3}He rose above $1000/L, the detectors became the dominant cost in the production of these systems. For example, a commercially available active well coincidence counter (AWCC) containing ~42 liters of ^{3}He sold for approximately $86K in 1995; today that same system would cost between $300K and $400K if the gas were obtained on the open market. Essentially, these systems were no longer cost effective, and the market for these systems began to decline rapidly. The events of 3/11 in Japan have also created a temporary lull in the need for new systems as nations reevaluate their nuclear power programs. Both of these factors, the high cost and the Fukushima event, would cause a significant decline the apparent need for ^{3}He alternatives, but which is the dominating factor and whether the decline is permanent is not immediately apparent.

2.1.1 Historic Usage

Based on conference proceedings, facility tours, and a personal knowledge of the safeguards market, it is possible to estimate the historical use of ^{3}He in safeguards applications. During the period from 1997 to 2010, we can confirm the use of some 28,000 liters of ^{3}He in safeguards-related applications (Fig. 1). Based on an assumed market visibility of 60% (which is quite optimistic), we can estimate a total use of some 49,000 liters of ^{3}He over the course of 14 years or about 3500 liters/year of ^{3}He. Roughly 50% of the ^{3}He used in safeguards-related applications was associated with neutron-based waste assay systems.

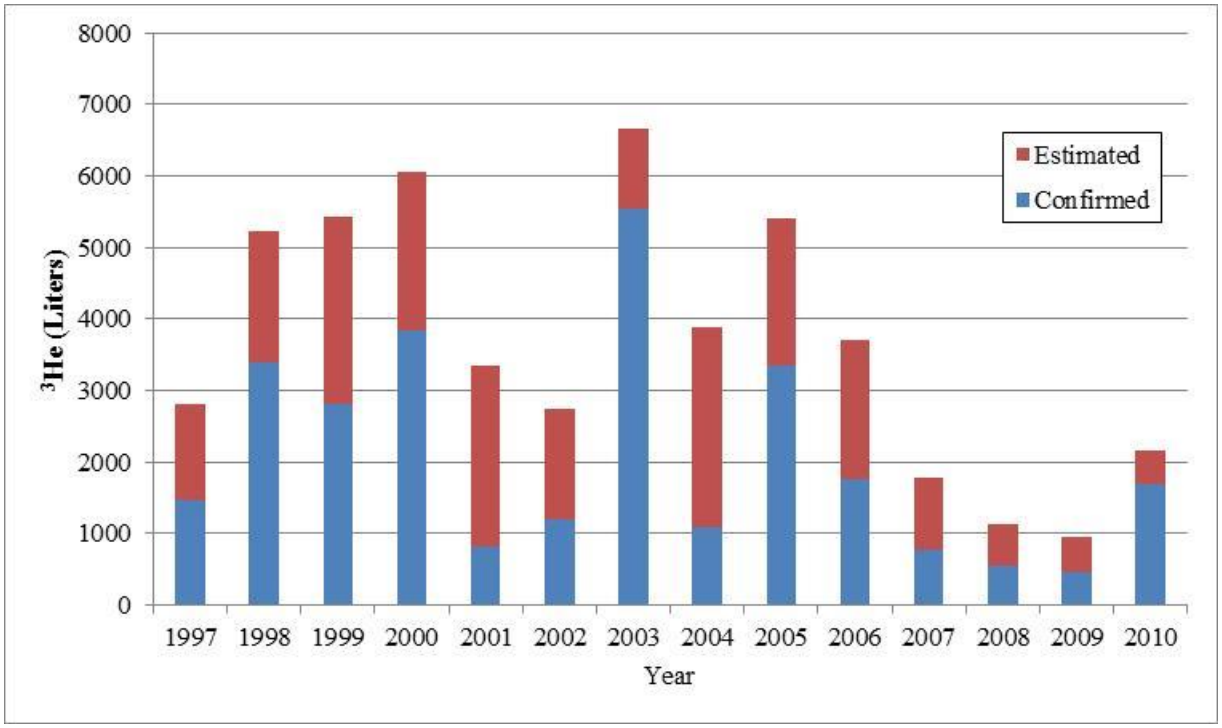


Fig. 1. Annual consumption of ^{3}He in safeguards and safeguards-related applications, 1997–2010.

2.1.2 Future Usage

Future needs for safeguards applications are obviously even more difficult to predict than the historic consumption is to validate. Queries to both the International Atomic Energy Agency (IAEA) and the European Atomic Energy Community (Euratom) have suggested that each of these international organizations foresees a need for approximately 250 liters of ^{3}He/year each. However, direct requirements from these agencies should not be mistaken for the actual safeguards demand for neutron counting systems. Historically, the production facilities have procured the lion's share of safeguards equipment in terms of cost and ^{3}He consumption. For example, consider the production facilities at Tokai. While US programs provided systems such as the PCAS, FPAS, and FAAS (i.e., the bulk of the unattended systems) the facilities procured directly, at the requirement of the IAEA, the WDAS, WCAS, FCAS, PSMCs, and other dual use equipment. The IAEA provides only a handful of small, portable systems such as the High Level Neutron Coincidence Counter (HLNCC-II) and PNCL. Direct procurement by regulatory bodies such as the IAEA and Euratom has historically been less than 10% of total safeguards demand.

Based on the historic consumption described in Fig. 1, we would have expected a continuing consumption of approximately 3500 liters/year. However, the increased costs of ^{3}He have caused a drop in the demand for ^{3}He, as evidence by the decline in demand from 2007 on. More recently, the events at Fukushima have altered the market dynamics by introduction of a perhaps temporary pause in the construction of new production facilities. An additional short-term factor influencing apparent demand is a robust recycling effort where excessed systems have been scavenged for their ^{3}He content. (Several thousand liters have been reclaimed since 2007.) Current consumption levels for safeguards purposes are estimated at 1000–1500 liters/year given current pricing and availability; however, it is expected that should ^{3}He costs return to previous levels (say <$500/liter), demand would return to near pre-2007 levels (2000 to 3000 liters/year).

3. GENERAL REQUIREMENTS FOR SAFEGUARDS APPLICATIONS [4, 5]

3.1 EFFICIENCY CONSIDERATIONS

A basic, albeit subjective, requirement for the alternative detector mechanism is that the neutron detector should provide a relatively high intrinsic neutron detection efficiency that is constant as a function emitted neutron energy. However, the use of coincidence counting, as well as practical considerations, requires that the active neutron detection volume be as compact as possible. The precision of a neutron coincidence measurement is generally limited by the accidental neutron coincidence rate, which is proportional to the coincidence gate time. Thermal neutron detectors such as the ^{3}He proportional tube require that the neutrons thermalize to yield high detection efficiencies. Since a thermal neutron travels 10 cm in 45 µs, the active detection volume for a thermal neutron detection technology should be limited to a thickness on the order of 10 cm to meet the timing requirements. Alternatively, detection mechanisms based on faster neutrons can tolerate larger active detection volumes, but the size of the assay system may become prohibitively large in terms of floor space, floor loading, and practicality of sample handling.

3.2 GAMMA-RAY SENSITIVITY

Unlike applications such as portal monitoring, where the typical gamma-ray exposure rate from the item of interest is near background, a safeguards system will generally be used for the assay of plutonium containers with gamma-ray surface exposure rates of up to 5 mSv/h (500 mR/h). However, higher gamma-ray exposure rates are not uncommon. For example, storage canisters of plutonium can produce surface exposure rates of up to 100 mSv/h (10 R/h), and systems used for measurement of spent fuel components or waste may have to accommodate exposure rates up to 10 Sv/h (the latter being relatively rare). Evaluation of the suitability of an alternative technology must also consider any modifications such as internal or external gamma-ray shielding that is required for proper operation of the detection system.

3.3 MATERIAL SAFETY CONSIDERATIONS

Safety requirements can vary dramatically from facility to facility. The viability of an alternate detection technique must also include a full safety analysis of the technology including not only the detection material but any associated requirements for the measurement system such as moderators, shielding, supporting framework, and electrical requirements. For example, the most obvious detection alternatives, BF_3 and liquid scintillation detectors, are both considered to be hazardous materials, and acceptability will be facility dependent. A system based on these techniques would likely require a remediation plan. The ideal neutron detection technology will be based on low hazard materials.

3.4 RELIABILITY AND MAINTENANCE

A typical safeguards system is installed within an active process facility with limited access. In many instances the equipment operators or inspectors have only occasional access to the instrumentation. Instrumentation used in unattended remote monitoring applications may have to operate without intervention in time frames of 6 months to 2 years. Evaluation of the mean time between failures (MTBF) must be considered for the collection of neutron detectors and associated electronics. Maintenance considerations for the neutron detection system as a whole must be considered, and required maintenance activities must be limited to recurrences longer than the typical inspection cycle (e.g., longer than 1 year intervals). The ^{3}He-based systems, with their long history in safeguard applications, were known to have MTBFs that were often far longer than the expected operating life of a typical nuclear facility. The new technologies will not have that pedigree, and consideration should be given to incorporation of detector level state of health monitoring.

Reproducibility of Components. Replacement of components in the field becomes complicated and time consuming when the detector assemblies are not identical. If the installation of a spare results in a change in efficiency, recalibration of the entire system may be required. To avoid this costly recalibration activity, the replacement detector technology should be based on reproducible detector modules.

3.5 ROBUST AND PRACTICAL

The measurement system will be installed within an operating facility, typically in an industrial environment. The development of the replacement detection technology must consider seismic qualification and flammability, vibrational, and environmental requirements of the facility. The system cannot be so fragile that normal plant operations would introduce noise or cause physical damage to the detectors. Additionally, the assembled detector must allow sample loading and maintenance access. For instance, a design that requires a hoist to load a sample would not be desirable for an attended assay system. Routine maintenance should not require complete disassembly of the detector assembly to replace a simple component. Finally, facility floor space is generally limited and expensive. Detection systems based on the alternative technologies should not be significantly larger than their ^{3}He counterpart.

3.6 INFRASTRUCTURE CONSIDERATIONS

Because the ^{3}He proportional tube was successfully applied to many different neutron application areas, the wide range of neutron detection systems and applications areas could be supported with a limited infrastructure. That is, the same basic physics, electronics, application software, operational protocols, and maintenance procedures could be used across a large installed base of neutron detection systems. A replacement neutron detection technology should be applicable to a significant portion of the safeguards application area to minimize associated development, training (of operators and measurement “experts”), and maintenance costs.

Scalability. The neutron detection systems used for safeguards encompass a wide range of container sizes and physical form factors. A suitable replacement technology should be scalable, allowing construction of measurement systems from small milliliter-sized sample vials to large containers with volumes of several thousand liters.

3.7 AFFORDABILITY

Affordability of a safeguards assay system must be considered in light of alternate technologies and the cost of not performing the measurement at all. For the near term, limited quantities of ^{3}He are available at a cost many times higher than just a few years ago. A reasonable expectation is that the cost of the replacement technology should be lower than that of a system based on ^{3}He at current pricing. Unfortunately, even with market pricing of $4000/liter of ^{3}He, this may not be the case. System based on alternative technologies with equivalent performance to the ^{3}He reference unit may be beyond the typical safeguards budget The cost of replacement technologies may drive target performance values to less desirable levels, degrading the worth of the safeguards measurement.

3.8 LIFE CYCLE COSTS

The cost of the replacement technology cannot be based simply on the cost of the neutron detector assemblies. The cost of the replacement technology should also consider

- system level design of the replacement systems;
- calibration requirements;
- required spares;
- training;
- procedures for operation, maintenance, and calibration;
- new electronics required;
- algorithms and software development;
- disposal cost (especially if hazardous materials are used); and
- the level of experience that is required for routine use and data interpretation.

4. FIGURE OF MERIT

A figure of merit (FOM) has been applied to neutron coincidence counting systems as a means to compare the relative performance of two or more different but similar designs. The mostly commonly used FOM for comparison of neutron coincidence counting systems is defined as

$$FOM = \varepsilon/\sqrt{\tau}$$

where

ε = neutron detection efficiency,
τ = characteristic die-away time.

This FOM is generally considered to be inversely proportional to the measurement precision for standard coincidence counting applications, where a larger FOM equates to smaller measurement uncertainty. However, this FOM only applies to the coincidence (doubles) rate for nonmultiplying samples and even then is only an approximate representation.

To examine the FOM, we must examine the uncertainty in the measured doubles rates, D. For the traditional ^{3}He-based neutron coincidence counting system, the electronics coincidence gate width, G, is set to 1.257·τ to provide the optimal measurement precision, yielding a doubles gate fraction of ~0.66. The uncertainty in the measured doubles rate is given by the following

$$\sigma_D \cong \sqrt{2S^2G + D}/\sqrt{t} = \sqrt{2S^2 1.256\tau + D}/\sqrt{t}, \tag{1}$$

where

t = acquisition time,
S = singles neutron count rate,
D = doubles rate,
G = coincidence gate width.

To evaluate the uncertainty in terms of efficiency and die-away time, we consider the point model equations for singles and doubles rates for a traditional neutron coincidence counting system

$$S = m_{240} \cdot \Phi \cdot M \cdot \varepsilon \cdot \overline{\nu_{s1}} \cdot (1+\alpha)\ , \tag{2}$$

$$D = \frac{m_{240} \cdot \Phi \cdot M^2 \cdot \varepsilon^2 \cdot f_d}{2} \cdot \left\{ \overline{\nu_{s2}} + \left(\frac{M-1}{\overline{\nu_{i1}}-1} \right) \cdot \overline{\nu_{s1}} \cdot \overline{\nu_{i2}} \cdot (1+\alpha) \right\} \tag{3}$$

where

m_{240} = the $^{240}Pu_{eff}$ mass,
Φ = the spontaneous fission rate per gram ^{240}Pu,
M = the self-leakage multiplication,
E = the neutron detection efficiency,
f_d = the doubles gate fraction,
α = the ratio of uncorrelated to correlated neutron emission,
$\overline{\nu_{s1}}, \overline{\nu_{s2}}$ and $\overline{\nu_{i1}}, \overline{\nu_{i2}}$ = the spontaneous and induced fission prompt factorial moments, respectively.

If we simplify the point model equations for the nonmultiplying case, we find

$$S = m_{240} \cdot \Phi \cdot \varepsilon \cdot \overline{\nu_{s1}} \cdot (1+\alpha)\ , \tag{4}$$

$$D = \frac{m_{240} \cdot \Phi \cdot \varepsilon^2 \cdot f_d}{2} \cdot \overline{\nu_{s2}}, \tag{5}$$

From which we can rewrite the doubles rate uncertainty equation (1) as

$$\frac{\sigma_D}{D} \cong \frac{\sqrt{2 \cdot \left(m_{240} \cdot \Phi \cdot \varepsilon \cdot \overline{\nu_{s1}} \cdot (1+\alpha)\right)^2 \cdot 1.256 \cdot \tau + \left(\frac{1}{2} \cdot m_{240} \cdot \Phi \cdot \varepsilon^2 \cdot f_d \cdot \overline{\nu_{s2}}\right)}/\sqrt{t}}{\frac{1}{2} \cdot m_{240} \cdot \Phi \cdot \varepsilon^2 \cdot f_d \cdot \overline{\nu_{s2}}} \tag{6}$$

which simplifies somewhat to

$$\frac{\sigma_D}{D} \cong \overline{\nu_{s1}} \cdot \frac{\sqrt{[2\cdot(1+\alpha)^2\cdot 1.256]\cdot\tau + \left(\frac{\overline{\nu_{s2}}\cdot f_d}{2\cdot m_{240}\cdot\Phi\cdot\overline{\nu_{s1}^2}}\right)}}{\frac{1}{2}\cdot\varepsilon\cdot f_d\cdot\overline{\nu_{s2}}\cdot\sqrt{t}} = a \cdot \frac{\sqrt{\tau + \frac{b}{m_{240}}}}{\varepsilon\cdot\sqrt{t}} \tag{7}$$

where we can now see the general inverse relationship $FOM \cong \varepsilon/\sqrt{\tau}$ applies to **large**, nonmultiplying masses.

$$\frac{\sigma_D}{D} \cong \lim_{m_{240}\to\infty}\left(a \cdot \frac{\sqrt{\tau + \frac{b}{m_{240}}}}{\varepsilon\cdot\sqrt{t}}\right) = \frac{a}{\sqrt{t}} \cdot \frac{\sqrt{\tau}}{\varepsilon} \tag{8}$$

Unfortunately, large mass, nonmultiplying samples are very rare occurrences and would not be assayable within a typical neutron coincidence counting system. So we see that the traditional FOM is flawed. To illustrate the limitations of the FOM, we have calculated the expected doubles rate measurement precision as a function of plutonium mass for a series of hypothetical mixed oxide (MOX) containers for three different counting systems, each with a FOM = 2.6 (i.e., the same as HLNCC-II). The three fictional assay systems are characterized by:

- System 1: HLNCC-II equivalent assay system, efficiency 17.8%, die-away time = 42 µs.
- System 2: Lithium-coated plastic scintillation slab, efficiency 9.2%, die-away time = 12.74 µs.
- System 3: Liquid scintillation slab, efficiency 0.7%, coincidence gate = 100 ns (equivalent to 80 ns die-away).

While the fictitious assay systems may have unrealistically low detection efficiencies, what is important here is that each would yield the same FOM. Fig. 2 shows the expected doubles rate precision as a function of plutonium mass ($^{240}\mathrm{Pu}_{\mathrm{eff}}/\mathrm{g} = 0.25$, $\alpha=1$). As can be seen in the plot, the expected precision is equivalent for the two slower counters, while the precision for the fast scintillator is markedly poorer. For completeness, and since we are ultimately interested in the mass results, not the coincidence rates, Fig. 3 plots the expected precision in the reported plutonium mass for the same three example systems. We see the same general behavior in the mass performance results.

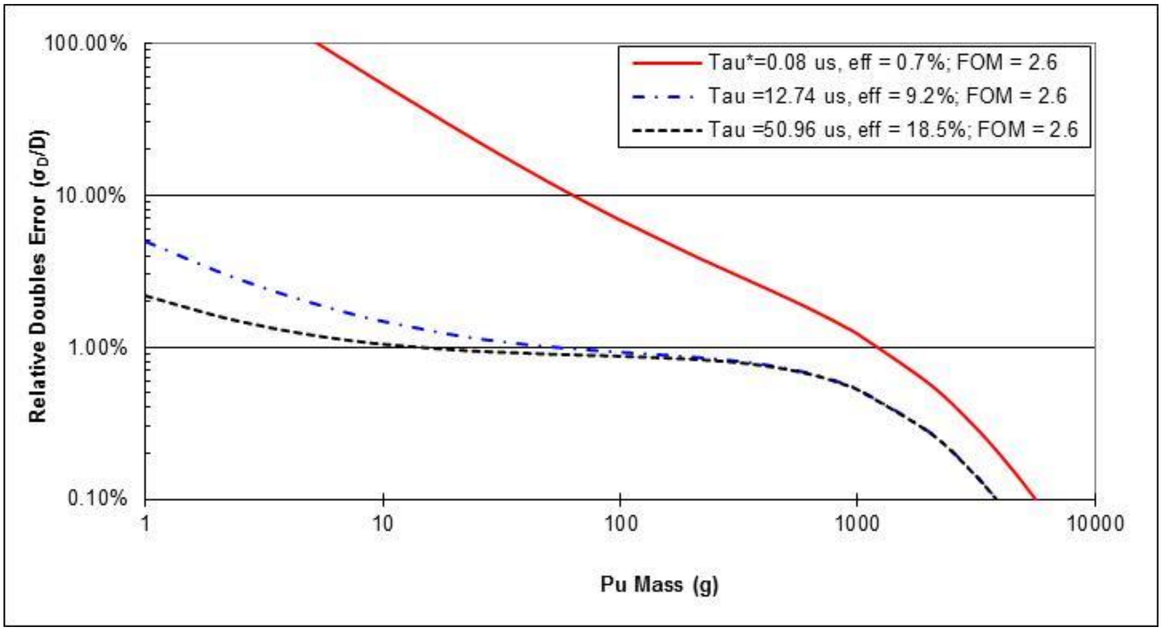


Fig. 2. Doubles rate precision for three systems with equal FOM but different efficiencies and die-away times. The solid red line represents the expected performance of a low-efficiency liquid scintillation detector using an effective die-away time = coincidence gate/1.256.

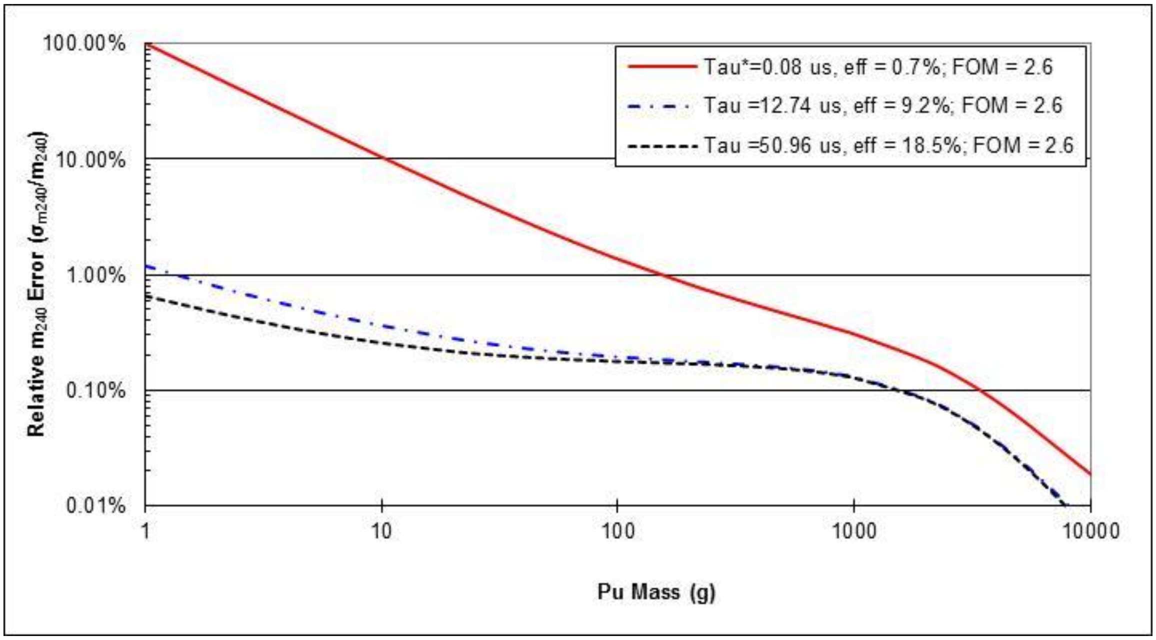


Fig. 3. Coincidence assay precision in the plutonium mass results for three systems with equal FOM but different efficiencies and die-away times. The solid red line represents the expected performance of a low-efficiency liquid scintillation detector using an effective die-away time = coincidence gate/1.256.

To better illustrate the limitation of FOM, we also calculated the expected measurement precision for three different plutonium masses as a function of efficiency for a fixed FOM. That is, we vary the die-away time along with the efficiency to keep an FOM of 2.6. Fig. 4 shows that the FOM is not sufficient to specify the performance needs for an assay system. A minimum efficiency is also required to achieve a specified performance goal.

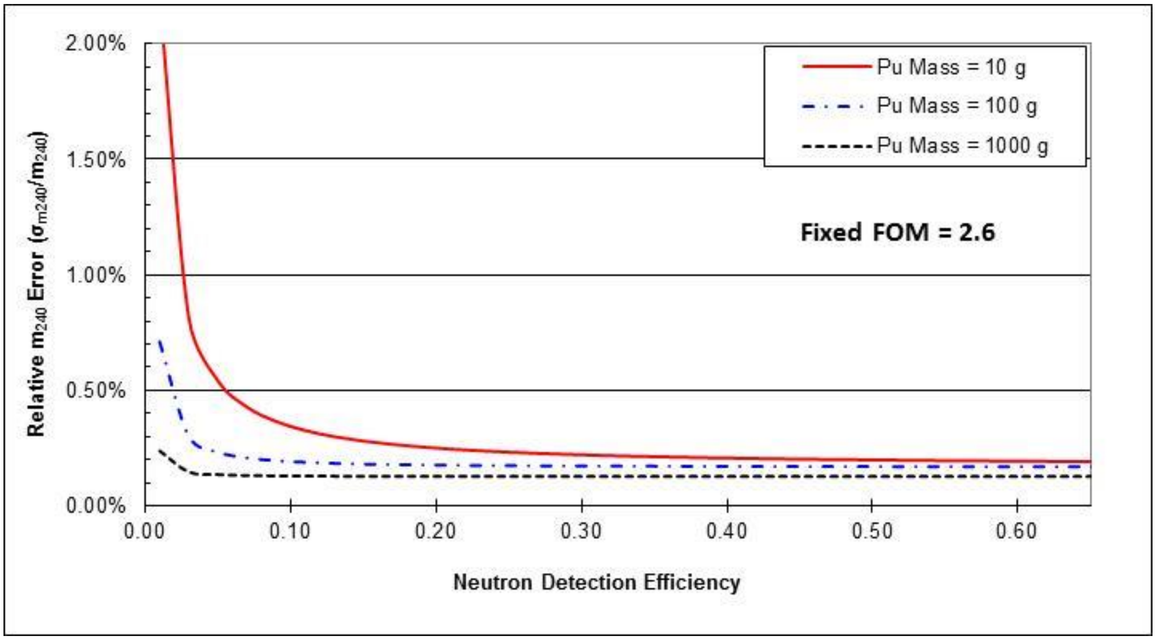


Fig. 4. Coincidence assay precision in the plutonium mass results for three plutonium masses. In this case, the neutron detection efficiency and die-away time vary to provide a constant FOM=2.6 (equivalent to the HLNCC-II). We see here that in order to cover the dynamic range of the HLNCC-II (~10 to 1000 g Pu) requires not only that we match the FOM; it also requires a minimal efficiency (10% in this case).

The FOM can aid in the evaluation of similar detection solutions for a specific assay problem where there are a limited number of variables to consider. However, it is necessary to remember that a figure of merit is a quick comparison tool that is generally intentionally simplified for ease of understanding. To properly evaluate the expected performance of an assay system, it is necessary to examine the measurement response over the full range of sample properties expected for the assay campaign.

4.1 PERFORMANCE COMPARISONS FOR MULTIPLICITY COUNTING

As already discussed, for the ^{3}He-based neutron coincidence system, the measurement performance is dependent on both the neutron detection efficiency and the time required for a neutron to be thermalized and detected, the die-away time. For the traditional neutron coincidence measurement (two neutron counting), the measurement precision at high count rates is proportional to 1/FOM = $\sqrt{\tau}/\varepsilon$, where ϵ is the efficiency and τ is the characteristic die-away time. For multiplicity counting systems, measurement precision is somewhat more complicated, but it is roughly proportional to $\tau/\varepsilon^{3/2}$. Good measurement performance requires both high efficiency and a short die-away time. The plots in Fig. 5 illustrate the importance of the die-away time on measurement precision as a function of mass for a typical scarp MOX measurement. While for coincidence counting of clean product materials high efficiency is more important than a short die-away time, the reverse is true for multiplicity assay of scrap materials where the uncorrelated events dominate the measurement.

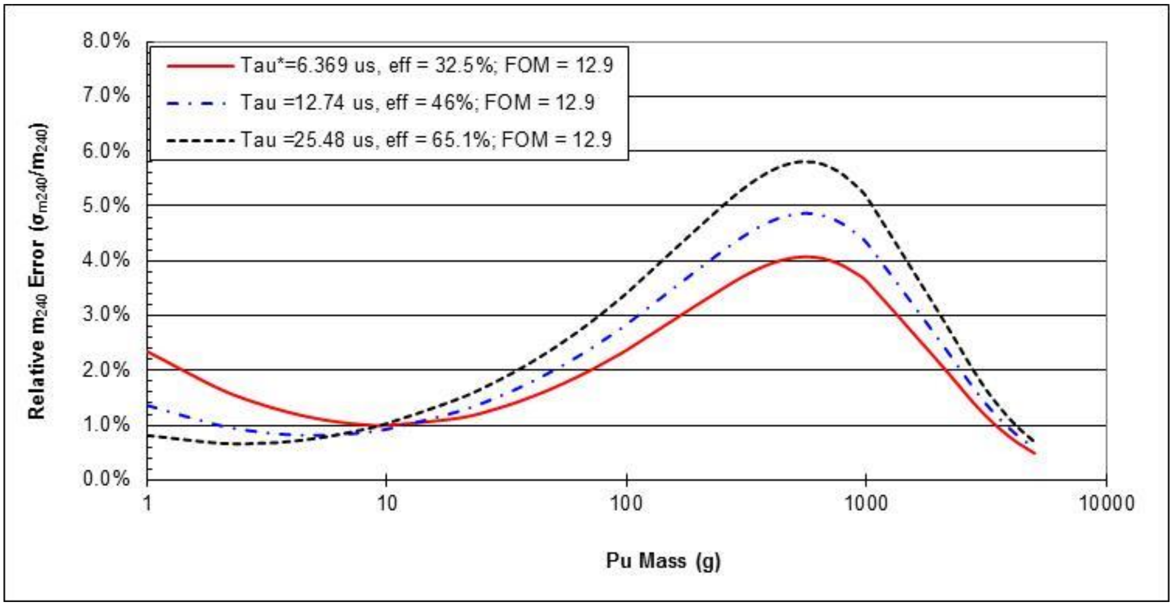


Fig. 5. Measurement performance for three multiplicity counting systems, each with FOM = 12.9 for MOX scrap with an alpha of 5. Due to the high count rates encountered with the scrap materials, both high efficiency and short die-away times are required, but in this case the shorter die-away time is more important.

5. EVALUATION OF ALTERNATIVE TECHNOLOGIES

5.1 REPRESENTATIVE ASSAY SYSTEM—THE HIGH LEVEL NEUTRON COINCIDENCE COUNTER

The High Level Neutron Coincidence Counter (HLNCC-II) is a passive neutron coincidence well counter, and is a staple of the safeguards field for the measurement of 1–10,000 g plutonium oxide and MOX powders in storage or process cans [6]. The HLNCC-II is a commonly deployed counter (more than 36 systems have been installed worldwide) but it is also a basic design that provides only modest assay performance (Table 2). For the evaluation of alternative detection technologies, the HLNCC-II represents an entry-level system—that is, any viable alternative neutron detection technology must be able to replace this system to be anything more than a niche technology or specialty system. A photograph and diagram of the commercially available Canberra JCC-31 [7] are presented in Fig. 6.

Table 2. HLNCC-II basic specifications

Pu mass range	0 to 3 kg—typical
$^{240}Pu_{eff}$ /g	5–40%
Alpha value	0.7–1.1—must be known
Assay cavity	40.6 × 17.0 cm H × diam
Nominal max container*	27 × 12 cm H × diam
Footprint	73.7 × 34 cm H × diam (including wheels)
Weight	55 kg
Gamma-ray tolerance	5—10 mSv/h—typical maximum
^{3}He per system	18 liters
Target Performance Values [3]	
Random error	1—5%
Systematic error	0.5—3%
Assay times (minutes)	10

*Size limit to fill height effects to <1%.

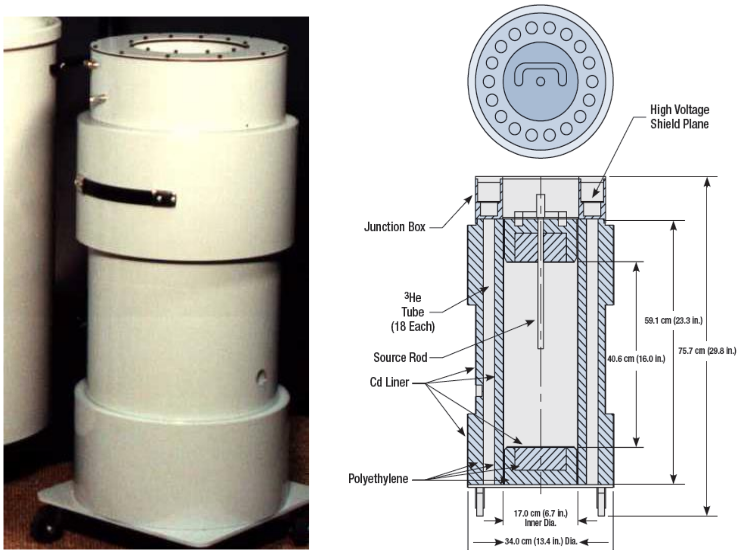


Fig. 6. The standard ^{3}He-based HLNCC [7]. The unit consists of an HDPE moderating assembly and a single ring of 4 atm ^{3}He proportional tubes.

5.2 EFFICIENCY CONSIDERATIONS

Brief discussions of the analyses for BF_3 and boron-lined proportional tubes are provided in the following sections as examples of the ongoing evaluation process. In terms of the general considerations detailed above, both tube types have been in use for several decades; they are robust designs with limited gamma-ray sensitivity; they are scalable; and their behavior is well understood and would require minimal effort to incorporate into the existing safeguards infrastructure. Table 3 provides a basic comparison of the boron-based proportional tubes with the ^{3}He proportional tubes.

Table 3. Comparison of the ^{3}He and BF_3 proportional tubes, in the context of typical nondestructive assay systems for safeguards, waste, and process control applications

Parameter	^{3}He	BF_3	Boron-Lined
Reaction mechanism	$^3He(n,p)^3H$	$^{10}B(n,\alpha)^7Li$	$^{10}B(n,\alpha)^7Li$
Thermal cross section (barns)	5330 (^{3}He)	3840 (^{10}B)	3840 (^{10}B)
Typical enrichment	NA	90% ^{10}B	90% ^{10}B
Typical partial pressures in NDA system	4–10 atm	0.9 atm	NA
Quench gases	Ar/CH_4, Ar/CO_2, CO_2, N_2	BF_3	Ar/CH_4, Ar/CO_2, CO_2
Typical tube diameters	6–51 mm	13–51 mm	8–51 mm
Gamma insensitivity	Good	better	better

5.3 BF_3 PROPORTIONAL TUBES

The BF_3 proportional tube has been in use as a neutron detector for several decades, but its widespread use has been limited because of the chemical properties of the BF_3 gas. BF_3 is a hazardous and corrosive material; in the presence of moisture, it forms fluoroboric and fluoric acids. While BF_3 gas is safely contained within the stainless steel walls of the proportional tube, there are both perceived and genuine safety concerns associated with these tubes. However, these tubes are currently manufactured and used on a commercial basis, so it may be possible to mitigate the safety concerns.

Performance as well as handling limitations place a practical limit on the fill pressure of <1 atm on these tubes, such that a BF_3-based system cannot achieve the same high levels of performance as a system using ^{3}He tubes. The impact of the pressure limitation of the BF_3 tube is best considered in terms of the transmission of a thermal neutron through a standard tube. For example, a typical ^{3}He tube with 4 atm partial fill pressure and diameter of 25.4 mm will absorb 77% of all thermal neutrons incident normally to the surface of the tube. In contrast, a 25.4 mm diameter BF_3 tube with the maximum practical fill pressure will absorb only 18% of the thermal neutrons. In addition to the lower overall efficiency, this means that on average a neutron will need to travel much farther prior to detection, resulting in a longer characteristic die-away time for the BF_3 based system.

The Monte Carlo N-Particle eXtended (MCNPX) transport code [8] was used to examine various potential configurations for a BF_3-based alternative to the standard HLNCC. This cursory study examined various arrangements of the assay cavity and tube positions. Ultimately, a two ring arrangement using 2 in. diameter BF_3 tubes was selected as being the most cost effective configuration to achieve performance similar to the standard unit. A screen shot of the tube arrangement from the MCNPX simulations is shown in Fig. 7.

The expected efficiency for the hypothetical BF_3-based neutron counting system is approximately 26%, which is much higher than the standard HLNCC. However, as can be seen in Fig. 8, the coincidence die-away time for the BF_3-based system (85 μs) is much slower, degrading the measurement precision and requiring the higher detection efficiency to compensate. Fig. 9 shows the expected relative precision in the Reals coincidence rate as a function of plutonium mass for MOX materials from 1 to 7000 g. The expected performance values are effectively equivalent for the purposes of this exercise.

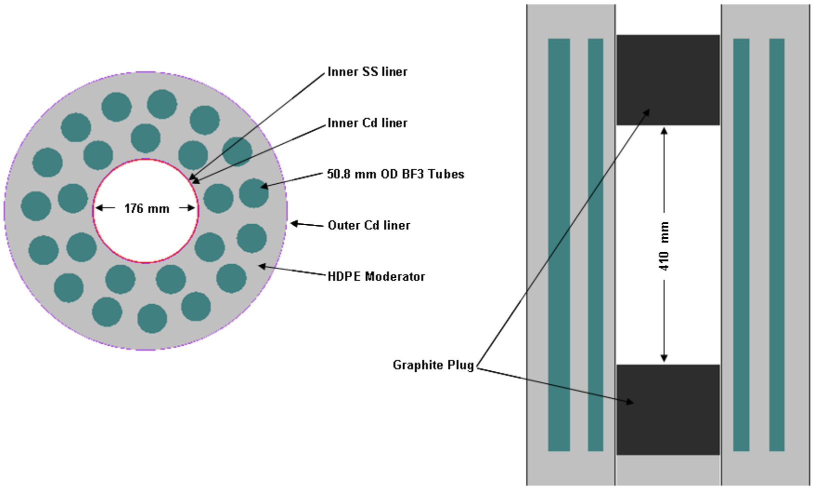


Fig. 7. Screen shot of the MCNPX input file for the potential BF_3 replacement HLNCC. This alternate design would be larger, and it would require the use of 2 in. diameter tubes and an increase in the number of tubes compared to the standard ^3He-based system.

The feasibility of creating BF_3 proportional tube equivalents of several standard neutron counting systems was examined via MCNPX simulation. These included several well counters, drummed waste assay systems, and a material accountancy glove box system. The study indicates that BF_3 equivalent systems can be constructed for nondestructive assay (NDA) systems where the value of $\sqrt{\varepsilon}/\tau$ is less than 7.5 for the ^3He-based system.

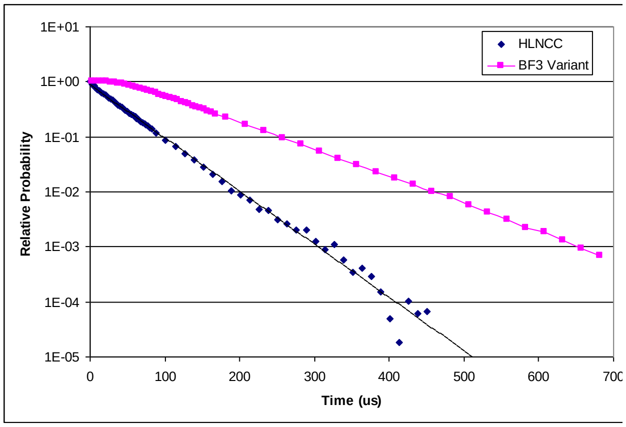


Fig. 8. MCNPX-generated die-away time plots for the standard HLNCC and the BF_3-based counter. Even with the much higher detection efficiency, the response of the BF_3 system is much slower than for the ^3He-based counter.

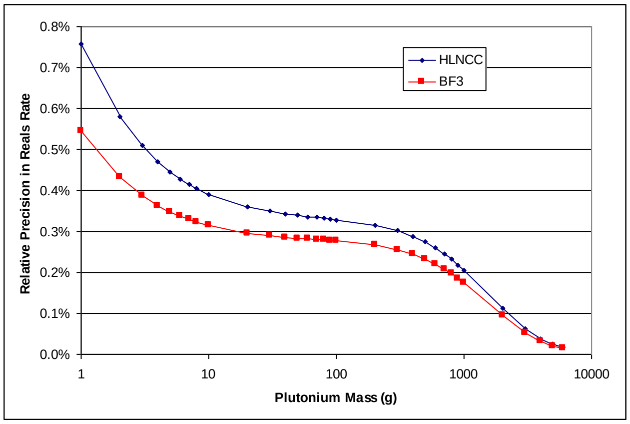


Fig. 9. Estimated relative precision as a function of plutonium mass for clean MOX powder for the standard HLNCC and the proposed BF_3-based system.

5.3.1 Safety Considerations for the BF_3 tubes

BF_3 could potentially replace ^3He in many existing safeguards applications, however, the BF_3 is a corrosive and hazardous material. Because BF_3 is used in many commercial applications (e.g. electronic chip manufacture) there are several well established industrial safety limits associated with the gas.

- The Permissible Exposure Limit (PEL) established by the Occupational Safety and Health Administration (OSHA) is 1 ppm (3 mg/m^3) ceiling.

- The American Congress of Governmental Industrial Hygienists (ACGIH) sets the Threshold Limit Value (TLV) of 1 ppm ceiling.

- The National Institute of Occupational Safety and Health (NIOSH) has established an Immediately Dangerous to Life and Health (IDLH) exposure level for BF_3 of 25 ppm.

To illustrate the potential hazard level associated with BF_3, consider the impact of a single BF_3 proportional tube vented within a closed room. A typical 1 atm, 5 cm diam, 60 cm long BF_3 tube contains approximately 3 g of BF_3. Assuming uniform dispersal, the PEL and TLV limits would be exceeded for any room smaller than 1000 m^3 or roughly 16 m × 16 m ×4 m (50 ft × 50 ft × 13 ft) in extent. The BF_3 released from the rupture of a single tube would be sufficient to be considered immediately dangerous to life if it were opened inside a typical closed office (12 ft × 12 ft × 10 ft).

Within the safeguards community, there have been events when ^{3}He tubes have been breached. For example:

- A handheld JSP-12 handheld survey instrument was inadvertently dropped down a flight of stairs, resulting in the release of the ^{3}He from the tubes.

- A field modification performed on an AWCC resulted in a hole being inadvertently drilled through a ^{3}He proportional tube and release of the ^{3}He gas.

Although these occurrences were unlikely and uncommon, damage to the proportional tubes (^{3}He or BF_3) in the field is possible and must be considered a credible event. It has been argued that since the BF_3 tubes are filled to slightly less than 1 atm, the release of the gas will be slow, and danger to health is actually quite low. However, at this time BF_3 tubes are generally not allowed in nuclear fuel fabrication facilities

5.4 BORON-LINED PROPORTIONAL TUBES

The ^{10}B-lined proportional tube has been in use as a neutron detector for several decades but its widespread use has been limited by the low detection efficiency achievable from traditional designs. Unlike the BF_3 proportional tube, the ^{10}B-lined tube is not considered a hazardous or corrosive material. The neutron sensitive material, the boron, is deposited in a thin layer on the inner wall of the proportional tube. The proportional gas is a common type such as Ar/CH_4 or Ar/CO_2. When a neutron is absorbed by the ^{10}B, two energetic charged particles are created (^{4}He and ^{7}Li), traveling in opposite directions. This means no more than one of the two reaction products will be emitted in the direction of the proportional gas (Fig. 10). The range of the ^{4}He and ^{7}Li particles in the boron layer is very short, 3.8 and 1.7 microns respectively, limiting the maximum usable coating thickness for which an absorbed neutron will produce an output pulse. The short range of the particles in the boron layer limits the useable thickness that can be applied to a surface and hence limits the intrinsic efficiency of the tube. There are a number of strategies under investigation to increase the useable surface area per unit volume. For instance, bundling five tubes of 8 mm diam would provide 1.6 times the overall surface area of a 25.4 mm diam tube and fit within the same overall volume (Fig. 11).

As with the BF_3 tubes, MCNPX was used to simulate the performance from a series of conceptual designs based on various configurations of boron-lined proportional tubes. Configurations using 2, 3 and 4 rings of tubes were examined, with tube diameters of 8 mm to 50.8 mm. Examples of the configurations examined are shown in Fig. 12. For each configuration the boron thickness was varied to determine the optimal thickness based on the predicted measurement precision. The performance of the models for the boron-lined tubes was benchmarked against measurements made on a slab neutron detector constructed with boron-lined proportional tubes. The relative accuracy of efficiency measurements is estimated to be +/-10% based on the benchmark results. Achieving neutron detection efficiencies in excess of 20% for a well counter configuration proved difficult with the arrangements considered. For each of the compound and bundled tube configurations depicted in Fig. 12, it was possible to contrive a system providing the same performance as the ^{3}He-based HLNCC.

The preliminary results indicate that boron-lined equivalent systems can be constructed for NDA systems where the value of $\sqrt{\varepsilon}/\tau$ is less than 5.5 for the ^{3}He-based system. However, it will be necessary to construct a full-scale prototype counter to confirm this result.

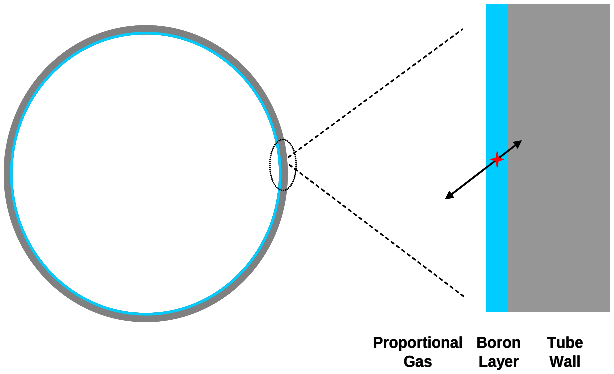


Fig. 10. When a neutron is captured in the ^{10}B layer, only one of the two reaction products will be emitted in the direction of the proportional gas. The other will be absorbed in the boron layer or the tube wall.

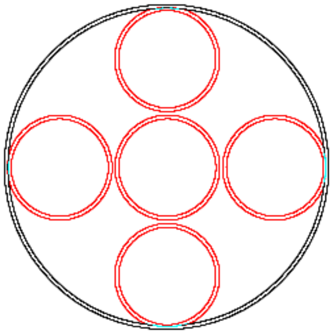


Fig. 11. A bundled tube concept designed to increase the useable surface area within the same volume of a single larger tube.

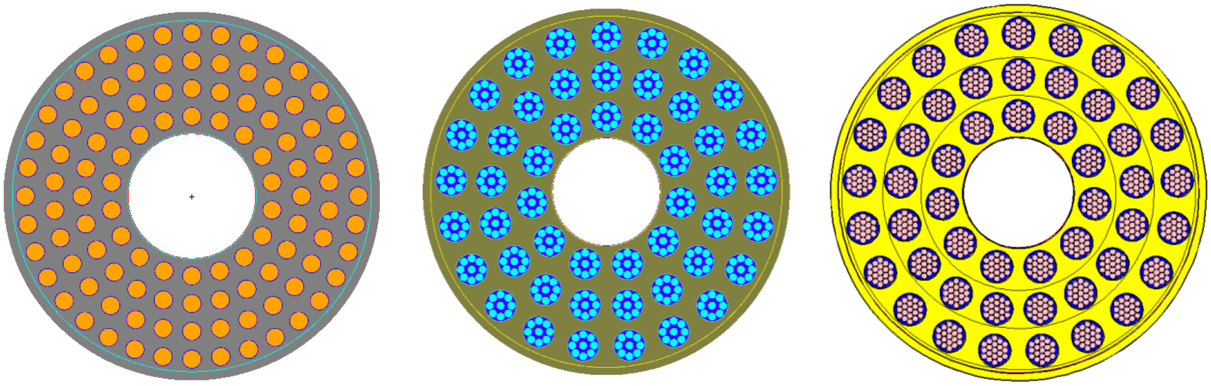


Fig. 12. Screen shots of the MCNPX input files for three potential B_{10}-lined alternative designs that would provide performances equivalent to a traditional ^{3}He-based HLNCC. The design on the left uses four rings of 1 in. diam tubes; the center design is based on the compound tube arrangement offered by Centronic, LLC; and the design on the right is based on bundles of 19 tubes of 8 mm diameter. Each of these alternate designs will be larger and require more tubes than the standard ^{3}He-based system.

5.5 BORON STRAW DETECTORS (SMALL DIAMETER PROPORTIONAL TUBES)

The discussions above suggest that the key to improved performance for the boron coated tubes is to distribute a large number of very narrow diameter tubes throughout the moderating assembly. A small diameter ^{10}B lined proportional tube has been developed by Proportional Technologies Inc. that provides increased active surface area per unit system volume. These detectors were dubbed “straw detectors” due to early fabrication techniques in which the active boron coating was deposited on a thin metal strip that was then wound in a helical fashion (similar to a paper drinking straw) to form a cylinder [9]. These detectors are now fabricated by forming the substrate material over a mandrel, but the name has been retained. The small diameter (8 mm) of the straws allows a high ratio of active surface area to detector volume and a reduction in boron coating thickness to provide improved detection efficiency in comparison to the larger diameter proportional tubes discussed in Section 5.4. a proto-type neutron coincidence counting detector was designed and fabricated by PTI using ~800 straw-based proportional tubes arranged within a moderating HDPE annulus (Fig. 13).

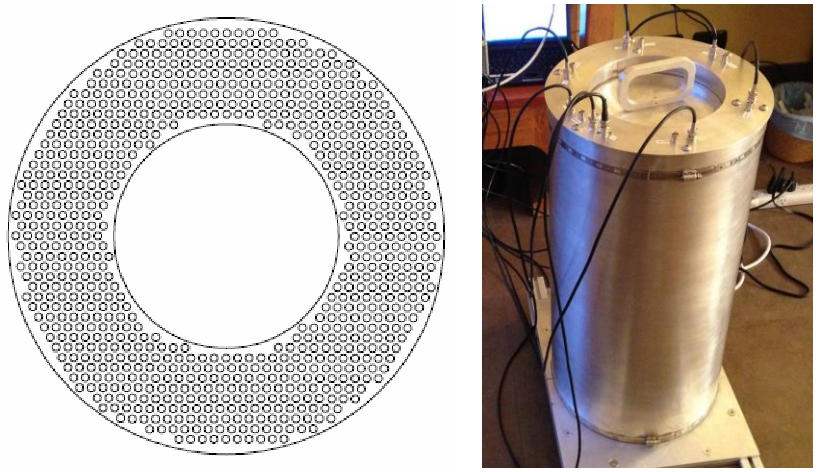


Fig. 13. Sketch of the straw-based neutron counting system (left) [9] and a photograph of the prototype neutron coincidence counter, a functional equivalent to the HLNCC-II system.

Their prototype system sought to replicate the performance of the HLNCC-II system, including its assay cavity dimensions and external footprint. The system's performance was recently demonstrated at the ^{3}He Alternatives for International Safeguards Workshop that took place at the Joint Research Centre (JRC) in Ispra, Italy, in October 2014 [10]. Preliminary results from the workshop indicate that the straw-based HLNCC-II equivalent equaled the measurement performance of the HLNCC-II. The counter was determined to provide a neutron detection efficiency of 13.6% with a die-away time of 26 µs, yielding an FOM of 2.66 compared to 2.75 for a commercial off-the-shelf (COTS) ^{3}He-based HLNCC-II.

5.5.1 Boron Straws as a Potential Replacement Technology

The straw detectors offer certain advantages relative to other potential alternative detectors such as BF_3, liquid or plastic scintillators:

- Of the detector types evaluated at the ^{3}He Alternatives for International Safeguards Workshop, only the straw detector based system was able to match the performance of the ^{3}He-based equivalent.
- The prototype system was of similar footprint and weight to the ^{3}He system. With the potential exception of the Symetrica blade design, all the candidate replacement technologies will require larger footprints to meet the performance of their ^{3}He counterpart.
- Minimal impact on infrastructure: the straws are proportional tubes and are compatible with existing acquisition electronics (shift-registers and list-mode) and software (e.g., INCC). They are effectively a drop-in replacement for ^{3}He detectors.
- Gamma-ray sensitivity: The boron straw detectors are expected to be less sensitive to gamma-rays than other candidate technologies and will have a performance that is similar to the ^{3}He systems.
- Safety: The straws use a low pressure (<1 atm) quench gas, are nontoxic, and pose no new hazards.
- Transportation: The straws will not need to be shipped as pressure vessels

However, there are still some outstanding concerns with this technology:

- Reliability: unlike the decades old BF_3 and traditional ^{10}B-lined proportional tubes, the straw detectors (and in particular the straws used in this prototype system) have a relatively limited operational history. The sheer number of straws required to fabricate a relatively simple system such as the HLNCC-II equivalent (804 straws versus 18 ^{3}He tubes) will require extensive long-term testing before its suitability for safeguards applications can be determined.
- Scalability: It is estimated that an AWCC equivalent system (26% efficient with 50 µs die-away) will require >1700 straws. Expansion to larger, more efficient systems will require a very large numbers of detectors. Scaling to these larger systems will need to be demonstrated.
    - PTI is developing a complex star-shaped straw detector design that further increases the active surface area to detection volume. In principle, such a detector could reduce the number of individual detectors required to achieve a given performance level.

5.6 SUMMARY OF EXPECTATIONS FOR THE BORON-BASED PROPORTIONAL TUBE CONFIGURATIONS

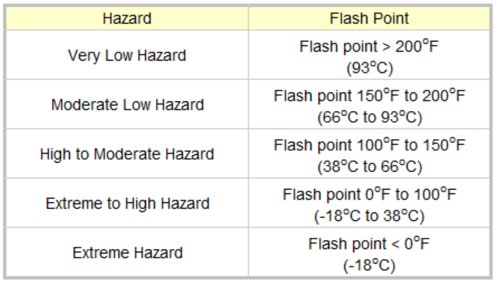
Based on the MCNPX results, the feasibility of constructing a boron proportional tube–based equivalent for several standard neutron counter arrangements is summarized in Table 4. The MCNPX simulations indicate that BF_3 could provide replacement counting systems for a large fraction of the typical counting configurations, while the boron-lined tubes will likely only be suitable for applications requiring lower performance. The models also indicate that the more complex tube structures provide improved performance; however, the cost effectiveness of the complex tubes must still be evaluated.

Table 4. Standard neutron counting systems and the possibility of constructing a BF_3 or boron-lined tube-based replacement system

System Description	Standard ^{3}He System Performance				BF_3 Variant Feasible	B-lined Variant Feasible	B-lined Straw Variant Feasible
	Neutron Detection Efficiency (%)	Die-Away Time (μs)	Gate Width (μs)	Sensitivity (Pu-240) (Reals/s/g)			
Slab, Well, and Collar Counters							
Portable slab counter (@ 30 cm)	2.2	50	64	NA	Yes	Yes	Yes
Universal fast breeder reactor	7.0	22	64	3.4	TBD	TBD	Yes
UNCL—coincidence collar	14.6	54	64	12	Yes	Yes	Yes
HLNCC	17.9	42	64	18	Yes	Yes	Yes
Curved slab counter	19.4	75	95	21	Yes	Yes	Yes
Flat squared counter	24.4	56	64	32	Yes	No	Yes
AWCC—active well	24.9	52	64	36	Yes	No	Yes
INSV—inventory sample	35.0	45	64	72	TBD	No	TBD
OSL—on-site laboratory	40.0	54	64	88	TBD	No	TBD
PSMC—multiplicity counter	54.0	50	64	175	No	No	TBD
Drum Counters							
WCAS—Waste Crate Assay	16.6	74	128	13.5	Yes	Yes	Yes
^{252}Cf drum shuffler	17.5	80	128	17	Yes	Yes	Yes
Waste drum assay system	19.4	79	128	20	Yes	Yes	Yes
IWAS—passive/active drum	27.0	44	128	47	Yes	No	TBD
HENC—drum assay system	30.8	52	128	52	Yes	No	TBD
HNMC—hexagonal multiplicity counter	51.0	28	32	137	No	No	TBD

6. SCINTILLATION COUNTERS

There are several distinct types of scintillation detectors under evaluation as potential replacement technologies for safeguards applications. The various neutron scintillation detectors have been in wide use in experimental physics applications for many years; however, unlike the ^{3}He and boron proportional detectors, they have not been used extensively in the harsh industrial environments encountered in international safeguards applications. These detectors will only be discussed briefly in this report due to the relative immaturity of the technologies when applied to international safeguards applications.



Hazard	Flash Point
Very Low Hazard	Flash point > 200°F (93°C)
Moderate Low Hazard	Flash point 150°F to 200°F (66°C to 93°C)
High to Moderate Hazard	Flash point 100°F to 150°F (38°C to 66°C)
Extreme to High Hazard	Flash point 0°F to 100°F (-18°C to 38°C)
Extreme Hazard	Flash point < 0°F (-18°C)

- High pressure noble gas detectors—recoil detection
  Efficiency is based on high fill pressure (typically greater than 100 atm)
  - Detects fast neutrons (charged particle recoil)
  - Generally inefficient, requiring large detector volumes

- Liquid—recoil detectors (e.g., NE-213)
  - Fast neutron detection (proton recoil)
  - High intrinsic detection efficiency
  - Gamma sensitive—requires PSD
  - Historically toxic or carcinogenic and low flash point.
    Newer, safer detector materials are available
    (e.g., EJ309 with flash point 144°C).
  - Prone to leakage

- Liquid —capture gated detectors (e.g., BC523A)
  - Thermal/Fast neutron detection (neutron capture/proton recoil)
  - High intrinsic detection efficiency
  - Gamma sensitive—requires pulse shape discrimination (PSD)
  - Toxic or carcinogenic and low flash point (~-8°C)
  - Prone to leakage

- Solid—plastic scintillators
  - Fast neutron detection (proton recoil)
  - Medium intrinsic detection efficiency
  - Gamma sensitive—requires PSD
    PSD-capable plastics are a relatively recent advancement.
    A first use of these detectors for safeguards applications was conducted at Oak Ridge National Laboratory (ORNL) during FY 2015 [11, 12].

- Solid—Glass Scintillators
  - Fast neutron detection – capture on ^{6}Li (NE912) or ^{10}B (NE110)
  - Low intrinsic detection efficiency
  - Very gamma sensitive—requires pulse height/shape discrimination

- Solid—Coated/Plastic Scintillators
  - Thermal neutron detection (capture ^{6}Li)
    Typically Li mixed in scintillating matrix such as ZnS applied as a coating a light pipe
  - Medium intrinsic detection efficiency (depending on configuration)
  - Low to medium gamma sensitive—requires pulse height/shape discrimination.
  - Prototype system designed and fabricated and demonstrated by Symetrica, Inc., at the ISPRA ^{3}He workshop in 2014 [10]

The technologies in the list above are diverse, but they are all scintillation based detector technologies and they all ultimately function by the detection of light emitted in a secondary reaction following the impact of the neutron on the detector assembly. Historically, these detectors would require the use of a photomultiplier tube (PMT)-based electronics chain with the following disadvantages that will need to be overcome for routine use of any of these technologies:

- **Temperature effects.** The PMTs and associated electronics are more susceptible to temperature than the proportional tube-based technologies, leading to larger efficiency variations and complication of the PSD process.
- **Footprint**. The traditional PMT is large in comparison to the active detection volume such that the footprint of the scintillation-based system is expected to be far larger than the ^{3}He system it is intended to replace.
- **Reliability.** The PMT-based data acquisition systems are significantly more complex than the ^{3}He proportional tube based systems and are somewhat less robust on a per detection channel basis. This will lead to lower reliability than the safeguards community is accustomed to and will incur both economic costs and lead to a negative perception of the technology.
- **Data Rates.** The scintillation detectors are sensitive to both neutron and gamma rays, and a sophisticated analysis is required to separate the two signals. Historically, this processing was done during post-acquisition analysis. However, the combined neutron and gamma-ray count rates would be on the order of 10 times larger than encountered using the simpler proportional tube-based detectors. Much of this post processing will need to be performed on the fly, requiring the addition of high-performance pre-processors to the system.

To mitigate these issues, many detector developers and system integrators are evaluating the use of solid state PMTs. These compact devices would allow a reduction in the system footprint, potentially lower the system cost, and increase reliability.

Significant public and private development has been expended on adapting these scintillation detectors for use in international safeguards. The following sections discuss the observed performance of three of these technologies and their potential application to safeguards.

6.1 PSD PLASTIC SCINTILLATION DETECTOR FOR ACTIVE WELL COINCIDENCE COUNTING

The Uranium Neutron Collar (UNCL) and the AWCC provide quantitative mass analysis of bulk uranium items by means of active neutron interrogation and neutron coincidence counting. In both cases, the measurements suffer from poor signal-to-noise ratios and require long assay times to achieve the desired measurement precision. The quantitative assay of bulk uranium and plutonium materials in international safeguards applications has traditionally been carried out using moderate-to-high efficiency (in this case $\varepsilon > 10\%$) but relatively slow thermal neutron coincidence assay systems. However, the same assays may be accomplished using less efficient but faster scintillation detection systems while providing significant improvements in measurement precision. In this section, we present a brief summary of a recent study at ORNL, sponsored by the Office of Nuclear Safeguards and Security, demonstrating that an existing non-optimized, single detector slab consisting of an array of PSD-plastic fast-neutron detectors can equal the performance of the traditional UNCL and AWCC systems (much of this section is taken from reference [12] to be published at the 2015 meeting of the Institute of Nuclear Materials Management).

To demonstrate the feasibility and investigate the potential performance of the PSD-capable plastic scintillation detectors, an existing neutron imaging system was configured to simulate an AWCC. The benefits of the pixilated scintillation-based detection system include:

- **Improved signal to noise.** The limiting factor of the traditional ^{3}He based AWCC is the large accidental coincidence rate produced by the interrogating neutron flux from the Am(Li) sources. The fast scintillation detectors eliminate this background due to very short coincidence gating (100 ns vs. 64 µus for an ^{3}He-based system) and by means of pulse height discrimination taking advantage of the energy resolution of the scintillation detectors.

- **Reduced dead time.** Typical large-volume liquid scintillation detectors are effectively dead each time a neutron is detected. That is, if one detector from an eight-detector array detects a single neutron, it is no longer available to detect a second neutron from a given fission event. Although the detectors themselves are very fast, the eight-detector array would have a 12.5% minimum dead time. The pixilated PSD-plastic system uses a much finer array resulting in much lower dead times.

- **Reduced cross talk.** A unique problem to the recoil-based scintillation detectors is that a single neutron may be detected more than once. That is, a neutron may scatter in one detector, depositing sufficient energy for detection, and then still have sufficient energy to travel to a second detector and create a detectable signal. Because all the interactions from a single fission event take place within nanoseconds of each other, it is not possible to distinguish the twice detected neutron from two individual neutron events. The typical solution to this cross-talk issue requires a masking procedure to turn off the adjacent detectors likely to participate in a cross-talk event. This means that if two neutron events are detected within the coincidence time window within the group of detectors, they are counted only as a single event. The pixilated detector array minimizes the fraction of the detector included within the group and therefore results in fewer counting losses.

Elements of ORNL’s Large Volume AWCC [the end plugs, cavity liner and Am(Li) sources] were simply placed in front of the imaging system (Fig. 14). The low-energy interrogating neutron flux from the Am(Li) sources was removed by pulse height/PSD, resulting in essentially zero random coincidences. Keeping in mind that this system was in no way optimized for use in safeguards counting, the observed performance for the measurement of a series of enriched uranium standards was roughly equivalent to that of the fully optimized ^{3}He-based AWCC system as shown in Fig. 15. The figure presents the observed measurement precision using the non-optimized system along with the expected performance for a four-slab system arranged in a rectangular configuration about the uranium container. A factor of 2 to 4 improvement in measurement precision is possible within the same count time using the scintillation counting system.

The study also highlighted some potential downsides to the use of the scintillation detectors. These include:

- **Increased footprint.** The detector/PMT assemblies are much thicker than the corresponding detector wall in the ^{3}He-based system. An optimized PSD-plastic–based AWCC will likely be two times larger in width and length and slightly taller than the traditional ^{3}He system.

- **Increased weight.** The resulting system and support structures will be approximately five times heavier than its ^{3}He-based counterpart.

- **Neutron background sensitivity.** Background count rates were more difficult to quantify and control due to the open structure of the proof of concept system.

- **Gamma-ray sensitivity.** For the active neutron assay of LEU and HEU items, gamma-ray sensitivity is not expected to represent a significant interference. However, gamma-ray sensitivity is expected to be a concern for MOX items where gamma-ray exposure rates will typically range from 10 to 1000 mR/h. This may be mitigated by use of internal shielding but at the expense of additional increases in footprint and weight. A more complete system characterization including a detailed gamma-ray sensitivity study will be required before routine deployment is possible.

- **External gamma-ray backgrounds.** Operational nuclear facilities, and in particular storage areas where these systems would be routinely used, will have elevated background environments. External gamma-ray shielding may also be required.

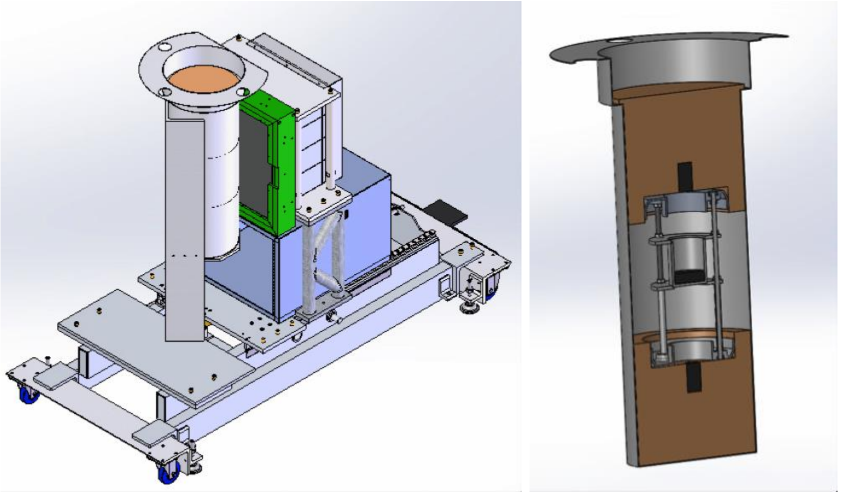


Fig. 14. The fast neutron imaging system configured for the proof of principle work (left). The instrument uses a modified Am(Li) source holder from the ORNL L-AWCC system. A cutaway of the sleeve showing the sleeve, the ^{241}Am(Li) plugs, and the mounting of the sources internal to the sleeve (right) [12].

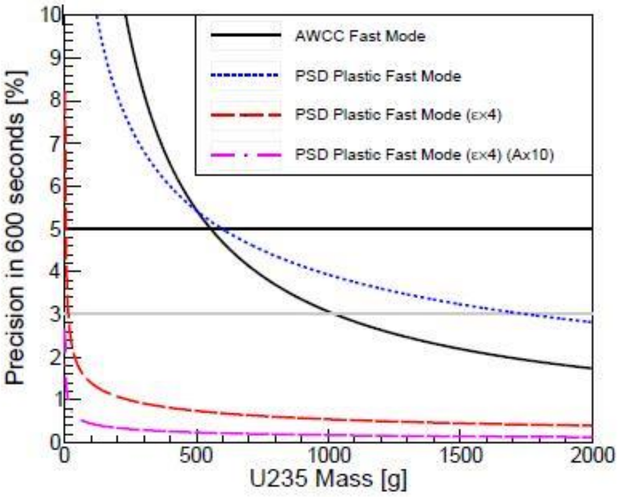


Fig. 15. The performance of the AWCC and PSD-plastic operated in "fast mode" with two interrogating sources in each end-plug each with activity of 5 × 10^4 neutrons·s^{-1} [12]. Also shown is the predicted performance for a reconfigured PSD-plastic system with four times the singles efficiency and with interrogating activities scaled by a factor of 10.

6.2 LIQUID SCINTILLATION DETECTION FOR (ACTIVE) NEUTRON COINCIDENCE COLLAR COUNTING

The IAEA, with the European Commission's JRC in Ispra and Hybrid Instruments, Ltd. (United Kingdom), has developed a liquid scintillator-based neutron coincidence counter for the assay of fresh

LEU fuel assemblies. The current standard assay system is the ^{3}He based UNCL. Similar to the AWCC, a large Am(Li) source is used to induce fission within the fuel assembly, and the coincidence neutron rate from the induced fission events is counted. The current ^{3}He-based system suffers from poor signal to noise ratio, and long count times are required to achieve the desired measurement precision. As with the fast-neutron, scintillation detector alternative for the AWCC, a scintillation detector equivalent for the UNCL offers the possibility to not only meet but to improve on the existing ^{3}He system performance. Performance results for the demonstration system tested at the 2014 ISPRA workshop did not include sensitivity or measurement precision results, but based on the results from the PSD plastic scintillation system discussed above, we would expect that the current precision targets for the ^{3}He-based system could be met in 1/10th the time using the liquid scintillation-based system.

Although the liquid (or plastic) scintillation counters are very susceptible to gamma-ray interference, the fresh LEU fuels have a minimal gamma-ray emission rate which can be easily shielded to the point where the scintillators can remove the contribution to the neutron signal. This application is an example of a niche solution for the new generation of neutron detection systems. At present, it is unlikely that a fast neutron scintillation system could be fielded for routine MOX assays, however, they do offer a promising solution to an existing problem not well addressed by the current generation of ^{3}He systems.

6.3 LI/ZNS COATED PLASTIC SCINTILLATION DETECTOR FOR PASSIVE NEUTRON COINCIDENCE COUNTING

Symetrica Ltd., demonstrated a partially populated HLNCC-II equivalent neutron coincidence counter based on a Li/ZnS coated plastic scintillation detector (Fig. 16). The system is composed of four sides, each containing two detector modules referred to as blades [13]. Detection is accomplished through neutron capture in Li. The charged particles released in the capture reaction cause light to be emitted from the ZnS matrix. ZnS is a very bright scintillating material, but it is also a very opaque to its own light. The coating must therefore be very thin to avoid attenuation of the emitted light. By stacking thin light pipes (paddles or blades) coated with Li/ZnS, it is possible to achieve a reasonably efficient thermal neutron detector panel where the plastic blades serve dual purpose as a neutron moderator and a means to channel the emitted light to the compact PMTs (actually solid-state readout silicon photomultipliers) located at the end of the blade. The thinness of each blade reduces gamma-ray sensitivity of the scintillator, setting this detector type apart from the other scintillator technologies. Testing at the 2014 ISPRA workshop illustrated that the arrangement can accommodate gamma-ray exposure rates in excess of 10 mR/h (which is very impressive for a neutron-sensitive scintillation detector).

Note that unlike the other scintillation alternatives, the blades function as a thermal neutron detector. The system properties are similar to the ^{3}He counterpart in that die-away times are on the order of tens of microseconds rather than nanoseconds. The system tested was not fully populated (only 25% of the detector assemblies were installed) due to budgetary constraints. However, the reported performance of the partially populated system (~8.5% efficient with a die-away time of <40 μs) suggests that the fully populated system would exceed the performance of the ^{3}He based HLNCC-II.

Fig. 16. Photograph of the Symetrica HLNCC equivalent neutron coincidence counter (left) and an individual detector assembly or blade (right) [13].

While the system demonstrated at 2014 ISPRA Workshop is considered as a prototype, the system performance is impressive overall for a scintillation-based detector system. Symetrica has addressed scalability to more efficient detection systems through MCNP modeling. Design and fabrication of an advanced counting system such as the Epi-Thermal Neutron Multiplicity Counter (ENMC), based on these detector assemblies, appears promising. However, modeling of scintillation detectors is challenging, and additional measurements are needed. Additionally, long-term stability, reliability, and environmental testing are needed prior to field deployment of the Li/ZnS-based scintillation detector.

7. USE CASE SCENARIOS

The concept of Use Cases has been introduced to aid in the evaluation of potential detection technologies relative to the needs of an international safeguards program. Neutron coincidence and multiplicity assay represent the primary method for quantification of Pu in MOX and account for the lion's share of the ^{3}He consumed in safeguards measurements. Potential neutron detection technologies must be evaluated in the context of the safeguard measurement to be performed and must consider a broad range of factors beyond detection efficiency. It is beneficial to consider the operation of the fully assembled assay systems (not just the neutron detector) as they would be used in a typical production facility. By consideration of the existing installed base and projected future facility needs, it is possible to gage the impact of a given detector technology and its value as a replacement to the current ^{3}He biased safeguards infrastructure.

The Use Case Scenario examines the capability of the candidate neutron detection technology to meet the safeguards objective for the system in which it will be used, on the materials for which it will be used, operated in the manner in which it will be used in the environment in which it will be installed.

7.1 NEUTRON DETECTION VS. ASSAY PERFORMANCE GOALS

Safeguards measurement objectives are normally discussed in terms of precision and accuracy within a prescribed measurement time. The performance of the assay system is generally discussed in terms of the properties of the collection neutron detectors within the system taken as a whole and described by a detection efficiency, die-away time and FOM. These metrics are valuable in the evaluation of a detection technology but are not sufficient to determine the ability of the system to achieve the safeguards objectives for the measurement. In neutron coincidence or multiplicity assay, a number of additional factors impact the measurement performance such as sample mass, plutonium isotopics, physical and chemical form of the special nuclear materials, and the sample matrix.

Examples of the application-specific factors that need to be considered in the evaluation of a candidate detection technology include:

- **Dynamic Range vs. Pu Mass Range.** Pu and MOX samples occur in many forms such as clean product grade materials to dirty, high-burn scrap. The measurement system must accommodate the sample sizes, sample mass range, neutron emission rates, and decay heats. It is not uncommon for a given assay system to accommodate neutron emission rates spanning five orders of magnitude. Detectors, electronics, and analysis software must be designed to accommodate the extreme count-rate ranges while meeting the target performance goals.

- Form Factor vs. Facility Constraints:

  - **Footprint Limitations.** Floor space is limited within active nuclear facilities and comes at a high price. As a facility is designed, constructed, and commissioned modifications to accommodate NDA measurement systems becomes progressively more difficult such that the NDA system must physically fit within the existing or allocated foot print and is subject to floor loading constraints. Physically larger, heavier systems are generally less desirable than smaller ones.

  - Practicality: Routine system operations cannot be onerous to the facility. That is operations such as loading and unloading samples must be achievable within the normal operating constraints of the facility (e.g. attended systems are generally loaded manually without the need for steps, ladders or overly extending the operator).

  - Seismic Qualification: The assay system will be installed within an active nuclear facility and must meet the structural requirements of the facility. Larger, heavier systems introduce more risk in a seismic event. The assay system must be physically robust and allow for proper anchoring when required.

  - Maintenance Access: Regardless of the projected reliability of a new technology systems must be accessible for maintenance purposes. Additionally, the NDA system cannot permanently block access to process areas needing periodic maintenance. The systems must in general be easily moveable or removable.

- Environmental Response vs. Facility Constraints
  Safeguards measurements are often carried out under industrial rather than laboratory conditions. Environmental factors such as temperature, humidity, dust, and vibration are generally poorly controlled, yet the measurement system must provide reliable results across the full range of these conditions. The relevant environmental factors must be identified and the system performance characterized against these conditions.

- Gamma Sensitivity vs. Sample Exposure Rates
  Gamma-ray exposure rates from plutonium and MOX materials can be very high. Contact exposure rates in excess of 2 mSv/h are routine, and rates in excess of 10 mSv/h are not uncommon. Detector technologies must be able to accommodate the maximum expected exposure for that application whether through inherent insensitivity to gamma rays, distance, or shielding. It must be remembered that increases to assay system size and weight required for gamma-ray tolerance must be considered in the parallel with form factor and seismic constraints.

- Gamma and Neutron Sensitivity vs. Background Exposure Rates
  Gamma and neutron backgrounds within an operating nuclear facility will be elevated relative to normal background levels. The sensitivity of the assay system to external sources of neutron and

gamma-ray backgrounds must be considered. External shielding necessary for the proper operation of the system is considered to be an integral part of the assay system and impacts the footprint and seismic evaluations.

To illustrate the value of this approach, we have selected a single Use Case Scenario as an example. In this case a simple neutron coincidence counting application performed in attended mode. The example was chosen because the assay system ultimately fielded for the application was an HLNCC-II of the same model used as the reference counter in the workshop discussed above.

7.2 PLUTONIUM INVENTORY MONITORING

The plutonium inventory monitoring systems provide quantitative mass assay of Pu-oxide and MOX containers of product and clean scrap materials. These are generally modest performance neutron coincidence systems with detection efficiencies of less than 25% and die-away times of less than 50 μs (typically $2 < \mathrm{FOM} < 3.5$) constructed for use in either attended or unattended monitoring and are used to verify the declared mass of plutonium within an item. Because of the large number of these systems (>100 in current use), this class, and the relative importance of the measurements, these counters represents an important category of safeguards systems that must be supported into the future. These counters individually require only a relatively modest volume of ^{3}He (20 to 100 L). Collectively they represent a total safeguards investment of several thousand liters. Three different types of coincidence counters that are considered plutonium inventory monitors are shown in Fig. 17.

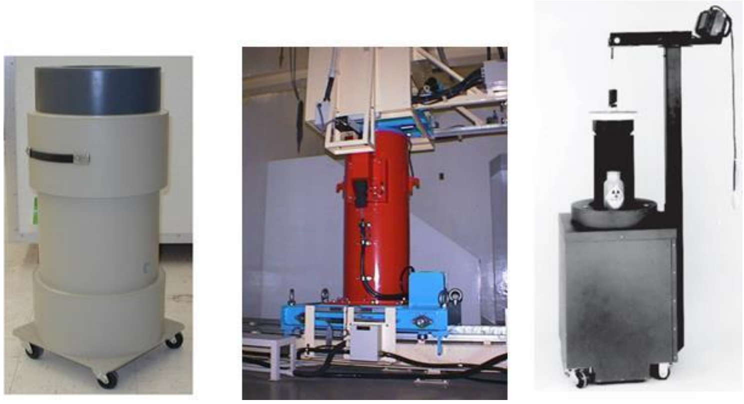


Fig. 17. Examples of Plutonium Inventory Monitoring Systems (from left to right the HLNCC-II [6], PCAS [14], and Flat Square [15] Counters).

As a typical Use Case for a plutonium inventory monitor, we consider the following real world example:

Measurement of product grade MOX containers for material balance within a storage vault.

The materials and performance goals in this example are typical of a MOX fuel manufacturing facility. Definition of the required assay system properties must consider a large number of factors. These factors are summarized as follows:

Sample Description:

- Material Forms: Pellets, Product Powders, Clean Scrap
  – Scrap: similar to product materials but may contain small impurities
  – Chemical form of each material type is well characterized and known
  – Pu isotopic abundances for each sample are available and well known

- Mass Range: 50–1200 g Pu in MOX
- $^{240}\mathrm{Pu}_{\mathrm{eff}}$/g ~0.36; ^{241}Am ~2%
- Container Sizes: 10–13 × <25 cm (OD × H)

Measurement Description:

Verification of operator plutonium mass declarations by neutron coincidence assay combined with declared isotopic data for each container. The agreed upon assay performance requirements for the measurement were typical of those encountered in MOX production facilities.

- Goal Measurement Performance:
  - $\sigma_{\mathrm{systematic}}$: <0.5%;
  - $\sigma_{\mathrm{random:}}$ <1% product, 2% scrap

- Measurement time: 1800 s

Complicating Factors:

- System storage location: Outside vault
  - Required Portable System (two-man carry, fit through standard door)

- Assay location: Inside vault
  - Elevated n/g backgrounds, nearly isotropic in distribution

Environmental Conditions:

- Nominally Environmentally Controlled: ΔT ±5°C, Humidity <90%

7.2.1 System Selection

In selecting the appropriate neutron assay system, the first step is to examine the various assay error contributions relative to the target performance goals. The well-characterized materials allow the use of known alpha, neutron coincidence counting analysis where we assume two unknowns in the neutron assay, the $^{240}\mathrm{Pu}_{\mathrm{effective}}$ mass, and the sample multiplication (i.e., multiplicity analysis is not required). We <u>assume</u> that the uncertainties due to the chemical form of the plutonium are negligible, as are the error contributions from the declared isotopic values. The dominant sources of error under such conditions are:

- Spatial uniformity of the neutron counter response:
  The neutron detection performance of the counter should not vary significantly (relative to the target performance values) across the container volume. The error contribution due to variations in material fill height and radial extent should not have a significant impact on the measurement result.

- Measurement precision:
  The measurement precision for an assay system is determined by the material characteristics, neutron detection efficiency and die-away time, background conditions, and the assay time allotted. The measurement precision is generally the largest contributor to the total measurement error.

- Linearity of the neutron detection performance as a function of neutron energy:

- An ideal counter will have a flat efficiency profile as a function of neutron energy. However, in this example, the neutron energy variation from item to item is small such that a linear energy response profile is not a key requirement. It is sufficient to require that the assay cavity be lined with cadmium to eliminate re-entrant thermalized neutrons from inducing fission within the sample.

- Calibration errors:
  Calibration error is generally limited by the quantity and quality of the available calibration standards. These standards must be representative of the items to be verified. In general, the calibration error will be small relative to the measurement precision and spatial uniformity effects.

In consideration of the systematic errors, for this example the only significant error term under the control of the system designer is the spatial uniformity of the neutron counter response. For a traditional ^{3}He proportional tube-based system, spatial uniformity is improved by increasing the assay cavity size within the limits of practicality and cost. The appropriate cavity size is determined either by examining measurement performance data of similar systems or by use of Monte Carlo modeling tools. However, as a general rule of thumb, for ^{3}He-based systems, the assay cavity height should be at least equal to the height plus diameter of the container, and the inner diameter of the cavity should be 1.5 to 2 times the diameter of the container.

The random uncertainty contribution is addressed by selection of an assay system with a sufficiently high FOM. In a normal background environment, an assay system with a FOM of ~1 would be sufficient to meet the performance requirement for this example. However, the elevated neutron background levels encountered in the measurement area drive up the required FOM. Modeling suggested that a figure of merit of greater than two was required. Systems providing neutron detection efficiency of 15% or more with a characteristic die-away time of 50 µs or less would meet this requirement.

The mass range, chemical form, and isotopic abundances allow us to estimate the maximum neutron emission and gamma-ray exposure rates. The detectors, acquisition electronics, and analysis computer must be able to accommodate the maximum expected exposure rates. The neutron emission rates and exposure rates are estimated from the sample mass range and chemical form:

- Expected neutron emission rate: <1.5E6 n/s
  The hypothetical 15% efficient neutron assay system would need to tolerate neutron count rates of 2.3E5 cps to accommodate the expected plutonium mass range.

- Gamma-ray exposure rates: <5 mSv/h

- The neutron assay system must be able to tolerate a significant gamma-ray exposure without misclassifying gamma rays as neutrons. The typical ^{3}He based coincidence counter can be operated at exposures between 5 mSv/h and 10 mSv/h, so the expected exposure rates would not have been a limiting factor.

The final requirement that the system be man-carried into and out of the assay area for use limits the physical size and weight of the system. The requirement limited total system mass to less than 50 kg and did not allow significant external shielding. (The measured neutron background count rate with the selected system was 1000 cps).

Rather than build a custom assay system to meet these needs, a COTS system (the HLNCC-II) was selected for use in this application. The HLNCC-II provides 18% neutron detection efficiency, a 50 µs die-away time meeting the measurement precision goals, the assay cavity 40 cm × 19 cm (H × ID) accommodates the expected range of container sizes. The system had been demonstrated to handle both the neutron counting rates from the sample and the elevated background conditions. Testing of the

HLNCC-II at the MOX facility demonstrated performances consistent with the measurement objectives [3].

Table 5. Average Biases and Standard Deviation by sample types.

Sample Type	Alpha Weight	Average Bias	Standard Deviation
Pellets	1.0	1.1%	0.8%
Scrap Powder	1.1	1.3%	2.1%
Feed Powder	1.0	0.0%	0.9%
Product Powder	1.0	0.6%	0.8%
Recycled Powder	1.0	-0.6%	0.5%
Scrap Powder	1.0	-2.7%	1.2%
All samples		**0.2%**	**1.6%**

The Use Case scenario allowed identification of an appropriate assay technology that would meet the target performance values for the specific measurement application. Examination of new detection technologies across a range of potential Use Cases facilitates assessment of the impact of the candidate technology against the needs for replacement neutron technologies in an international safeguards program taken as a whole. The Use Case approach allows the determination of whether the candidate technology has broad application or if it is suitable only for niche applications.

8. ALTERNATE METHODOLOGIES

The ^{3}He shortage has provided impetus to re-examine not only replacement neutron detection technologies but also to re-examine the general measurement methodology used for the safeguards measurements. These alternatives can either reduce the quantity of ^{3}He required to achieve a desired measurement outcome or provide a technique that does not require ^{3}He in the first place.

Two techniques under study at ORNL, the Direct Multiplication Measurement [16] and the Cadmium Subtraction Ratio Method [17], seek to provide alternative methods for determination of the sample's multiplication as a means to address multiplicity counting in an era where epithermal multiplicity counters [18] are cost prohibitive. Providing a secondary means for determination of multiplication allows the application of multiplicity analysis using modest neutron coincidence counting systems (e.g., boron-lined proportional tube-based systems) currently available as ^{3}He alternatives. Both of these techniques are in the development stage awaiting field trials.

Since the neutron coincidence counters were first introduced to international safeguards, they have been widely perceived as the de facto low-cost, most accurate solution due to the deep penetration of fast neutrons through most materials and the availability and low cost of ^{3}He. However, other neutron based technologies such as the ^{252}Cf shuffler that were previously considered to be not cost effective are now less expensive due to the relatively small amount of ^{3}He required for fabrication, and they could also be fabricated using currently available ^{3}He alternative detectors [19]. Other non-neutron-based, accurate measurement technologies such as the Tomographic Gamma Scanner have been introduced for scrap and waste assay applications and are now commercially available [20, 21]. These alternative technologies have long track records of successful operation but have not been adopted for international safeguards applications and so would incur cost to integrate into the existing safeguards infrastructure. They are sufficiently close in technology to other accepted technologies that this cost would be relatively modest. However, these techniques would not be general purpose solutions and would be considered niche solutions.

9. CONCLUSIONS

Numerous alternative technologies exist as potential replacements for ^{3}He proportional counters that may be suitable for neutron detection systems in international safeguards applications. Mature technologies such as the BF_3 or boron-lined proportional counters currently satisfy the measurement performance requirements of many of these demands. However, concerns such as safety, weight, footprint, or cost may cause the rejection of these technologies. An essential first step in assessing the suitability of any neutron detection technology is the determination of its fundamental properties. To fully demonstrate the applicability of the technology to a given safeguards application, it must be evaluated within the context of a complete assay system. This evaluation must include all relevant parameters impacting measurement performance, operability, maintainability, reliability, availability and life cycle cost. Furthermore, the evaluation should consider the potential applicability of the technology across a wide range of measurement solutions to minimize proliferation of niche solutions that are ultimately unsustainable.

More recently developed technologies such as the boron coated straw detectors and the Li/ZnS scintillation detectors are promising technologies that can potentially service a much larger fraction of the safeguards application space. The straw detectors represent a near drop in replacement to the ^{3}He proportional tube-based systems with the exception of the very high performance multiplicity and epithermal multiplicity counting systems. Here the limiting factor is expected to be the large number of individual detector tubes, connectors and associated wiring and electronics, for the largest systems the straws may become impractical. The Symetrica Li/ZnS-based thermal neutron scintillation detector (blade) has the potential to replace the ^{3}He based systems with the potential limiting factor of gamma-ray sensitivity (potentially requiring massive amounts of internal shielding). The high intrinsic efficiency and low die-away times (low for a thermal neutron counting system) can in principle provide sufficient performance to replace the flagship ENMC [18] system. For both of these very promising technologies, environmental and stability testing is required as well as detailed reliability and MTBF studies. However, these two technologies represent the most likely successors to the ^{3}He proportional tube.

The drive to develop a replacement to the ^{3}He tube has re-opened the dialog regarding what constitutes an acceptable safeguards solution to the measurement of uranium- and plutonium-bearing items. Techniques formerly considered off limits due to infrastructure constraints, such as the liquid scintillation detector, have been embraced by the IAEA. These studies have resulted in the potential introduction of niche measurement systems such as the liquid scintillation detector equivalent of the UNCL counter for the assay of LEU fuel assemblies, techniques that can offer dramatic improvements over the existing standard solution and potentially solve the throughput limitation that has plagued the safeguards measurement of the fuel assemblies.

Finally, although several very likely candidate technologies have been identified, it is necessary to remember that these candidates require formal, long-term vetting prior to adoption as the successor technology to the ^{3}He proportional tube applied to international safeguards.

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