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PNNL- 19050

BF_3 Neutron Detector Tests

Revision 0

RT Kouzes
JH Ely
AT Lintereur
ER Siciliano
ML Woodring

December 9, 2009

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Pacific Northwest National Laboratory
Richland, Washington 99352

Executive Summary

Radiation portal monitors used for interdiction of illicit materials at borders include highly sensitive neutron detection systems. The main reason for having neutron detection capability is to detect fission neutrons from plutonium. The currently deployed radiation portal monitors (RPMs) from Ludlum and Science Applications International Corporation (SAIC) use neutron detectors based upon ^{3}He-filled gas proportional counters, which are the most common large neutron detector. There is a declining supply of ^{3}He in the world; thus, methods to reduce the use of this gas in RPMs with minimal changes to the current system designs and detection capabilities are being investigated.

Reported here are the results of tests of the efficiency of BF_3 tubes at a pressure of 800 torr (1.07×10^{5} Pa). These measurements were made partially to validate models of the RPM system that have been modified to simulate the performance of BF_3-filled tubes. While BF_3 could be a potential replacement for ^{3}He, there are limitations to its use in deployed systems.

Model simulations indicate that three, one-atmosphere (760 torr, 1.01×10^{5} Pa) BF_3-filled tubes would be required to provide the same performance as one tube filled with three atmospheres of ^{3}He gas, with two tubes falling just below the required efficiency. The experimental measurements reported here on BF_3 tubes at 800 torr (1.07×10^{5} Pa) indicate that two such tubes may be marginally sufficient to replace one three-atmosphere ^{3}He tube in a standard SAIC moderator box. The disagreement between the experimental and simulated results is less than 15%. The BF_3 tubes tested require a substantially higher voltage (about 2200 V) to operate than ^{3}He tubes (about 1000 V), and the voltage increases with pressure. Thus, BF_3 tubes may be difficult to deploy to the field due to breakdown concerns in humid environments. Modifications to the SAIC electronics to operate these tubes would be necessary. Since BF_3 is toxic, there are also considerations about the acceptability of deploying this gas in the field.

It would be of value to explore the dependence of BF_3 efficiency as a function of gas pressure to lower pressures than tested so far.

Acronyms and Abbreviations

ANSI	American National Standards Institute
atm	atmospheres
BF_3	boron trifluoride
CBP	Customs and Border Protection
cps	counts per second
DOE	U.S. Department of Energy
MCNP	Monte Carlo for Neutrons and Photons Transport Code
Pa	Pascal
PNNL	Pacific Northwest National Laboratory
PolyBox	polyethylene moderator/reflector box
POV	personally owned vehicle
PVT	Polyvinyl Toluene (plastic) scintillation gamma detector
RPM	Radiation Portal Monitor
RSP	Radiation Sensor Panel
SAIC	Science Applications International Corporation

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1 Purpose

Radiation portal monitor (RPM) systems used for interdiction of illicit materials at borders include highly sensitive neutron detection systems. The main reason for having neutron detection capability is to detect fission neutrons from plutonium. The currently deployed RPMs from Ludlum and Science Applications International Corporation (SAIC) use neutron detectors based upon ^{3}He-filled gas proportional counters, which are the most common large neutron detectors.

Within the last few years, the amount of ^{3}He available for use in gas proportional counter neutron detectors has become more restricted, while the demand has significantly increased, especially for homeland security applications (Kouzes 2009). In the near future, the limited supply is expected to curtail use of ^{3}He; therefore, alternative neutron detection technologies are being investigated for use in the RPM systems being deployed for border security applications (Van Ginhoven 2009).

Reported here are the results of tests with BF_3 tubes at a pressure of 800 torr ($1.07x10^5$ Pa) to replace the 3 atmosphere (2280 torr) ^{3}He tubes within one of the SAIC units. This work supplements previous measurements on BF_3 for tubes at 900 torr ($1.2x10^5$ Pa) (Ely et al. 2009). Boron tri-fluoride tubes require a significantly higher operating voltage than ^{3}He tubes (2-3 kV versus less than 1 kV), even at the low pressures required to make operational proportional counters with this gas. Thus, one reason for testing the 800 torr ($1.07x10^5$ Pa) tubes was to determine if the operating voltage could be kept at a level that might be achievable in fielded systems. Another reason for these measurements was to validate models of the RPM system that were modified to simulate the performance of BF_3 tubes. While such tubes could be a potential replacement for ^{3}He, there are limitations to its use in deployed systems.

2 Test Hardware

2.1 SAIC RPM8 Neutron Detector

The SAIC RPM8 RSP employs a polyvinyl toluene (plastic scintillator) (PVT) for photon (gamma) sensitivity and ^{3}He tube detectors for neutron sensitivity. One or two neutron-detector tubes are located inside a polyethylene moderator/reflector "PolyBox" with exterior dimensions of 127 × 305 × 2210 mm (5.0″ × 12″ × 87″). The SAIC systems were purchased under a specification (Stromswold et. al., 2003) that requires a single radiation sensor panel (RSP) to meet the following requirements:

"A ^{252}Cf neutron source will be used for testing neutron sensor sensitivity:

- To reduce the gamma-ray flux, the source shall be surrounded by at least 0.5 cm of lead. To moderate the neutron spectrum, 2.5 cm of polyethylene shall be placed around the source.
- The absolute detection efficiency for such a ^{252}Cf source, located 2 m perpendicular to the geometric midpoint of the neutron sensor, shall be greater than <u>2.5 cps/ng</u> of ^{252}Cf. The neutron detector center shall be 1.5 m above grade for this test. (Note: 10 nanograms of ^{252}Cf is equivalent to 5.4 micro-Ci or 2.1×10^4 n/s,[1] since ^{252}Cf has a 3.092% spontaneous fission (SF) branch and 3.757 neutrons/SF.)
- The neutron detector shall not generate alarms due to the presence of strong gamma-ray sources. The ratio of neutron sensor gamma-ray detection efficiency to neutron detection shall be less than 0.001."

In addition, these systems are required to meet all aspects of the ANSI N42.35 standard (ANSI 2004). A summary of neutron detection systems in RPMs can be found in a PNNL report (Kouzes et al. 2007).

A single SAIC RPM8 RSP in its steel support stand and mounted on a prototype re-locatable base (shown in Fig.2.1) was used for the testing. On that base, the Steel Support Stand was approximately 200 mm above the pavement, and also approximately 200 mm from two large (220 mm diameter) steel bollards. The panel was oriented in the cargo configuration (that is with the electronics at the bottom of the panel). The RSP was modified by the replacement of the ^{3}He tube with the BF_3 tubes and tested.

2.2 BF_3 Tubes

The tests included one to four LND 253109, stainless steel tubes filled with BF_3 gas to 800 torr ($1.07\text{x}10^5$ Pa). Except for the different fill-gas, these are the same LND tube geometry used in the existing SAIC configuration. The BF_3 gas provides the boron atoms as the neutron target and also acts as the proportional gas that is used to detect the charged particles (a ^{7}Li ion and an α particle) that result from the $^{10}B(n,\alpha)$ reaction.

The BF_3 tubes were operated at 2300 V, which exceeds the maximum voltage that can be supplied by the SAIC RPM system. Thus, external electronics and multichannel analyzer were used to make the measurements instead of the SAIC system. The pulse shape from the BF_3 tubes is also not compatible with the SAIC electronics. The testing consisted of placing the tubes into the moderator box of a SAIC RSP. Figure 2.1 shows the RSP and the source holder, with the electronics to the right. Measurements with one and two 3-atmosphere ^{3}He tubes were also made with the external electronics for comparison purposes.

[1] The value of 2.1×10^4 n/s quoted in the reference has now been corrected to 2.3×10^4 n/s

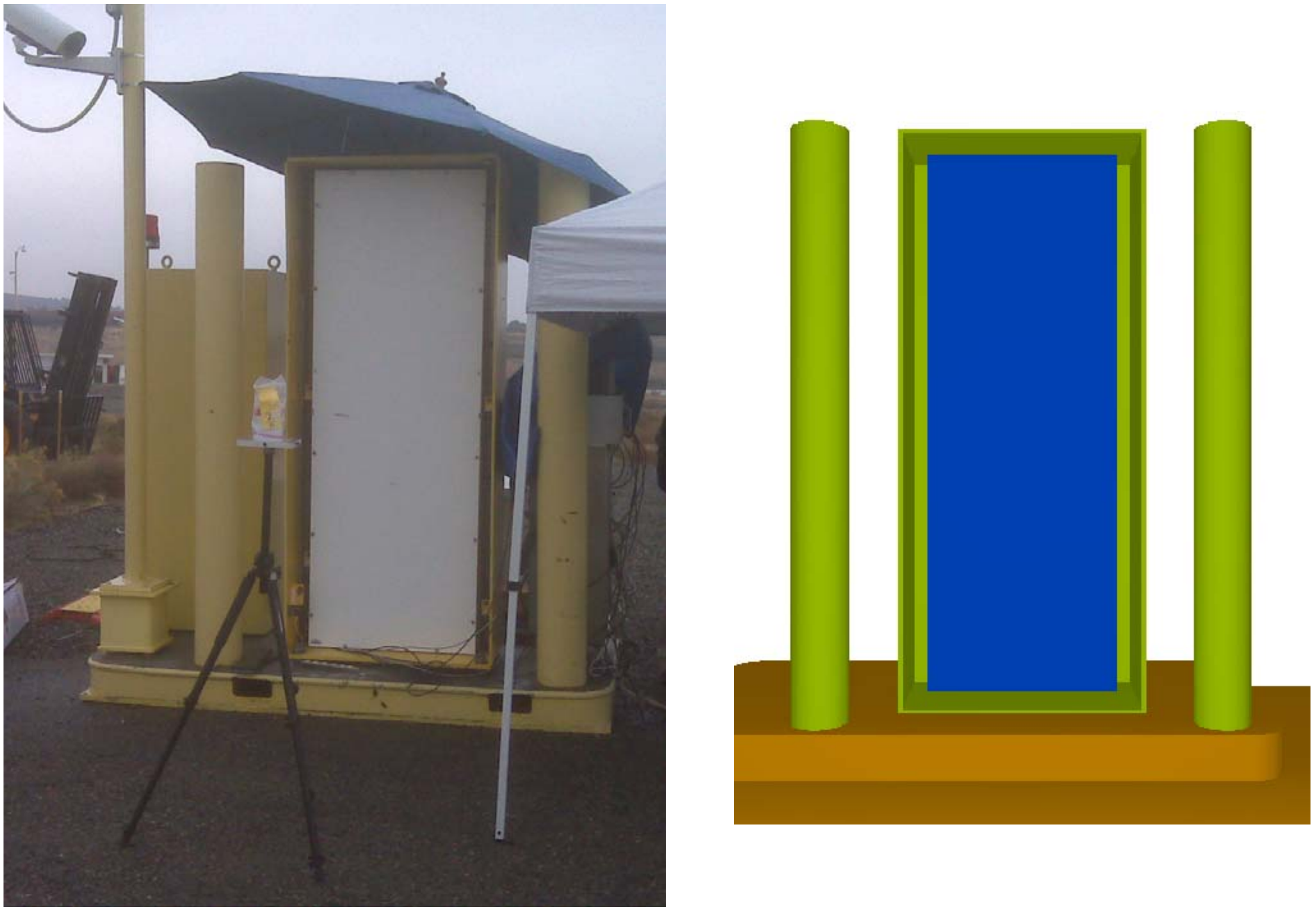

Figure 2.1. Configuration of Equipment Tested and Model System, Showing RSP In The Support Stand Mounted On The Prototype Re-Locatable Base.

2.3 Neutron Source

The neutron source used for this test was ^{252}Cf. The source was purchased from Isotope Products Laboratory (IPL) and was given a PNNL ID of 60208-44. This source was accurately calibrated by NIST and the ^{252}Cf activity was 21.9 ± 1.2 μCi on October 1, 2009. This corresponds to an estimated 41 ng of the pure isotope and an emanation rate of $9.5 \pm 0.5 \times 10^4$ n/s, using the 2.3×10^3 n/s/nanogram conversion factor stated above. On the day of the test, October 21, 2009, the ^{252}Cf activity was 21.6 ± 1.2 μCi. The total-source neutron emission rates on that day were calculated to be 9.3×10^4 n/s, with a margin of error of $\pm 0.5 \times 10^4$ n/s.

The source was in a shielded (25.4 mm poly moderator and 6.4 mm lead) configuration. For the tests, the source was placed on a tripod.

2.4 Test Facility

The test was performed at PNNL at the 331G Integration Test Facility located in Richland, WA. The test was performed on Wednesday, October 21, 2009.

3 Test Limitations

There were limitations for this test and results may change with different conditions.

- Only one test location was used, with the corresponding background. Since the testing was focused on net results (background subtracted) this should have little effect on the overall results.

- One BF_3 tube design from one vendor was used. Because of the simplicity of design, alternate vendor tubes are likely to have similar performance. For the previous 900 torr ($1.2x10^5$ Pa) measurements (Ely 2009), the BF_3 tubes used were GE-Reuter-Stokes model RS-P1-1672-205. These 51 mm diameter GE-RS tubes had the same (1830 mm) active height as the 800 torr ($1.07x10^5$ Pa) LND tubes, but used a slightly thicker (0.89 mm vs. 0.51 mm) stainless steel with the same outer diameter to give a slightly smaller active volume.

- One PolyBox design was used in the modeling and experiment. The intent of the test was to use the polyethylene box available in SAIC RPMs (which is very different from the Ludlum moderator).

4 Experiment Equipment and Setup

For the static test, the neutron source was located 2 m from the center of, and perpendicular to, the front face of the SAIC RSP corresponding to the RPM specification requirements (Stromswold, 2003). The height of the source was 1.35 m above the local grade to locate the source at the mid-height of the panel.

One configurations of the neutron source were used: the ^{252}Cf source with shielding in place as required in the specification (6.4 mm of lead and 25.4 mm of polyethylene). The shielded source provided data for the specification requirements. In addition to the neutron source measurements, background (no source) measurements were taken for each detector configuration.

For each source and detector configuration, data were collected over a period of five minutes. The data were collected with external electronics and software.

5 Modeling Results

In a previous report (Kouzes and Siciliano 2009), a series of calculations with the Monte Carlo N-particle neutrons and photons transport code (MCNP 2003) were reported for the currently deployed standard cargo (four-panel) RPM systems from Ludlum and SAIC mounted on a 0.25 m concrete curb. The simulations explored the effects of changing the ^3He pressure, as well as replacing it with BF_3. The effects of some simple modifications to the SAIC PolyBox in that model system were also simulated to estimate if such changes might improve its performance. However, only a comparison to the stock PolyBox measurements is reported here. These results include simulations for one and two ^3He tubes from LND and for one to four tubes of the same size and composition but filled with BF_3 gas, assuming 96% ^{10}B enrichment. For the ^3He gas, simulations were evaluated over the range from one to three atmospheres (760 to 2280 torr, 1.01×10^5 to 3.04×10^5 Pa) of fill-gas pressure. For the BF_3 gas, the pressure was evaluated for only one and 1.2 atmospheres. The results are shown in Figure 5.1.

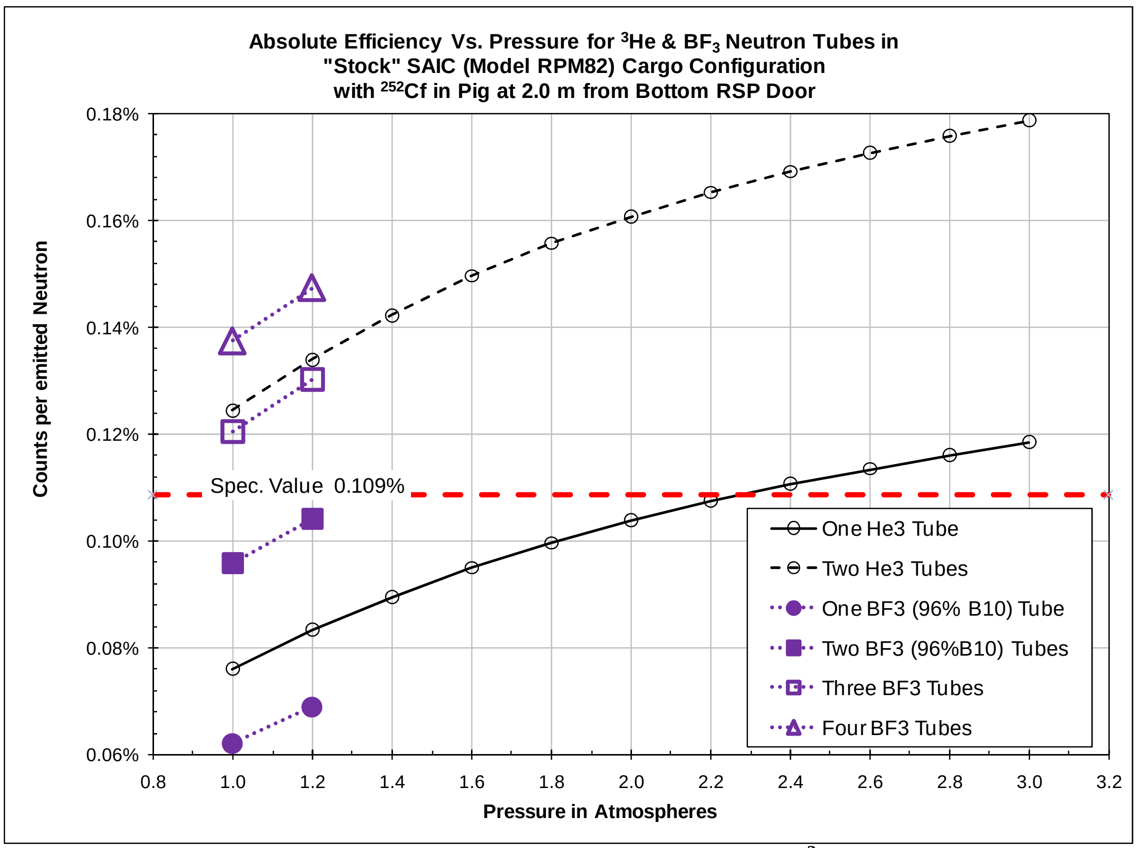

Figure 5.1. Absolute Detection Efficiency for Multiple ^3He and BF_3 Tubes

The model predicts that the BF_3 at one atmosphere in the Stock PolyBox has an absolute efficiency of 0.062% for the source in the polyethylene pig, compared to 0.076% for ^3He at one atmosphere, and 0.118% for ^3He at three atmospheres, which is the pressure of the currently deployed ^3He tubes. This can be compared to the required efficiency of 0.109%. These model results tend to suggest that one tube of

BF_3 at one atmosphere is about a factor of two less efficient than one tube of ^{3}He at three atmospheres; however, it ends up taking three BF_3 tubes in the PolyBox to unequivocally realize a greater efficiency than one ^{3}He tube because of interference effects between multiple, closely-spaced tubes.

From the above results, it is seen that it takes at least three BF_3 tubes at one atmosphere to exceed the absolute-efficiency requirement of 0.109%. Going from one to two to three tubes increases the efficiency from 0.062% to 0.096% to 0.120%, respectively. With the Stock PolyBox, even four BF_3 tubes cannot reach the efficiency of two, three-atmosphere ^{3}He tubes. Inserting more than four tubes requires modifying the Stock PolyBox.

To estimate the change in these results that might arise from the difference between the standard Cargo curb and the lower, bollard-equipped Re-locatable Base, a model of this base was constructed (shown in Figure 2.1). That version of the SAIC model was used to recalculate the one-to four BF_3 tube configurations, but at the measured pressures of 800 torr (1.07×10^5 Pa) and 900 torr (1.2×10^5 Pa). The results are given in Table 5.1, where the 800 torr (1.07×10^5 Pa) values were found to increase by 4-5% over the one atmosphere (760 torr, 1.01×10^5 Pa) original values shown in Figure 5.1, but the 900 torr (1.2×10^5 Pa) values did not change with respect to the 1.2 atmosphere (912 torr) values. The change from the standard Cargo configuration to the Re-Locatable Base at the same tube pressure was found to give at most a 1% increase. Changing from 760 torr (1.0 atm, 1.01×10^5 Pa) to 800 torr (1.07×10^5 Pa) also gave an increase, resulting in the net increase of 4-5%. But changing from 912 torr to 900 torr (1.2×10^5 Pa) gave a slight (~1%) decrease that was offset by the ~1% increase because of the prototype Re-Locatable Base.

Table 5.1. BF_3 tube model results for absolute efficiency versus pressure.

Pressure (torr)	One Tube	Two Tube	Three Tube	Four Tube
760 (standard)	0.062%	0.096%	0.120%	0.137%
800 (re-locatable)	0.065%	0.099%	0.125%	0.142%
900 (re-locatable)	0.069%	0.104%	0.131%	0.149%
912 (standard)	0.069%	0.104%	0.131%	0.149%

The simulation results reported here have slightly higher (2%-6%) efficiencies than reported earlier (Ely 2009) due to improved accuracy in the geometry of the RSP, LND tubes, and PolyBox components of the models.

6 Results and Data Analysis

The first measurements made were of the voltage plateau for the BF_3 tubes to verify the recommended operating voltage from the manufacturer. This was followed by measurements of the efficiency for one to four tubes.

6.1 Voltage Plateau

The operating voltage range recommended by the vendor was 2200 – 2450 V, thus the voltage plateau was measured from 2100 V to 2550 V, as shown in Figure 6.1. There was little variation in performance over this range, so an operating voltage of 2300 V was used in the subsequent measurements. This high operating voltage is a potential problem for fielded equipment where the unit is not hermetically sealed and humidity can cause voltage breakdown.

Figure 6.2 shows a spectrum of the pulse height from the two BF_3 tubes located in the SAIC polyethylene moderating box (in the steel housing). The observed peak is not as symmetric as that observed with ^{3}He tubes due to the physics of the tube.

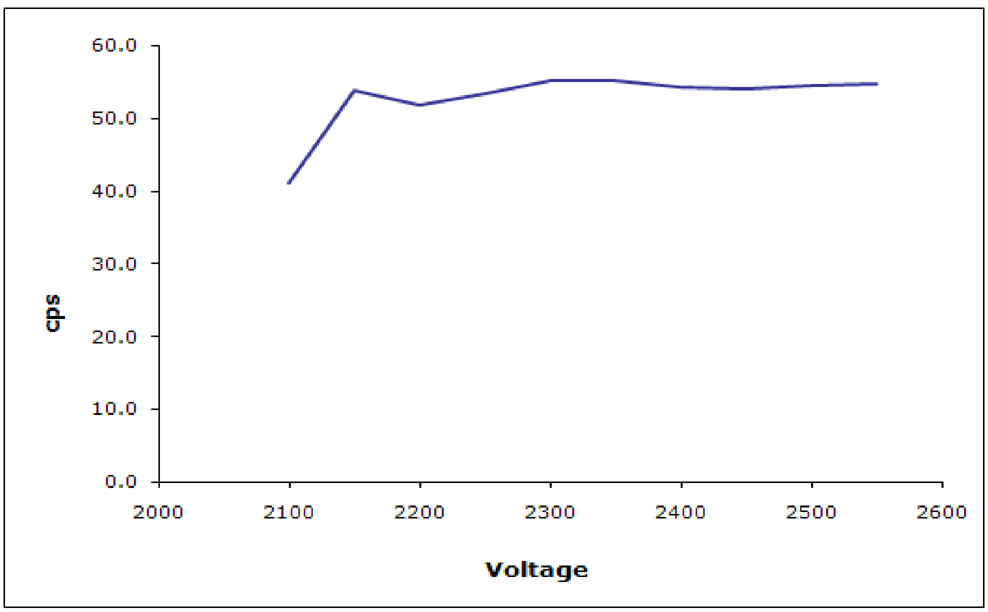

Figure 6.1. Measurement of counts per second versus operating voltage.

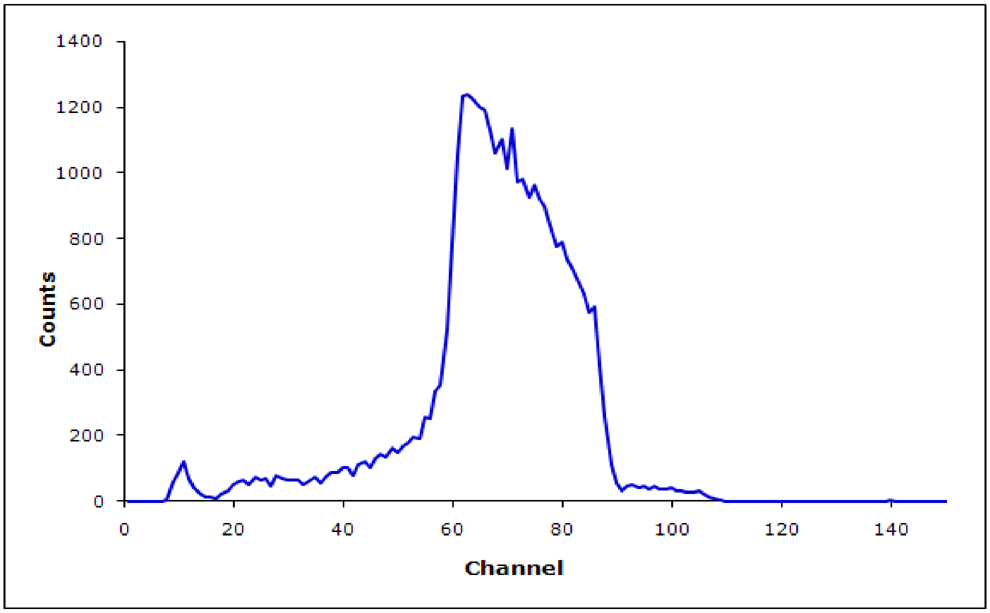

Figure 6.2. Pulse height spectrum from BF_3 tubes.

6.2 Absolute Efficiency

The absolute efficiency of the neutron assembly was measured for one to four tubes with the moderated source at a distance of 2 m from the detector. Since only two data acquisition channels existed, for the configurations with three and four tubes, only two of the tubes were instrumented. The analysis for the three and four tube measurements was performed by assuming that the tubes responded symmetrically. Thus, the uninstrumented one or two tubes were accounted for by adding the matching instrumented tube twice to the total counts. This potentially introduces a small error to the results. Table 6.1 shows the data obtained, including the counts per second (cps) values for the two tubes that were instrumented (cps/2-tubes) and for all the tubes (cps All Tubes) in the measurements (one pair plus one tube or two pairs). This total was converted to counts per second per nanogram of ^{252}Cf source material to compare with the specified value of 2.5 cps/ng. The last two columns show the values obtained previously with the 900 torr (1.2×10^5 Pa) BF_3 tubes (Ely, 2009), where the "Corrected" column has been scaled to correct for the recalibrated ^{252}Cf source strength (from 9 µCi to 8 µCi on the measurement date).

Table 6.1. Neutron count rates for tested configurations of BF_3 tubes.

# Tubes	cps/2-tubes	cps All Tubes	800 torr cps/ng	900[1] torr cps/ng	Corrected 900 torr cps/ng
1	65	65	1.6	1.4	1.58
2	107	107	2.7	2.3	2.59
3	86	131	3.3	2.7	3.04
4	75	150	3.7	3.0	3.38

[1] data from Ely, 2009

Figure 6.3 shows a graph of the absolute neutron efficiency, where the upper curve is the results from the 800 torr (1.07×10^5 Pa) tubes; the middle curve from the source-corrected 900 torr (1.2×10^5 Pa) tube (Ely, 2009); and the lowest curve from the simulations. The absolute efficiencies from the simulations are about 10-15% less than the measured values. The uncertainty in the latest results is dominated by the source strength uncertainty of 5.7%, while the earlier data had a 10% uncertainty. These latest results indicate that two BF_3 tubes at 800 torr (1.07×10^5 Pa) can (marginally) meet the required efficiency (horizontal red line in the figure) as specified by PNNL (Stromswold, 2003).

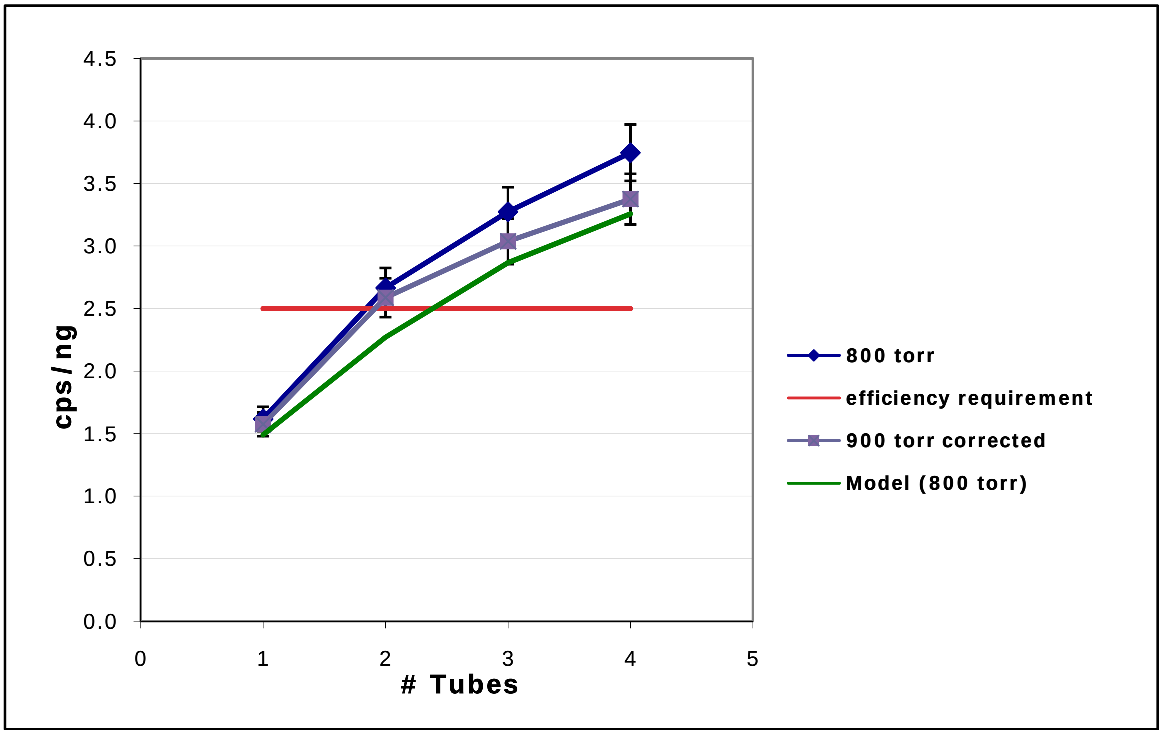

Figure 6.3. Absolute efficiency as a function of the number of BF_3 tubes.

This result differs from the result reported previously for the 900 torr (1.2×10^5 Pa) tubes (Ely, 2009) and this difference can be partially attributed to the larger source uncertainty in the earlier measurements. There may also have been an impact from the electronics used in the previous measurements. External electronics were used to supply the high voltage in both of the tests, but the multichannel analyzers differed between the 800 torr (1.07×10^5 Pa) and 900 torr (1.2×10^5 Pa) measurements, with potentially different dead times and thresholds.

The neutron source used in the measurements for the 800 torr (1.07×10^5 Pa) tubes (NIST referenced source) was used to cross calibrate the neutron source used for the previous measurements on the 900 torr (1.2×10^5 Pa) tubes. This cross calibration was accomplished by making measurements with both sources in the same geometry and a ^{3}He-based neutron detector. This measurement showed that the source strength used by Ely (2009) was apparently lower than implied by its calibration. The scaled calibration gives a source strength of 8 µCi at the time of the 900 torr (1.2×10^5 Pa) measurements (June 30, 2009) rather than the 9 µCi used in the previous paper (Ely, 2009). Ely's results were corrected with the new source strength and the scaled data for the 900 torr (1.2×10^5 Pa) tubes are shown in Table 6.1 and plotted in Figure 6.3. The two sets of measurements have overlapping uncertainties, but it was expected that the 900 torr (1.2×10^5 Pa) tubes would have a higher absolute efficiency than the 800 torr (1.07×10^5 Pa) tubes in disagreement with these results. This may be due to the electronics differences mentioned above. It would be of value to test BF_3 tubes at even lower pressures to see the dependence of efficiency on pressure. Higher pressures are of no interest since they require even higher voltages to operate, additional shipping limitations on pressurized cylinders come to play, and recombination in the gas ultimately limits the pressure.

7 Conclusions

The experimental measurements with BF_3 tubes at 800 torr (1.07×10^5 Pa) reported here indicate that two 800 torr (1.07×10^5 Pa) tubes may be sufficient to replace one three-atmosphere ^{3}He tube in a standard SAIC moderator box, but not for any lower BF_3 pressure. Because this conclusion is based upon results that appear just barely above the required level of performance, and because these results are affected by (un-avoidable) uncertainties in the source strengths, it may be prudent to assume that three one-atmosphere BF_3 tubes would be needed, as had been previously indicated. This would give a comfortable margin of error for the SAIC system. Higher pressure tubes have better efficiency but introduce other significant problems such as requiring higher voltage.

Another factor pointing towards the use of three tubes with lower pressure than 800 torr (1.07×10^5 Pa) is the voltage issue. The BF_3 tubes require a substantially higher voltage of about 2200 V to operate at 800 torr (1.07×10^5 Pa) compared to ^{3}He tubes at about 1000 V, which may be difficult to deploy to the field due to breakdown concerns in humid environments. Modifications to the SAIC electronics to operate these tubes would be necessary. Since BF_3 is toxic, there are also considerations about the acceptability of deploying this gas in the field.

It would be of value to explore the dependence of BF_3 efficiency as a function of gas pressure to lower pressures than tested so far.

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902 Battelle Boulevard
P.O. Box 999
Richland, WA 99352
1-888-375-PNNL (7665)
www.pnl.gov