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PNNL-22567

Boron-10 ABUNCL Active Testing

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July 2013

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Executive Summary

The Department of Energy Office of Nuclear Safeguards and Security (NA-241) is supporting the project *Coincidence Counting With Boron-Based Alternative Neutron Detection Technology* at Pacific Northwest National Laboratory (PNNL) for the development of a ^{3}He proportional counter alternative neutron coincidence counter. The goal of this project is to design, build and demonstrate a system based upon ^{10}B-lined proportional tubes in a configuration typical for ^{3}He-based coincidence counter applications.

This report provides results from testing the active mode version of the General Electric Reuter-Stokes Alternative Boron-Based Uranium Neutron Coincidence Collar (ABUNCL) at Los Alamos National Laboratory using sources and fresh fuel pins. The measurements indicate that a 1% doubles measurement can be made in about 30 minutes with this instrument. This time period may be acceptable for safeguards measurements by inspectors, but that operational question needs to be answered by the IAEA.

Acronyms and Abbreviations

ABUNCL	Alternative Boron-Based Uranium Neutron Coincidence Collar
AmLi	Americium-lithium neutron source
BWR	Boiling water reactor
cps	Counts per second
D	Doubles
DOE	U.S. Department of Energy
DU	Depleted uranium
ε	Detection efficiency
FOM	Figure of Merit
GE	General Electric
GERS	General Electric Reuter-Stokes
HDPE	High Density Polyethylene
IAEA	International Atomic Energy Agency
LANL	Los Alamos National Laboratory
LEC	Low-Energy Cutoff
LEU	Low-enriched uranium
MCA	Multi-Channel Analyzer
MOX	Mixed Oxide fuel
NIM	Nuclear Instrumentation Module
NIST	National Institute of Science and Technology
PHL	Pulse-Height Light
PNNL	Pacific Northwest National Laboratory
Pu	Plutonium
S	Singles
τ	Die-away time
TTL	Transistor-transistor logic
U	Uranium
UNCL	Uranium Neutron Coincidence Collar

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1. Introduction

The search for technological alternatives to ^{3}He is a major research area in nuclear security and safeguards due to the shortage of this gas in recent years [Kouzes 2010; Menlove 2011]. One of the important safeguards applications of ^{3}He has been for coincidence counting instruments. The Department of Energy Office of Nuclear Safeguards and Security (NA-241) is supporting the project *Coincidence Counting With Boron-Based Alternative Neutron Detection Technology* at Pacific Northwest National Laboratory (PNNL) for the development of a ^{3}He proportional counter alternative neutron coincidence counter. The goal of this project is to design, build and demonstrate a system based upon ^{10}B-lined proportional tubes in a configuration typical for ^{3}He-based coincidence counter applications.

General Electric (GE) Reuter-Stokes (Twinsburg, OH) developed a *passive* coincidence collar prototype based on arrays of ^{10}B-lined tubes using Ar fill gas. That prototype was loaned to PNNL for testing against the safeguards requirements for an Alternative Boron-based Uranium Neutron Coincidence Collar (ABUNCL) [McKinny 2012; Kouzes 2012]. It was also reconfigured for use in an *active* mode, and initial testing of the active configuration was also performed at PNNL [Kouzes 2013]. Subsequently, the ABUNCL was taken to Los Alamos National Laboratory (LANL) for testing with fresh low-enriched uranium (LEU) nuclear fuel.

The ABUNCL, as delivered by GE Reuter-Stokes, was configured in a passive boiling water reactor (BWR) coincidence collar configuration. This was modified to an active BWR configuration by replacing one side of the detector with a removable high-density polyethylene (HDPE) door. The shape and size of this HDPE block was not optimized, potentially increasing the die-away time of the system. Listed in Table 1.1 is comparative data characterizing the two ^{3}He-based active BWR configurations, and the ABUNCL.

The ^{3}He-based active BWR UNCL-II has the largest figure-of-merit (FOM) listed in the table, defined as FOM = ε^2/τ. The GE Reuter-Stokes active ABUNCL configuration initial experimental and modeling results were reported in [Kouzes 2013]. While the GE Reuter-Stokes design was targeting the UNCL-I FOM of 3.1, its performance falls below that value with a FOM of 1.7.

Based on the previous measurements at PNNL, the pre-delay time for the ABUNCL was chosen as 4 μs and the gate width was chosen as 100 μs. The preamplifiers (PDT10A/20A) all had a 150 keV threshold and the boron-lined tubes had an operating voltage of 750 V. Engineering students from Washington State University developed a mechanical lift for the ABUNCL that allows the system to be rolled around and lifted to the required height for measurements, as shown in Figure 1.1 in the lab at LANL.

The ABUNCL system was set up and initially tested at LANL on May 14, 2013, and tested with fresh fuel assemblies at LANL on June 11-12, 2013. Measurements were also made of the system stability between these dates. All data were acquired with a JSR-14 shift register (Canberra Industries, Meriden, CT) and a laptop running the INCC code [Krick 2009]. This paper reports on the results of the active testing activities with fresh nuclear fuel.

Table 1.1. Characteristics of UNCL and ABUNCL configurations [Kouzes 2012].

Detector	Total # Tubes, Configuration, & Fuel Cavity H x L x W (cm)	Total No. Moles	Efficiency (ε), Die-Away Time (τ) & FOM =ε^2/τ					
			Measurement Results			Model Results		
			ε	τ (μs)	FOM $(\%)^2/\mu s$	ε	τ (μs)	FOM $(\%)^2/\mu s$
UNCL-I Active BWR	18 ^{3}He tubes 3 Rectangular banks, 41.4 x 16.5 x 23.4	0.44	13.5%*			12.5%	50	3.1
UNCL-II Active BWR	16 ^{3}He tubes 3 Rectangular banks 41.3 x 16.5 x 16.5	0.39	15.3%** 15.4%* 13.5% (±10%)***	58**	4.0	14.9%	53	4.2
GE RS ABUNCL Active BWR****	56 ^{10}B-lined tubes 3 Rectangular banks 78.1 x 16.5 x 23.4	NA	9.4%	83	1.1	11.5%	77	1.7

* From [Menlove et al. 1990] using ^{252}Cf centered in sample chamber

** From [Croft et al. 2011] using ^{252}Cf centered in sample chamber

*** From [Canberra 2011] for JCC-72 using active measurement

**** From [Kouzes 2013] using a 150 keV low energy threshold

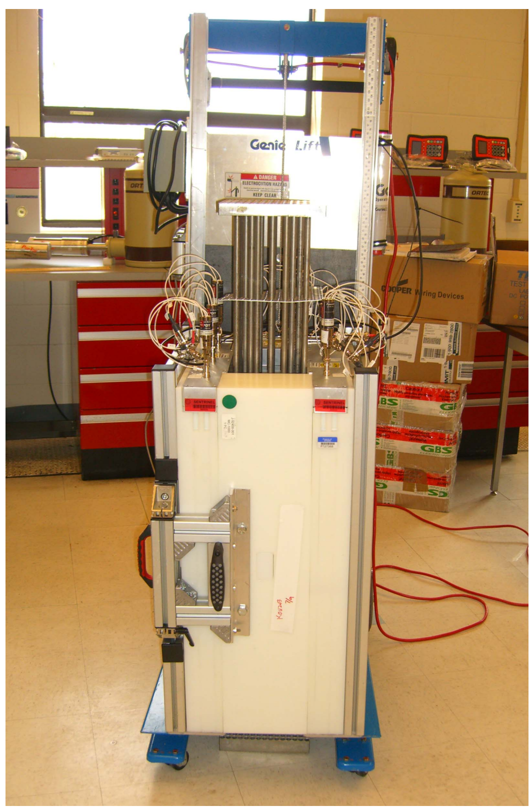

Figure 1.1. GE Reuter Stokes ABUNCL at LANL.

2. Measurements at LANL

All measurements discussed in this report were made at LANL in area TA-35, Building 2. After the system was assembled, the efficiency and the dead time were measured on May 14, 2013 in room C-154. Some extended runs were initiated to look at long-term stability, covering the period from May 14 to June 11, 2013.

Measurements on depleted uranium (DU) and LEU fuel rods (3.19% enriched) were made on June 11-12, 2013 in Room C-157.

The neutron sources used at LANL are summarized in Table 2.1, including two AmLi sources and two ^{252}Cf sources. LANL provided the neutron emission rate (determined from comparison to a standard source) and the derived source activity for the date of measurement, and stated that uncertainties for these source values were typically ~1.5%. The sources were enclosed in small metal pigs.

Table 2.1. Sources used at LANL.

Source	Number	Activity (μCi)	Neutrons (s^{-1})
^{252}Cf	A7-866	31.0	1.09×10^5
^{252}Cf	A7-867	55.6	2.11×10^5
AmLi	N-160	1.17×10^6	4.5×10^4
AmLi	N-161	1.17×10^6	4.5×10^4

Listed in Table 2.2 is the information on the initial measurements performed at LANL on May 14, 2013. The first measurement acquired was a background. The recorded singles rate was 9.7(1) cps (Run 1). The values in parentheses are the reported uncertainties in the last digit shown. All uncertainties are statistical as reported by the INCC code.

The data taken in Runs 4-7 with the ^{252}Cf sources was for the purpose of acquiring a dead time measurement. Run 5 was aborted when another source entered the room and was not used in the analysis. To account for any scattering or attenuation by the second source, each of the single source measurements were made with a "dummy" source of the same shield dimensions and composition as a real source also in place.

Table 2.2. Measurements at LANL on May 14, 2013.

Run	Configuration	Time (s)	Singles (s^{-1})	Doubles (s^{-1})
1	Background	300	9.7(1)	0
2	Background contaminated by source	-		
3	55.6 μCi ^{252}Cf in detector center	600	20304(60)	1805(140)
4	55.6 μCi ^{252}Cf with dummy source on inside door	3600	11607(2)	547(3)
5	55.6 μCi ^{252}Cf with dummy source on inside door	1080	11602(4)	546(6)
6	31.0 & 55.6 μCi ^{252}Cf source on inside door	3600	17478(2)	818(4)
7	31.0 μCi ^{252}Cf with dummy source on inside door	3600	5964(1)	278(2)
8	Two AmLi sources inside source location	300	4947(4)	1(4)

Listed in Table 2.3 is the information on long-term drift measurements made between May 14, 2013 and June 11, 2013 in Room C-154. Gaps of some hours to a day exist between measurements. The values in parentheses are the reported uncertainties in the last digit shown.

Table 2.3. Long stability measurements at LANL from May 14 to June 11, 2013.

Run	Configuration	Dates	Time (s)	Singles (s^{-1})	Doubles (s^{-1})
9	Two AmLi sources inside source location	May 14 - 20	487,200	4893.6(2)	1.2(1)
10	Two AmLi sources inside source location	May 20 - 22	181,800	4899.5(2)	1.5(2)
11	Two AmLi sources inside source location	May 22 - 28	501,600	4880.6(2)	1.2(1)
12	Two AmLi sources inside source location	May 28 - June 4	597,000	4879.4(2)	1.5(1)
13	Two AmLi sources inside source location	June 5 - 11	483,000	4863.7(2)	1.4(1)

Listed in Table 2.4 is the information on the measurements performed at LANL on June 11-12, 2013, in Room C-157. The values in parentheses are the reported uncertainties in the last digit shown. The “Doubles % Error” is the uncertainty listed in the “Doubles” column divided by the rate listed. It is desirable that measurements of a complete fuel assembly have a doubles precision of ~1%, which dictates the amount of time required for measurements. The analysis of the data from these measurements is discussed below.

Table 2.4. Measurements at LANL on June 11-12, 2013.

Run	Configuration	Time (s)	Singles (s^{-1})	Doubles (s^{-1})	Doubles % Error
1	Background	1000	25.9(1)	0.01(1)	100.0%
2	72 DU pins with AmLi source	1480	2758(1)	14(1)	0.8%
3	56 DU plus 16 center LEU pins with AmLi source	3950	2916(1)	32.2(7)	1.9%
4	56 DU plus 16 outside LEU pins with AmLi source	2950	2951(1)	37.4(7)	7.1%
5	56 DU plus 16 rear LEU pins with AmLi source	2875	2875(1)	33.2(7)	2.2%
6	56 DU plus 16 front LEU pins with AmLi source	2970	2970(1)	39.9(6)	1.9%
7	72 DU pins with no source	45660	43.3(2)	2.60(2)	2.1%
8	40 DU plus 32 LEU pins with AmLi source	3570	3096(1)	55.9(7)	1.5%
9	8 DU plus 64 LEU pins with AmLi source	2700	3276(1)	80.8(1)	1.3%
10	72 LEU pins with AmLi source	2700	3299(1)	86.0(9)	1.1%
11	72 LEU pins with no source	1800	51.2(3)	3.66(7)	1.0%
12	72 LEU pins with AmLi source shifted to front	1800	3206(1)	90(1)	1.1%
13	72 LEU pins with AmLi source shifted to back	1800	3331(1)	82(1)	1.2%

3. Efficiency and Dead Time Measurements

The efficiency of the ABUNCL active configuration (3 detector slabs) was measured at LANL on May 14, 2013, by placing a ^{252}Cf source (A7-867) at the center of the detector volume and measuring the net singles rate (Run 3 in Table 2.2), giving 9.6(5)%, compared to 9.4(5)% measured at PNNL [Kouzes 2013].

The dead time of the system was measured using the two-source method [Henzlova 2010]. The two ^{252}Cf sources were used individually and together to get the singles and doubles rates (Runs 4, 6 and 7). In this method, three measurements of singles (S) and doubles (D) are made: 1) source 1 and a dummy source (to include the effect of scattering) placed next to each other are measured, 2) source 1 and source 2 together are measured, and 3) source 2 and the dummy source are measured. The dead time correction parameter, A, is then found iteratively as discussed below.

The dead time corrected doubles are given by [Henzlova 2010]:

$$D_{corr} = De^{\delta S}$$

where

$$\delta = A + \frac{A^2}{4} S$$

in which A is the dead time correction parameter determined experimentally. By using the singles from source 1 (S_1), singles from source 2 (S_2), singles from the combination (S_{1+2}), doubles from source 1 (D_1), doubles from source 2 (D_2), and doubles form the combination (D_{1+2}), the parameter A can be varied until the following expression is true:

$$D_1 e^{\delta S_1} + D_2 e^{\delta S_2} = D_{1+2} e^{\delta S_{1+2}}$$

Based on the measured data listed in Table 2.2, the result for the dead time parameter δ was found to be 9.20×10^{-7}, corresponding to 920 ns.

4. Long Term Drift

Between May 14, 2013 and June 11, 2013, long measurements (2-6 days each) were made with two AmLi sources in place in the source cavity in the HDPE door (Runs 9-13 in Table 2.3). The total acquisition time was over 26 days. While the individual singles rates for the five runs had smaller statistical errors, the standard deviation between these measurements was 0.2%, indicating some variation in efficiency over time.

The air temperature above the ABUNCL was monitored during the entire measurement period using an EL-USB-2-LCD temperature, humidity, and dew point data logger. The record of the temperature (sampled every five minutes, giving over 15,000 data points) in Rooms C-154 and C-157 is seen in Figure 4.1. The average air temperature during this period was 24.6°C, with a minimum of 22.0°C and a maximum of 32.5°C. The spikes seen in the data are real transients that occurred around 7:30 AM each day, presumably due to air conditioning variations, having durations of tens of minutes. Note that the temperature had two dips of a couple degrees in the middle, and an increase of a couple degrees toward the end of the time period.

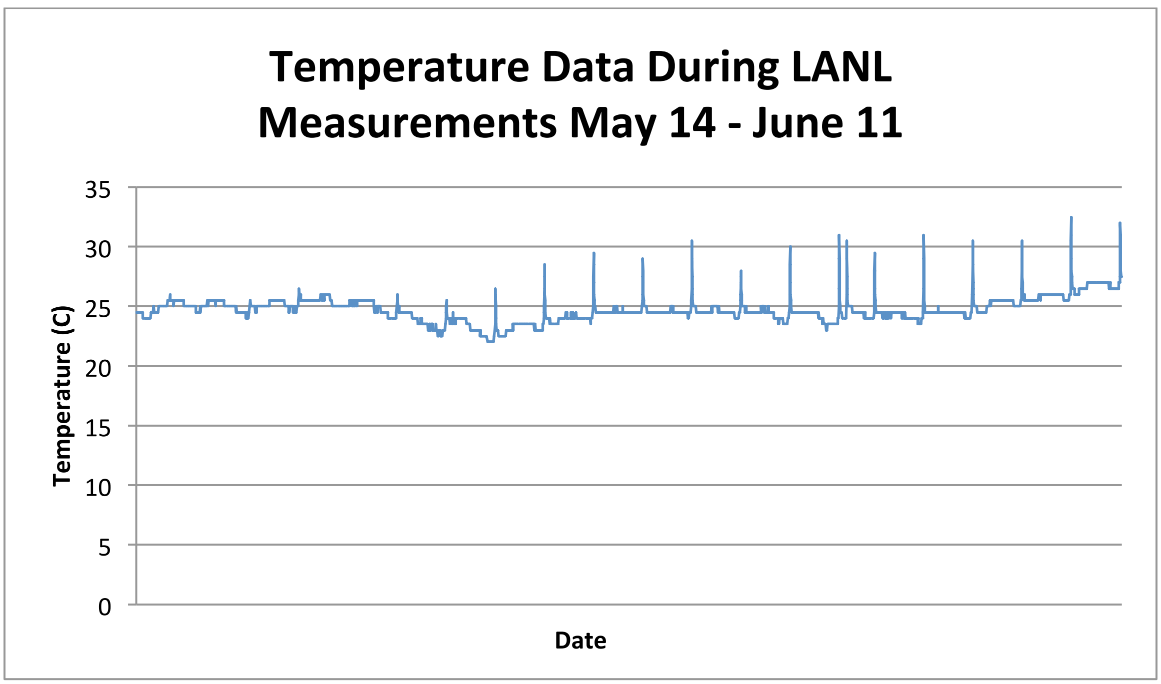

Figure 4.1. Temperature variation at LANL from May 14 to June 11, 2013 measured every 5 minutes.

The singles counts accumulated during the period from May 14 to June 11, 2013, are shown in Figure 4.2. Each value is counts per 600 s. The vertical bar indicates a 1% variation. There was almost a 1% swing in count rate over this one-month period from the highest to the lowest count value (well outside of statistical variation). There appears to be an anti-correlation in the counts with temperature variations observed, which could be explained as due to thermal expansion of the detector system, a shift in the electronics threshold affecting the efficiency, and temperature dependence in the neutron capture cross-section on boron (the thin-target thermal neutron cross-section decreases by ~0.17%/°C)[1] [Henzlova 2012]. The short temperature spikes do not seem to be reflected in the count rate data. The long duration temperature swings shown in Figure 4.1 (maximum of 5°C) seem to anti-correlate with the change in singles rate shown in Figure 4.2. If the temperature deviations of a few degrees seen in Figure 4.1 are responsible for the change in efficiency, then it appears that the singles rate changes by about 0.25%/°C. It is unclear if the drop in efficiency over the entire time period is due to a slow downward drift in efficiency or is just due to environmental temperature changes. About two thirds of the change could be attributed to the cross-section temperature dependence.

This variation is perhaps an order of magnitude larger than that seen for ^{3}He-based systems; thus, stabilizing the system against temperature changes may be needed. If the temperature in a facility is stable for many hours, this temperature dependence may not be a problem since detector efficiency can be determined from neutron singles measurements of a calibration source. A longer-term measurement in an environmental chamber should be performed to observe if the ABUNCL efficiency trend continues downward or correlates only with temperature.

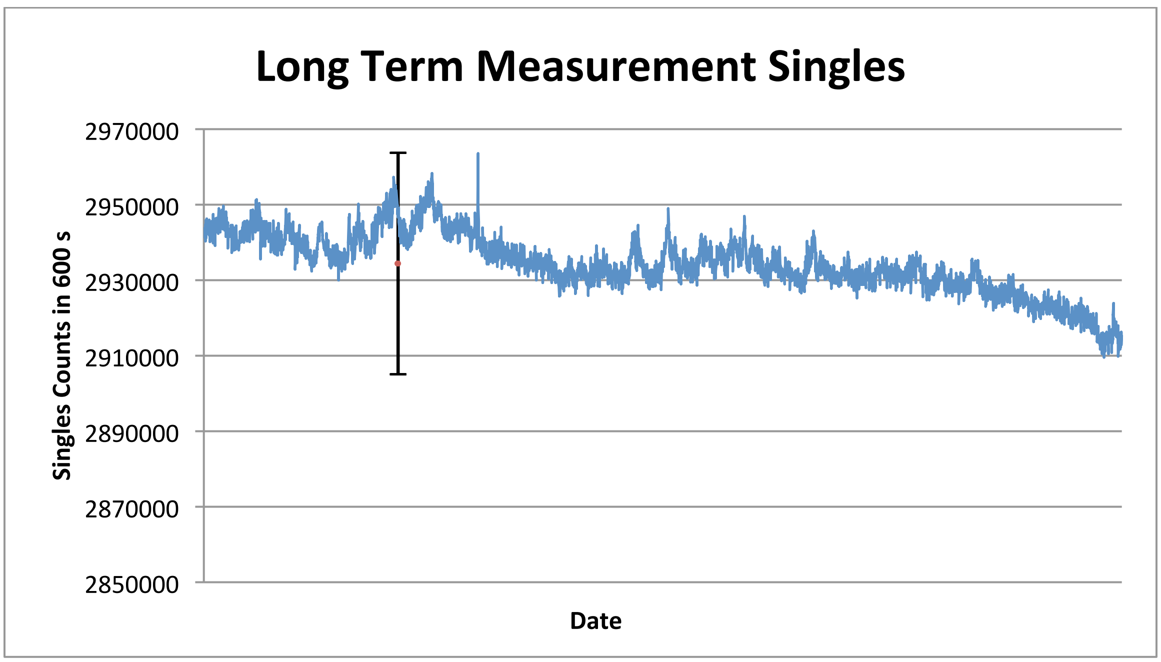

Figure 4.2. Singles counts at LANL from May 14 to June 11, 2013 measured every 10 minutes.

[1] Since the capture cross-section (σ) varies as the inverse of the speed (v), and $v \sim \sqrt{T}$, where T is temperature, then $d\sigma/\sigma \sim -dT/2T$. At room temperature (~300K), $d\sigma/\sigma = 0.17\%$ per degree C.

5. Fresh Fuel Measurements

A series of measurements was made on June 11-12, 2013, on DU and LEU fuel arranged in a support rack, as seen in Figure 5.1 from above. The ABUNCL was arranged approximately on the center of the fuel assembly, both vertically and horizontally (the hole in the ABUNCL for fuel is rectangular rather than square). The fuel pins (138 cm long with 128 cm of fuel in each pin, 1 cm in diameter) were arranged in a 9 x 9 array (spaced at 1.4 cm), which is consistent with a BWR assembly. There were 9 “guide” holes in the array, resulting in a total of 72 holes for fuel pins.

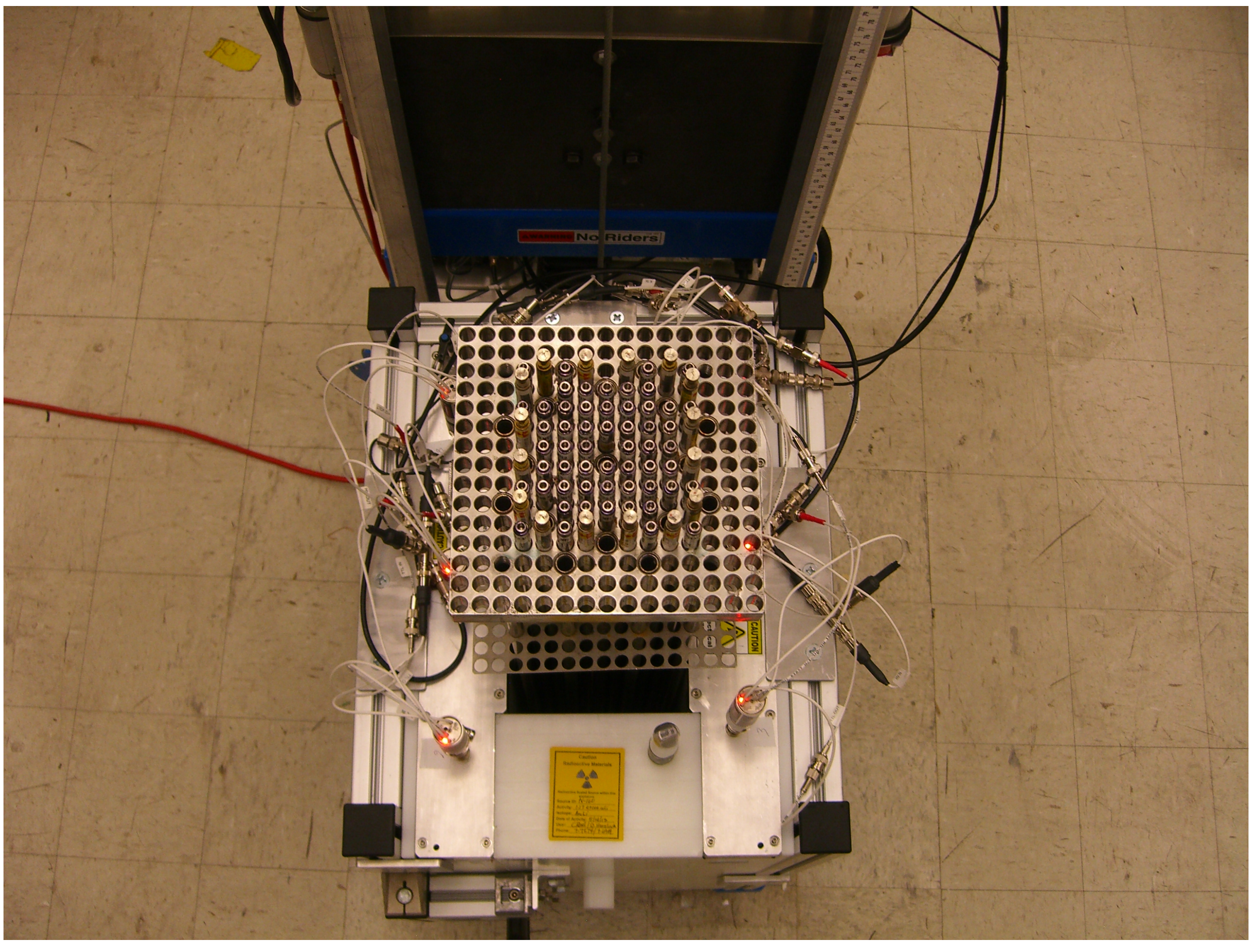

Figure 5.1. ABUNCL at LANL with fuel pins loaded (DU in the center, LEU on outside).

For all measurements, all 72 fuel pins (DU or LEU) were in place. A number of different configurations were measured, including all DU and all LEU fuel, and mixtures of each. Figure 5.2 shows a schematic of the various configurations measured. The outer line represents the 9 x 9 space. The black squares represent the guide holes, and the blue squares represent the LEU pins. All other white holes within the 9 x 9 array were filled with DU fuel pins. Configuration 0 was all DU pins. Measurements were made on 8 LEU pins (Configurations 1 and 2), 16 LEU pins

(Configurations 3, 4, 5, and 6), 32 LEU pins (Configurations 7), 64 LEU pins (Configurations 8), and 72 LEU pins (not shown).

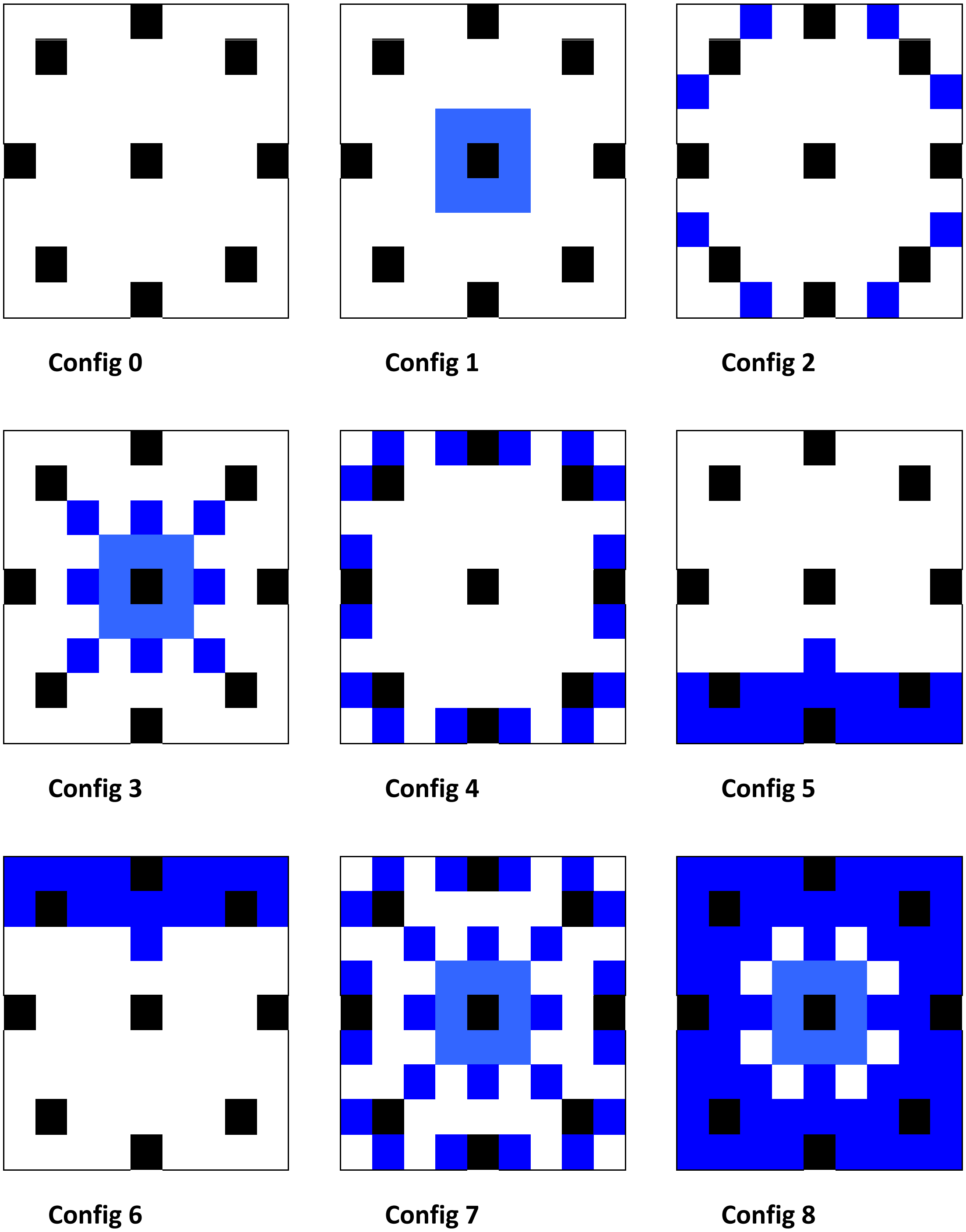

Figure 5.2. Fuel configurations used during measurements, where blue represents LEU pins, black are guide tubes, white are DU pins.

The data results from these measurements are summarized in Table 2.4. The doubles rates are plotted in Figure 5.3, where the red line is for no AmLi source and the green line is with the AmLi source loaded in the source holder location. Error bars are smaller than the markers. There is a general trend for a larger signal with more LEU pins, as expected. Based on such measurements, the mass of uranium in the assembly could be inferred if the system were calibrated with a reference fuel assembly to about 1% precision.

For the 16 LEU pin data, placing the pins at the center or the rear of the assembly (away from the AmLi sources) reduces the signal compared to the other locations (around the outside, or at the front near the AmLi source). Moving the LEU pins from the front to the back of the pin assembly causes a 17% change in the doubles rate. This is a smaller effect than seen in the specific ^{3}He-based UNCL geometry tested in 1981 [Menlove 1981], though that geometry was different than current UNCL designs.

For the 72 LEU pin case, moving the entire assembly to the front (near the AmLi source) increases the signal, while moving it to the rear (away from the AmLi source) reduces the signal, with a 9% change in doubles rate.

As listed in Table 2.4, the measurement precision for the full LEU fuel load was 1.1% in 30 minutes. For a full DU load, it was about 2.2% in an hour. The desired precision for a LEU fuel assembly is a 1% measurement in 15 minutes. The fact that the ABUNCL takes twice as long to make a comparable measurement as a ^{3}He-based UNCL may be acceptable to an inspector since longer times are usually needed to load and unload the fuel in the instrument.

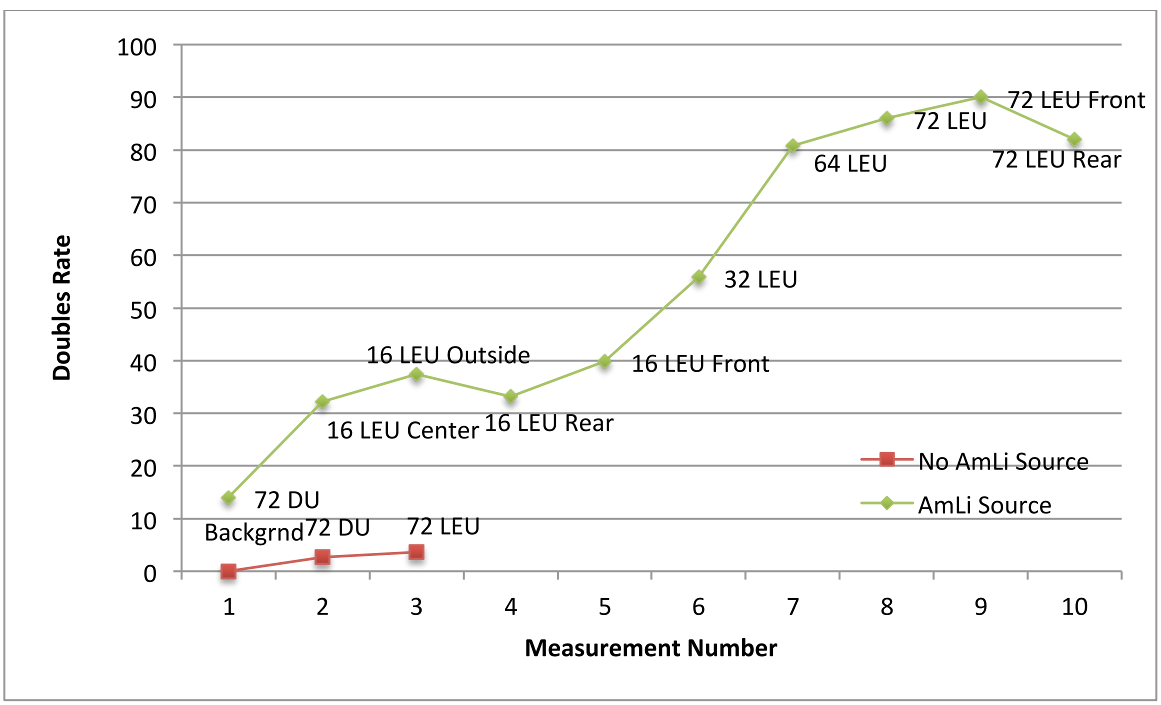

Figure 5.3. Doubles rates from fuel measurements.

6. Conclusions

This report discussed the characterization of a full-scale boron-based ABUNCL coincidence counter developed by GE Reuter-Stokes for applications in safeguards under the project *Coincidence Counting With Boron-Based Alternative Neutron Detection Technology*.

Long dwell measurements were made on the system to determine stability. A decrease in efficiency was observed over a one-month period that correlated with a slow increase in average temperature. Measurements in an environmental chamber should be performed to determine if the changes were caused by temperature variation or if there is a slow decrease in efficiency due to other causes.

Results were provided from measurements on combinations of DU and LEU fuel from 72 DU pins up to 72 LEU pins in various combinations. The measurements at LANL indicate that a 1% doubles measurement can be made in about 30 minutes. This time period may be acceptable for safeguards measurements by inspectors, but that operational question needs to be answered by the IAEA.

Modeling with MCNPX [Pelowitz 2011] is planned for comparison to the measurements reported here. Each of the fuel configurations will be modeled to compare to these measurements. This will validate the predictive capability of the model for planning of future measurements.

7. Acknowledgements

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