Estimation of Performance of an Active Well Coincidence Counter Equipped with Boron-Coated Straw Neutron Detectors – 13401

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ABSTRACT

He-3, a very rare isotope of natural helium gas, has ideal properties for the detection of thermal neutrons. As such it has become the standard material for neutron detectors and sees ubiquitous use within many radiometric applications that require neutron sensitivity. Until recently, there has been a fairly abundant supply of He-3. However, with the reduction in nuclear weapons, production of tritium ceased decades ago and the stockpile has largely decayed away, reducing the available He-3 supply to a small fraction of that needed for neutron detection. A suitable and rapidly-deployable replacement technology for neutron detectors must be found. Many potential replacement technologies are under active investigation and development. One broad class of technologies utilizes B-10 as a neutron capture medium in coatings on the internal surfaces of proportional detectors. A particular implementation of this sort of technology is the boron-coated "straw" (BCS) detectors under development by Proportional Technologies, Inc. (PTi). This technology employs a coating of B-10 enriched boron carbide (B_4C) on the inside of narrow tubes, roughly 4 mm in diameter. A neutron counter (e.g. a slab, a well counter, or a large assay counter designed to accommodate 200 liter drums) could be constructed by distributing these narrow tubes throughout the polyethylene body of the counter. One type of neutron counter that is of particular importance to safeguards applications is the Active Well Coincidence Counter (AWCC), which is a Los Alamos design that traditionally employs 42 He-3 detectors. This is a very flexible design which can accurately assay small samples of uranium- and plutonium-bearing materials. Utilizing the MCNPX code and benchmarking against measurements where possible, the standard AWCC has been redesigned to utilize the BCS technology. Particular aspects of the counter performance include the single-neutron ("singles") detection efficiency and the time constant for the decrease in neutron population in the counter following a fission event (a.k.a. the die-away time). Results of the modeling and optimization are presented.

INTRODUCTION

He-3 is a very rare isotope of natural helium gas which has, over the past few decades, become the standard material for neutron detectors. It is chemically inert and nontoxic; detectors that utilize He-3 have a very low sensitivity to gamma rays; and, up until relatively recently, it was inexpensive. The gas is a decay product from tritium, and was thus maintained in ready supply for decades as tritium was continually being produced to support nuclear weapons stockpiles.

However, with the reduction in nuclear weapons, production of tritium ceased decades ago and the stockpile of He-3 has largely decayed away, reducing the available supply to a small fraction of that needed for neutron detection applications.

With its increasing scarcity, the cost of He-3 has increased drastically. At the time of this writing, the per-liter price of He-3 is about 35 to 40 times higher than it was a decade ago. This has driven up the cost of neutron counting instruments used for waste assay and materials safeguards to the point where a substantial majority of the cost of such instruments is in the He-3 gas used for the detectors.

Alternative strategies are needed. There are some applications for which there isn't likely to be an acceptable replacement, in which case a sensible strategy seems to be to limit the use of the remaining He-3 to those applications. Many technologies are under active investigation and development in a wide-spread effort to find a potential replacement that is rapidly deployable and has acceptable performance in neutron-detection applications. The majority of the technologies currently under investigation utilize either Li-6 or B-10 as neutron capture targets coupled with some means of detecting the reaction products from the neutron capture events and converting them to a measurable electrical signal. One broad class of technologies utilizes B-10 as a neutron capture medium in coatings on the internal surfaces of proportional detectors. A particular implementation of this sort of technologies, Inc. (PTi) [1]. This technology employs a coating of B-10 enriched boron carbide (B₄C) on the inside of narrow tubes, roughly 4 mm in diameter. A neutron counter (e.g. a slab, a well counter, or a large assay counter designed to accommodate 200 liter drums) could be constructed by distributing these narrow tubes throughout the polyethylene body of the counter.

The Active Well Coincidence Counter

One type of neutron counter that is of particular importance to safeguards applications is the Active Well Coincidence Counter (AWCC), which is a Los Alamos design that traditionally employs 42 He-3 detectors. A standard version of this counter is manufactured by Canberra Industries via a technology transfer from Los Alamos. This counter has a very flexible design which can accurately assay small samples of uranium- and plutonium-bearing materials [2,3].

The counter consists of a cylindrical body, roughly 50 cm in diameter and 70 cm tall (19" x 28") made of high density polyethylene. Embedded in the polyethylene body are 42 He-3 tubes arranged in two concentric rings of 21 tubes each. Each tube is 2.54 cm in diameter with roughly 51 cm active length (1" x 20") and filled with He-3 at 4 atmospheres absolute pressure. The inside of the counter is equipped with two AmLi sources which provide interrogating flux to induce fissions in samples containing substantial amounts of U-235 or U-233. The inside of the counter is

also equipped with two polyethylene rings ("donuts"), one nickel reflector ring, and a complete shell of cadmium lining over the entire inner surface of the counter. These various inserts serve to optimize the sensitivity of the counter to the induced fission neutrons from the uranium in the sample. These inserts as well as the AmLi interrogation sources are removable to provide multiple modes of operation (active versus passive, fast neutrons versus slow neutrons) and to accommodate different sample sizes. Figure 1 below shows a photograph and cutaway drawings of the standard Canberra Industries JCC-51 AWCC.



Figure 1. Photograph and cutaway drawings of Canberra Industries JCC-51 AWCC.

The AWCC is most commonly operated in three configurations:

- "Active, fast" AmLi sources, both polyethylene donuts, nickel ring, and cadmium inserts are all present. The assay chamber is roughly 23 cm in diameter and 21 cm high (9" x 8"). The response is optimized for the higher energy induced fission neutrons. This is used for samples with a high uranium content.
- "Active, thermal" AmLi sources are present, but the cadmium liner inserts are removed. The polyethylene donuts, nickel ring, and various spacers in the end plugs can be removed to accommodate larger samples. The assay chamber can be as small as in the previous description, or as large as 23 cm in diameter and 35 cm high (9" x 14"). This is used for samples with lower uranium content.
- "Passive, thermal" the same configuration (and possible assay chamber sizes) as above,

but the AmLi sources are also removed. This is used for samples containing plutonium, where the spontaneous fission signature doesn't require the active interrogation.

DESCRIPTION OF MODELING EFFORT

To assess the feasibility of utilizing the PTi BCS technology as a He-3 replacement in the AWCC design, a campaign of design and modeling using the monte carlo physics code MCNPX [4] was conducted.

Benchmarking the modeling technique

As a first step, the neutron response was measured for a small BCS-equipped slab fabricated by PTi. This slab is a demonstration unit intentionally made to be small for easy portability and demonstration purposes. The slab is roughly 15 cm wide, 13 cm deep, and 46 cm tall (6" x 5" x 18"). It is constructed as a layered polyethylene slab embedded with 196 boron straws. The B₄C thickness is 0.85 μ m. The measurement was performed with the slab suspended 100 cm above a flat concrete floor with a calibrated Cf-252 fission neutron source located 30 cm from the center of a broad face of the slab. The measured singles neutron absolute efficiency was 0.00323 with roughly 1% relative uncertainty due to counting statistics.

Note that there are three physical process that must take place from the time a thermal neutron strikes the boron coating on the inside of a straw and the recording of an actual neutron event (i.e. a "count") in the acquisition electronics. Discussing these three events illustrates key aspects of optimizing and modeling these sorts of counters. The three processes are:

- The thermal neutron must be captured in the boron layer on the inside of the straw. The likelihood of this happening depends on the B-10 neutron capture cross section, the degree to which the boron in the B₄C layer is enriched in B-10, and the thickness of the layer itself. From this standpoint alone, a thicker layer is better it provides more (a higher spatial density) of B-10 capture targets that could potentially yield a signal. In a broader sense, this likelihood also depends on how much B₄C is concentrated in a given volume of the slab. This is the primary intent behind the straw technology many small-radius tubes packed together provide a high volume concentration of B-10 capture targets.
- The reaction products from the capture reaction (B-10 + n → He-4 + Li-7) must then straggle their way out of the B₄C layer, into the gas-filled center of the straw volume, and deposit energy into the gas. From this standpoint alone, a thicker layer is *worse* the thicker the layer, the less likely the helium or lithium ions are to straggle into the gas volume and the less energy they're likely to have if they do. Given this trade-off, there is an optimum thickness for the B₄C layer which balances the competing effects of "more capture targets" versus "shallower straggling thickness." It is important to note that this

straggling effect will entail some loss – not every capture event in the B_4C layer will yield ionization events in the gas.

 The moving helium or lithium ions strike the gas and create ionization pairs which are then multiplied by the applied high voltage, ultimately yielding a shower of electrons which create a signal on the anode wire down the axis of the straw. The resulting electrical signal must be large enough to clear a lower threshold in the amplification electronics. This threshold is necessary to reject spurious noise, gamma ray events, etc. in the signal chain. This, too, entails some loss – not every ionization event in the gas will yield a countable signal.

A model of the demonstration slab was developed using MCNPX. A common technique for modeling these sorts of detectors is to tally the neutron captures (the first of the three processes in the above list) with MCNPX's F4 tally, and then apply an empirical scaling factor to account for straggling and electronic losses (the second and third processes in the above list). For the models discussed here, the pulse height spectrum in the proportional gas was calculated directly by explicitly tracking and tallying the heavy-ion reaction products with a combined F6 / F8 / PHL tally structure. This allowed for explicit accounting for not only the capture in the B_4C layer, but also the losses due to straggling and the imposition of an electronic threshold. In this particular case, the electronic setup of the slab performed by PTi established an electronic threshold of roughly 70 keV. Using this as a threshold on the MCNPX-calculated pulse height spectrum for the simulated measurement yielded a calculated singles neutron absolute efficiency of 0.00333 with roughly 0.5% relative uncertainty due to the monte carlo statistics. This provides excellent agreement with the measurement, and bolsters the expectation that this modeling approach can be used to at least roughly design and optimize a counter configuration.

Modeling the AWCC

There were multiple initial objectives for redesigning the AWCC with boron straws. It was desirable to keep the internal assay cavity size and design the same as for a standard He-3 AWCC, and somewhat less so to keep the external footprint the same as well. It was also very desirable to achieve measurement performance as close as possible to that for a standard He-3 AWCC. The idea of "performance" was quantified by calculating the singles efficiency and the die-away time (DAT) for a pointlike Cf-252 source centered in the assay cavity. In the models, the efficiency was found using the same tally technique described above for the slab benchmark calculations. The die-away-time is the average lifetime for a neutron emitted inside the assay cavity – ultimately the neutron is either captured (e.g. in the B₄C lining or in the hydrogen in the polyethylene body) or it escapes from the counter body altogether. The DAT was estimated from the model by fitting a single decaying exponential (i.e. $\exp(-t/\tau)$) to the time histogram of the F4 capture tally in the B₄C lining. Also, to provide a single overall metric of performance, a commonly-used figure of merit

(FOM) [5,6] $-\varepsilon^2/\tau$ – was calculated, where ε is the singles efficiency and τ is the die-away-time.

The model was constructed by placing the boron straws in a hexagonal lattice with a 1 cm pitch. Experience at PTi has shown that this is a recommended rough value. Other than replacing the two rings of He-3 with a lattice of boron straws, the other features and dimensions of the AWCC – the internal dimensions, structure, and contents of the assay cavity, as well as the external footprint – were identical to that for a He-3 AWCC. When laid out this way, the model utilized 1466 straws. Figure 2 immediately below depicts a horizontal cross-section illustration of the counter body, assay cavity, and the layout of the straws.



Figure 2. Horizontal cross-section illustration of AWCC counter and layout of boron straws.

A key parameter in the design and optimization of the counter configuration is the thickness of the B_4C layer. To investigate the effects of varying the layer thickness, the AWCC model was configured with the polyethylene donuts and nickel ring in place but with no cadmium liner inserts (thus configured, the counter has the smallest assay chamber size and is configured for detection of thermal neutrons). Calculations were run for a range of layer thicknesses. The performance of the counter drops off rapidly outside of a range between 1.5 μ m and 2.0 μ m. The calculated data for several thickness values within this range are presented in Table I below. The uncertainty on the FOM values is estimated to be roughly 0.5%. As expected, a thicker layer provides a lower efficiency due to straggling losses, but also provides a shorter DAT due to the greater overall concentration of B-10 capture targets.

B ₄ C Thickness (µm)	Absolute Efficiency	DAT (µs)	FOM (ϵ^2/τ)
1.50	0.2316	51.4	0.001044
1.65	0.2260	48.6	0.001051
1.70	0.2243	47.8	0.001053
1.75	0.2226	47.0	0.001054
1.80	0.2205	46.3	0.001050
1.85	0.2185	45.5	0.001049
2.00	0.2128	43.5	0.001041

Table I. Calculated performance values for AWCC (small chamber, thermal configuration) versus B_4C layer thickness.

Proceeding with performance estimates of other counter configurations, the decision was made to use a layer thickness of $1.50 \,\mu\text{m}$ – this does not maximize the FOM value but still gives a FOM close to the (broad) maximum and provides a slight preference for efficiency in the overall trade-off between efficiency and DAT.

DISCUSSION OF PERFORMANCE ESTIMATES

Models were run for a Cf-252 neutron source in the center of the AWCC with the straws configured as described in the preceding section. Three counter configurations were run:

- Both polyethylene donuts, nickel ring, and the cadmium inserts are all present. The counter is optimized for distinguishing the fast induced fission neutrons from uranium in the sample. This configuration also provides the smallest assay cavity size, and is notated in Table II below as "Small, fast."
- Both polyethylene donuts, and the nickel ring are present; however, the cadmium inserts are removed. In this case the counter is optimized for detecting thermal neutrons, and still has the smallest possible assay cavity size. This configuration could be used in active mode for samples with relatively little uranium content, or in passive mode for plutonium-bearing samples. This is notated in Table II below as "Small, thermal."
- The polyethylene donuts, nickel ring, and the cadmium inserts have all been removed. In addition, all the spacers in the end plugs have been removed. This provides the largest possible assay chamber. As with the above configuration, this is optimized for detecting thermal neutrons and would be used for low-uranium samples or plutonium samples. This is notated in Table II below as "Large, thermal."

For each of the three configurations, the single neutron absolute efficiency and the DAT were estimated from the MCNPX model of the AWCC equipped with straws. The resulting performance values are presented in Table II below along with equivalent measured performance values for a typical He-3 AWCC.

	He-3 AWCC (typical)			Boron straw AWCC (model)		
Counter	Absolute			Absolute		
Configuration	Efficiency	DAT (µs)	FOM (ϵ^2/τ)	Efficiency	DAT (µs)	FOM (ϵ^2/τ)
Small, fast	0.24	51	0.00113	0.1792	39.6	0.000811
Small, thermal	0.30	63	0.00143	0.2316	51.4	0.001044
Large, thermal	0.37	57	0.00240	0.2755	45.0	0.001687

Table II. Measured and calculated performance values for AWCC in various configurations.

For all three counter configurations, the calculated single neutron efficiency for the boron straw equipped AWCC is about 75% that of the He-3 AWCC; however, the estimated DAT for the straw equipped AWCC is also roughly 20% less (faster). Overall, this places the FOM values for the straw equipped AWCC approximately 40% lower than those for the He-3 AWCC. This seems like an acceptable compromise. This is especially true if it's noted that the FOM is roughly indicative of the neutron coincidence assay variance, if all other factors (sample type, count time) are held constant. The assay variance is also roughly proportional to the assay count time. Thus, the same degree of precision as obtainable with a He-3 AWCC is possible with the counter as designed here by counting for approximately 40% longer.

Note that the calculated performance values are for the straws laid out on a 1 cm pitch, which has 1466 straws in the counter body. Further optimization could be explored by going to a tighter straw spacing. A pitch of 0.8 cm was examined briefly. This generally increased the efficiency by about 4%, and decreased the DAT by about 20%. The overall effect raises the FOM values much closer to those for the He-3 AWCC, but at the cost of roughly 60% more straws. In addition, PTi are currently developing a "corrugated straw" approach whereby the straws are not circular in cross section, but rather more star-shaped in cross section. This serves to increase the spatial concentration of B-10 capture targets and should have the same effect as increasing the pitch of the straws. This is a very new development and its specific effect on designs such as the AWCC described here have yet to be evaluated.

Clearly new developments in the technology are ongoing and need to be evaluated. Exactly where the optimum point lies on the trade-off between performance versus cost and complexity, ultimately depends on the assay needs and context. However, the fundamental approach of using boron coated straws in small or medium sized coincidence counters such as the AWCC appears feasible.

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