A REVIEW OF NEUTRON DETECTION TECHNOLOGY ALTERNATIVES TO HELIUM-3 FOR SAFEGUARDS APPLICATIONS

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ABSTRACT

Existing neutron based systems used for the detection of Special Nuclear Material (SNM) in Safeguards application are typically based on ^{3}He proportional counters requiring relatively large volumes of the gas (up to several thousand liters per system). Because of the severe ^{3}He shortage, a replacement technology for neutron detection is required in the near future. There are competing demands for ^{3}He gas from other industries, as well as medical, security and research fields. It is estimated that the total ^{3}He annual demand is now approximately 65,000 liters compared to total worldwide annual supply of 15,000 liters. As a result of this shortage there is a major drive for alternative detection technologies to be developed for wide scale deployment as a long term replacement for ^{3}He.

There are three basic requirements for neutron detectors for safeguards applications involving detection and measurement of SNM: 1) high absolute detection efficiency, 2) low intrinsic gamma ray sensitivity of the detector and 3) maintaining neutron detection efficiency when simultaneously exposed to high gamma ray exposure rate. In addition, safeguards sensors may need to meet specific requirements of the application area including 1) security and integrity of data collection; 2) backward compatibility with existing systems and data collection methods; 3) long term electronic stability in harsh environments; 4) low or zero maintenance; 5) operability in unattended mode; 6) easy for field inspectors to operate with minimal training/specialization; and 7) applicability in a wide range of applications to allow agencies to standardize their designs.

Several alternative detectors are currently available as near term replacement for ^{3}He that could potentially meet the above requirements. These include Boron trifluoride (BF_3) filled proportional counters, ^{10}B lined proportional counters, ^{10}B doped plastic scintillators, ^{6}Li loaded optical fiber scintillators and ^{6}LiF/ZnS(Ag) screens with a bulk, wavelength-shifting light-guide. The capabilities of these alternatives is reviewed with respect to the long term requirements for safeguards systems such as drum and crate assay system, slab counters, Pu/MOX canister verification and well counters. Performance is compared to the ^{3}He baseline technology with any potential technical capability improvements that each alternative could offer also being considered.

INTRODUCTION

Innovative safeguards technologies are required to enhance future capabilities in verification of nuclear materials and activities [1]. New technologies of interest include the capability to detect diversion of nuclear material from declared facilities; to detect undeclared nuclear material and activities; and to verify compliance with arms control treaties and agreements related to the control, production, or processing of nuclear material [2, 3].

Existing neutron based systems for the detection of Special Nuclear Material (SNM) utilize ^{3}He proportional counters. Because of the current ^{3}He shortage, there is growing need to develop a replacement or alternative technology for neutron detection [7]. 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. It is estimated that the total ^{3}He annual demand is ~65,000 liters, while total worldwide annual supply is ~15,000 liters (the gas accumulates from the decay of tritium). The US DOE plans to make available ~ 8,000 liters each year on a limited basis [10].

Table 1 illustrates the volume of ^{3}He required to furnish a typical range of safeguard assay system units. Meeting future safeguards needs with exclusive reliance on ^{3}He would put significant strain on this limited resource.

Table 1. ^{3}He Gas Volume Required for Proposed Systems per Unit

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

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.

TECHNICAL REQUIREMENTS

There are three basic requirements for neutron detectors relevant to safeguards applications, primarily relating to detection and measurement of SNM: 1) high absolute detection efficiency, 2) low intrinsic gamma ray sensitivity of the detector and 3) detector must retain the same neutron detection efficiency when simultaneously exposed to high gamma ray exposure rate.

In addition, safeguards detectors need to meet specific requirements of the application area, this can include 1) security and integrity of data collection e.g. seals, immunity to 'spoofing'; 2) backward compatibility with existing systems and data collection methods such as universal NDA data acquisition modules; 3) long term electronic stability in harsh environments; 4) low or zero maintenance 5) operability in unattended mode; 6) easy for field inspectors to operate with minimal training/specialization; and 7) applicability in a wide range of applications to allow agencies to standardize their equipment designs.

RELEVANCE TO SAFEGUARDS

Accurate measurement and verification of nuclear materials contributes to global nuclear security. Some of the most relevant neutron based techniques that are used for safeguards include passive neutron assay (total neutron, coincidence and multiplicity methods) and active neutron (e.g. the "Active Well", differential die-away and californium shuffler techniques).

Generally speaking, the passive mode is usually preferred in safeguards because the detection limits are normally of the order of gram levels of plutonium. Active assay systems tend to be more frequently encountered in waste sentencing instruments which require very low detection limits (milligram levels). Passive techniques are therefore expected to dominate the future ^{3}He demand. Systems that require high efficiency in coincidence/multiplicity mode are the most urgent development need, because no existing neutron detector design (other than hazardous BF_3) can provide the necessary detection efficiency in a close-packed geometrical arrangement.

Methods such as multiplicity counting enable highly accurate accountancy measurements to be performed (e.g. for physical inventory verification at processing facilities). To achieve high levels of precision, the instruments must achieve a very high intrinsic efficiency and thus require large volumes of ^{3}He. This situation calls for research and development into alternative detectors that can be used as a 'like for like' (or superior) replacement.

The targeted system/application areas for evaluation of alternative detectors include:

- In-situ glovebox assay (slab counters), used for hold-up/accountancy.
- Pu canister (scrap) verification assay, often used in MOX facilities.
- Waste drum and crate assay, such as systems provided at Rokassho Reprocessing Plant in Japan [4].

The present shortage of ^{3}He has the potential to have a significant impact on the capability of safeguards agencies and as result there are several on-going multi-national research programs focused on this issue [11, 12]. Research efforts should be prioritized towards currently available technologies that are potentially capable of meeting all of the safeguards instrumentation performance requirements.

ALTERNATIVE NEUTRON DETECTORS

The main advantages of ^{3}He detectors for neutron detection are the very high efficiency (resulting from its high thermal neutron absorption cross section) and good discrimination of the gamma signal. Other benefits include inflammability, non-toxicity, physical robustness and long operational lifetime. Some potential alternative detectors either available or under development are as follows:

Boron trifluoride (BF_3) filled proportional counters. These counters are similar to ^{3}He but with lower efficiency. BF_3 filled detectors are direct replacements and were commonly found in non-destructive assay 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 this technology being largely discontinued. 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.

^{10}B lined proportional counters. As with BF_3, these 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 safeguards applications is that they are a factor of approximately 7 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 '4 pi' 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.

^{10}B lined 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' [14] or interior 'baffles'. Another approach is to pack multiple coated "straw" cathodes [16] 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.

Semiconductors. Despite on-going research in improved materials, these crystals have limitations in size and subsequently low overall detection efficiency. As a result the application areas are usually niche such as in high neutron flux monitoring.

^{10}B doped plastic scintillators. This detector comprises a plastic scintillator material embedded with particles of boron-containing compounds. They have high efficiency, are economical to mass produce and have a very fast pulse decay time which provides higher counting rate capability. Their high gamma sensitivity is the major issue that needs addressing. They are generally limited for use in detecting fast neutrons, but in some cases are suitable for thermal neutrons and their fast response time provides potential advantages in pulsed neutron active neutron applications.

^{6}Li coated argon filled detectors. Efficiency and gamma sensitivity issues need to be addressed, but this is potentially a very cost effective alternative in large area applications [17]. Essentially an ionization chamber with a thin foil of lithium lining a high-density polyethylene cavity filled with argon. Development efforts are on-going, targeted at creating efficiencies similar to conventional ^{3}He tubes, at a much reduced cost.

Liquid organic scintillators. Traditionally used for fast neutron applications, examples include lithium solutions. Typical usage scenarios include large volume applications where pulse shape discrimination capability is needed e.g. for suppression of the gamma response. Concerns over wide scale safeguards deployment include low thermal neutron efficiency, long term safety and radiation damage effects.

^{6}Li-loaded glass fibers. These 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.

Scintillator loaded plastic fibers. Non-scintillating fibers are 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.

Scintillating crystal composed of 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 [15]. Combined with their high light yield and high gamma energy equivalent of the neutron peak (>3MeV) this technology is very promising for safeguards 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.

^{6}LiF/ZnS(Ag) screen with a bulk, wavelength-shifting light-guide. 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 [9], 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 (e.g. TTL output). Electronics can also be adapted to provide a gamma ray count rate as a separate data channel.

PERFORMANCE METRICS

Absolute efficiency is the most important performance metric for passive neutron counting. A typical passive neutron safeguards counter varies in efficiency from 4% (large area slab counter for glovebox assay) to 55% (can contents verification). A typical scenario is a counter that comprises 6 'walls' surrounding the item of interest i.e. in a '4 pi' geometry.

In order to investigate suitability of the various alternatives, the following arrangement should be considered as a standard test platform:

- A slab shall be constructed with dimensions 40 cm x 100 cm containing detection and moderation materials.
- The efficiency of an individual slab shall be at least 1.6% for a ^{252}Cf neutron source at a distance of 50 cm. This would achieve the necessary efficiency for most safeguards applications including coincidence/multiplicity counting. For example, a cubic chamber that has an internal volume of 1m^3 with 6 slabs per side (arranged '3 wide' and '2 deep' in all walls, roof and floor) as illustrated in Figure 2, would achieve an efficiency that exceeds 30% (accounting for the fact that slabs in the outer layer will be less efficient compared to the inner layer).
- In order to achieve low detection limit (~ 0.1 -1.0 g Pu typical of a safeguards application), the slab must achieve very low counts in a 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 150,000 n/s (equivalent to 500 grams of commercial grade PuO2) and must also provide accurate measurement of SNM in the co-presence of the following gamma radiation fields:

    - Unshielded gamma rays produced from 500 grams of commercial grade PuO_2 (including associated gamma emissions from the ^{241}Am decay product of ^{241}Pu) placed at a distance of 50 cm from the detector slab,
    - Dose rate field of 200 mR/hr (2 mSv/hr) produced from (a) ^{137}Cs, (b) ^{60}Co.

- In a 10 mR/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.

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

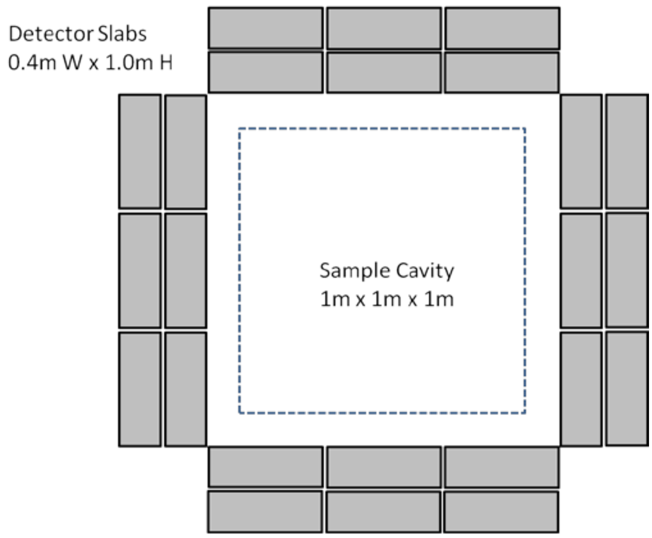

Figure 1. Schematic of a 'typical' 4-pi geometry neutron assay safeguards application

The above performance would be the nominal equivalent of a slab monitor of equal dimension comprising 8 ^{3}He detectors with a 4 atmosphere fill pressure, 2.54 cm diameter, 90 cm active length. This detector array may be considered the baseline currently used in Safeguards.

Additional capabilities to be investigated include:

- Potential application in active interrogation mode (passive mode is more commonly used in Safeguards application, but active mode can in some cases be a requirement where a direct measure of fissile material is required, for example if the passive signal is masked by unknown levels of interference e.g. from ^{244}Cm).
- Suitability of matrix correction techniques. Determine applicability of existing and new matrix correction methods. The primary methods to examine are referred to as “Ring-Ratio” and “Add-A-Source” [5]. In the Ring Ratio method, a matrix correction factor is defined as a function of the ratio of neutron count rates (emitted from the sample) in two or more rings of detectors. The outer detectors are more highly moderated than the inner ring and therefore will have a different response depending on the degree of thermalization that has occurred within the sample. In the Add-A-Source method an external ^{252}Cf source is used to determine a correction factor.
- Chamber die-away time is of importance when designing a passive neutron coincidence / multiplicity counting system. Fast neutrons must be moderated and reduced to thermal energy in order to be captured for counting. In a traditional system, using ^{3}He proportional counters, this is achieved by embedding tubes in a moderating material. As a result, chamber die-away times are usually 20-55 μs. It is possible to achieve fast die-away times as low as 6 μs with a well counter lined with detectors made from ^{6}LiF/ZnS(Ag) scintillator mixed with hydrogenous material [18]. Faster die-away times could allow improved precision for high (alpha,n) standards such as materials with enhanced ^{241}Am.
- Addressing software/electronics/mechanical upgrades required for existing system designs. Suitability for use with existing software such as INCC should be evaluated.
- Practical considerations such as large scale production of the detector modules, safety and durability, transportation, testing at the target nuclear facility.

In addition to technical performance, the lifetime cost, overall size, availability and scalability needs to be addressed in comparison to the baseline technology. New capabilities of the detector such as spectroscopic capability, improvements to matrix correction and dual gamma/neutron data acquisition are additional factors to consider in future research.

To evaluate the performance of the new sensor compared to current baseline, three primary criteria shall be considered: 1) absolute neutron detection efficiency in (a) singles mode, (b) coincidence mode, 2) intrinsic efficiency of gamma rays detected as neutrons; and 3) Gamma Absolute Rejection Ratio [6] in the presence of neutrons (GARRn).

Additional technical criteria to consider are: efficiency profile across the detector’s surface, timing characteristics; count rate dynamic range; susceptibility to electromagnetic interference; and mechanical robustness/reliability (mean time between failures).

CONCLUSIONS

The majority of existing neutron detection systems used for nuclear safeguards and security are based on ^{3}He. In recent times, the ^{3}He market has encountered significant supply and demand problems. This has resulted in order-of-magnitude price increases in commercial stocks and restrictions in supply from government sources. As a consequence, many manufacturers are currently unable to provide long term assurance on detector supply that would be necessary for long term project planning on safeguards related instrumentation projects.

Wide-scale application of alternative neutron detector technologies in Safeguards application has the potential to improve the capabilities of global nuclear security agencies without requiring depletion of the US government's limited supply of ^{3}He.

Table 2 summarizes the pros and cons of available neutron detectors and evaluates potential safeguards fields for which each type of detector has the capability to be currently deployed.

New materials such as CLYC scintillators offer intriguing prospects; in particular for dual gamma/neutron counting applications. However, it is unlikely that a single neutron detector will be developed in the next 5-10 years that exceeds the performance, cost and reliability of ^{3}He for all safeguards application. Of the currently available neutron detection technologies, BF_3-filled proportional detectors, ^{10}B-lined tubes, ^{6}LiF/ZnS(Ag) and ^{6}Li scintillating glass fiber can offer a range of capabilities as immediate replacements for ^{3}He detectors in many applications.

Table 2. Comparison of Available Neutron Detection Options (relative to ^{3}He)

Detector	Pros	Cons	Possible safeguards application
BF_3 tubes	Cheap (~$1k/tube). Mature technology. Plug-in replacement of ^{3}He. Low gamma sensitivity.	Lower neutron efficiency. Toxic, corrosive gas. Long term degradation issues. Transportation subject to strict regulations, prohibited in some locations.	Medium Efficiency Neutron Counting. Coincidence counting
^{10}B lined tubes	No toxicity or degradation. Plug-in replacement of ^{3}He. Can be set for very low gamma sensitivity.	Intrinsically poor counting efficiency (can be improved by tubelets, baffles etc.). Expensive (~$10k- 20k/tube).	Low-Medium Efficiency Neutron Counting. High gamma flux. RPMs
Scintillating glass fiber	Formed to various shapes. Scalable to large areas. Very high neutron efficiency. Simultaneous neutron/gamma measurement.	Very high gamma sensitivity. Relatively high cost for large area applications.	Low maintenance neutron verification in low gamma fields.
Non scintillating plastic fiber	High neutron efficiency. Good neutron-gamma separation (scintillator is very thin).	Cost is very high	Low maintenance neutron verification in high gamma fields.
B loaded plastic scintillator	Readily available. Good for fast neutron detection and active neutron applications.	Neutron detecting area is limited by the need to preserve optical clarity in the scintillating region. High gamma sensitivity.	Fast neutron detection. Active neutron.
$^{6}LiF/ZnS(Ag)$ with wavelength shifting light guide.	Very high neutron efficiency The scintillation is bright. Can discriminate neutron and gamma events in software/electronics. Materials can be chosen with different relative gamma and neutron sensitivity Can perform neutron spectroscopy Simultaneous neutron/gamma measurement. Can achieve shorter chamber die-away time then He-3 based chambers.	Not yet mass-manufactured. Neutron-gamma ray signal separation limits application in very high gamma flux.	Low-High efficiency neutron counting in low gamma fields. Neutron spectroscopy. Coincidence / multiplicity counting of high (alpha,n) materials. Neutron imaging applications (due to malleability and intrinsic position sensitivity).
CLYC scintillator.	Pulse shape and high gamma effective peak (>3 MeV) benefit gamma discrimination Simultaneous neutron/gamma measurement with medium resolution gamma spectroscopy.	Currently in development. Neutron-gamma ray signal separation may limit applications in very high gamma flux.	Further evaluation required.

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