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Performance Characterization of FB-Line Neutron Multiplicity Counter and Large Neutron Multiplicity Counter

N. Boyle

August 2025

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EXECUTIVE SUMMARY

Savannah River National Laboratory’s (SRNL) Nuclear Measurements group was tasked with characterizing the performance of two neutron multiplicity counters located at SRNL. Characterization measurements were made to determine the gate width, pre-delay, deadtime parameters, triples and doubles gate fractions, detector efficiency, and operating high voltage for the Large Neutron Multiplicity Counter (LNMC) and the FB Line Neutron Multiplicity Counter (FBLNMC). The parameters were determined, shown below, and were, as to be expected, slightly different than the previous calibrations, which were performed over 20 years ago. Several Pu samples were measured to validate the characterizations of the FBLNMC and LNMC. The measurements determined the sample Pu-240 mass within <2% deviation for the pure plutonium samples and ~8% for the mixed oxide sample. The pure Pu samples had significantly better accuracy compared with the impure mixed oxide sample due to the lack of induced fission or alpha,n neutrons from impurities. Overall, the characterization of the neutron multiplicity counters, and the determination of their operability has been completed successfully.

FBLNMC Calibration Parameter Comparison		
	Previous Calibration	SRNL Calibration
Efficiency	57.8%	58.4% ± 0.37%
Die-Away Time [μSec]	50.4	47.3
Pre-Delay [μSec]	3	3
Gate Width [μSec]	32	32
High Voltage [V]	1680	1680
Deadtime Coefficient A	0.2102	0.2102
Deadtime Coefficient B	0.002	0.002
Multiplicity Deadtime	50	50
Doubles Gate Fraction	0.4426	0.4447
Triples Gate Fraction	0.1919	0.1978

LNMC Calibration Parameter Comparison		
	Previous Calibration	SRNL Calibration
Efficiency	41.9%	41.7% ± 0.26%
Die-Away Time [μSec]	54.9	48.5
Pre-Delay [μSec]	3	3
Gate Width [μSec]	32	32
High Voltage [V]	1680	1680
Deadtime Coefficient A	0.0866	0.0866
Deadtime Coefficient B	0.0429	0.0429
Multiplicity Deadtime	24.5	24.5
Doubles Gate Fraction	0.4130	0.4249
Triples Gate Fraction	0.1820	0.1888

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1.0 Introduction

Savannah River National Laboratory’s (SRNL) Nuclear Measurements group was tasked with characterizing the performance of two neutron multiplicity counters located at SRNL. The detectors are referred to as the FB-Line neutron multiplicity counter (FBLNMC), and the Large Neutron Multiplicity Counter (LNMC), often referred to as the 30-gallon neutron multiplicity counter. The FB-Line neutron multiplicity counter “was developed to measure impure plutonium at the Westinghouse Savannah River Site FB-Line Facility” [2]. The FBLNMC was designed to have high thermal neutron detection efficiency, ~57%, at the time of fabrication with 113 He-3 tubes located concentrically in four rings around a central open sample chamber. The axial and radial profiles were designed to have less than 2% variability across the chamber allowing for measurements with a wide variety of geometries to be performed. The FBLNMC was designed to measure impure samples of plutonium with masses from a few tens of grams to several kilograms; this lower limit of detection will be important when comparing the performance with a plutonium standard discussed later. The effects of impurities on the sample measurements will also be discussed later. Figure 1-1 depicts the side and top-down views of the FBLNMC [2].

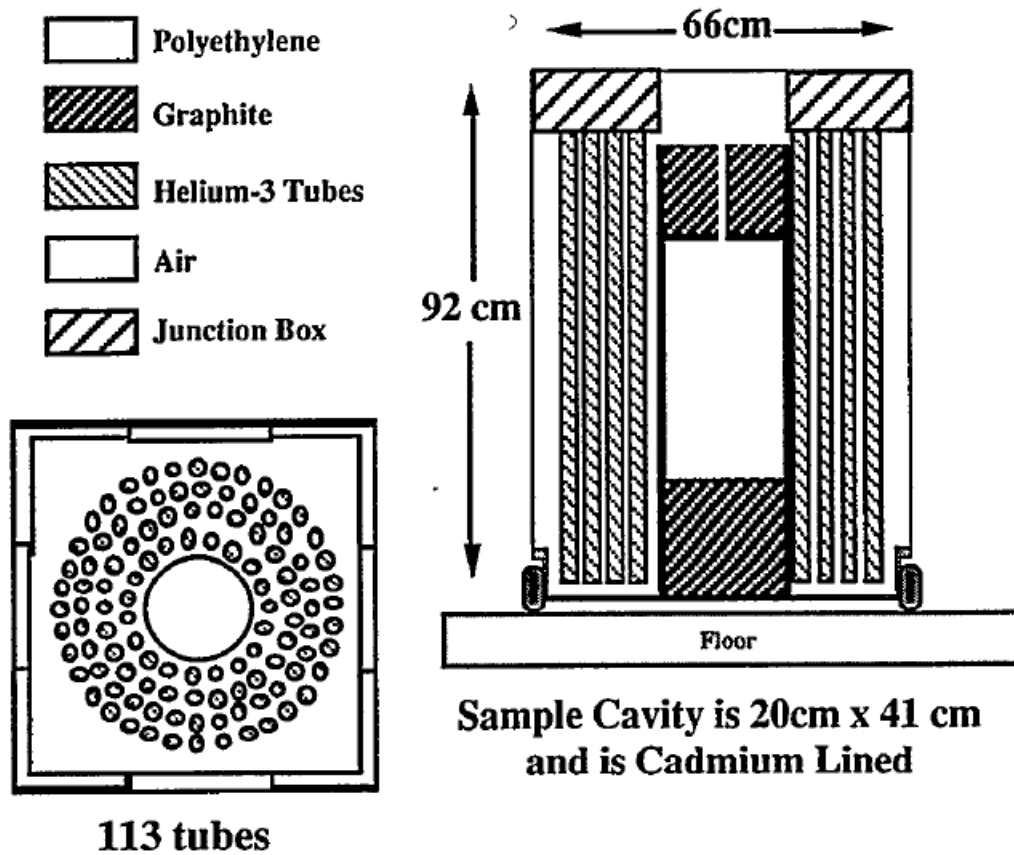


Figure 1-1: Side and Top-Down View of the FB-Line Neutron Multiplicity Counter [2].

The LNMC was designed to measure samples with significantly larger volumes such as 30-gallon drums or ATR400 long term storage containers, hence the nickname. The large volume samples of the LNMC required the design to load samples from the side instead of the top and therefore significant design changes were required. These design changes included a hexagonal shape for the moderation and aluminum corner reflectors to provide a uniform spatial response across the sample chamber. Additionally, the number of

He-3 tubes was reduced to three rings to save costs. The thermal neutron detection efficiency of the chamber at the initial characterization was 42% with a die-away time of 55 μ Sec. Figure 1-2 depicts the side and top-down view of the LNMC [2].

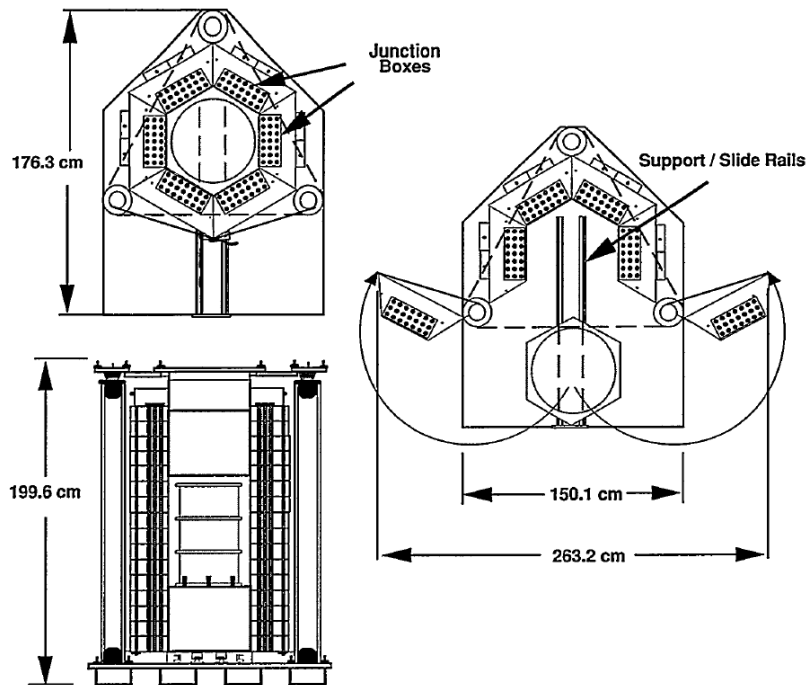


Figure 1-2: Side and Top-Down View of the Large Neutron Multiplicity Counter [2].

Two LNMC's were made, one is in Lawrence Livermore National Lab's (LLNL) Nuclear Materials Facility where it is used for inventory verification. This instrument has recently been suffering from an unknown issue that has caused degradation in the performance. The other LNMC was installed in the Rocky Flats Environmental Technology Site to assist the International Atomic Energy Agency (IAEA) in measuring excess weapons materials [2]. The instrument was transferred to SRNL. SRNL's nuclear measurements group was tasked with characterizing the FBLNMC to determine the operability, current detector calibration parameters (e.g. die-away time, gate fractions, pre-delay, efficiency, etc.), and accuracy of the FBLNMC. If the FBLNMC was still in operable conditions, it would be shipped to LLNL to be used as a replacement for their LNMC. Additionally, SRNL was tasked with characterizing the LNMC located at SRNL to determine if there were any issues associated with the system in hopes that it could help diagnose the issues plaguing the LLNL LNMC.

Neutron multiplicity is a defining characteristic of an isotope that undergoes fission, wherein the number of neutrons emitted during that fission event can be defined by a unique probability distribution function for that specific isotope. Because fission can be spontaneous or induced, the multiplicity distribution of the initiating event, i.e. spontaneous fission or induced fission, is also unique to that reaction for each isotope. It is necessary to know the spontaneous and induced multiplicity distributions for the isotopes of interest because these distributions are utilized to calculate the mass of the isotopes measured. Figure 1-3 shows the spontaneous multiplicity distribution of Pu-240, which is typically considered the main isotope of interest for Pu-239 content confirmation and verification, and the Pu-239 induced fission multiplicity distribution, a source of background for highly multiplying samples discussed further in this report [2].

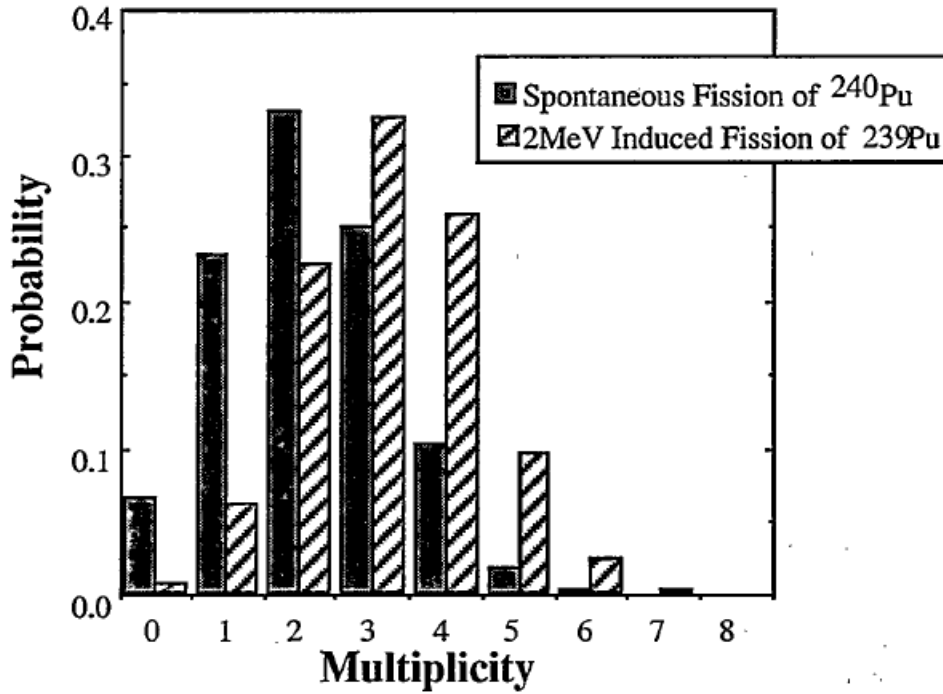


Figure 1-3: Spontaneous Fission Multiplicity Probability Distribution of Pu-240 and Induced Multiplicity Distribution of Pu-239.

Characterization of a neutron multiplicity counter is a measurement intensive procedure that requires a wide variety of neutron sources with low, and/or well-known, neutron emission from alpha,n reactions. Californium-252, a spontaneous fission (~3% branching ratio) neutron emitter, has been traditionally used as a calibration source in neutron multiplicity measurements because it has been extensively studied, the spontaneous and induced moments of multiplicity are well known, i.e. isotopic constants that define the probability of neutrons emitted per fission from either induced or spontaneous fission decay events, and the Cf-252 sources are geometrically similar even with varying levels of activity which allows for no bias from the point-model assumption traditionally used in multiplicity measurements. SRNL as part of this characterization procured 10 Cf-252 sources from Eckert & Ziegler with activities ranging from 0.3 – 100 uCi as of 11/15/2024. Cf-252 has a half-life of 2.645 years which impacts some of the measurements up to 12% depending on the time of the final measurement acquisition date, therefore the measurements discussed in this report are all decay corrected to the certificate date.

Briefly, neutron multiplicity counters operate by measuring the number of coincident thermal neutron detections within a time gate, known as the gate width, and comparing that coincidence with the uncorrelated background via the Rossi- α distribution. The Rossi- α distribution was developed to assess statistical fluctuations in nuclear reactor noise and has a relatively average distribution with respect to time if no fission neutrons are present [2]. If fission neutrons are present, the excess in the distribution is defined by the exponential die-away of the fission neutron signature due to the time required for moderation to thermal energies and subsequent absorption in the detector/moderating materials. Figure 1-4 shows a histogram of the Rossi- α distribution associated with a neutron detection stream from a source with correlated fission neutrons [2].

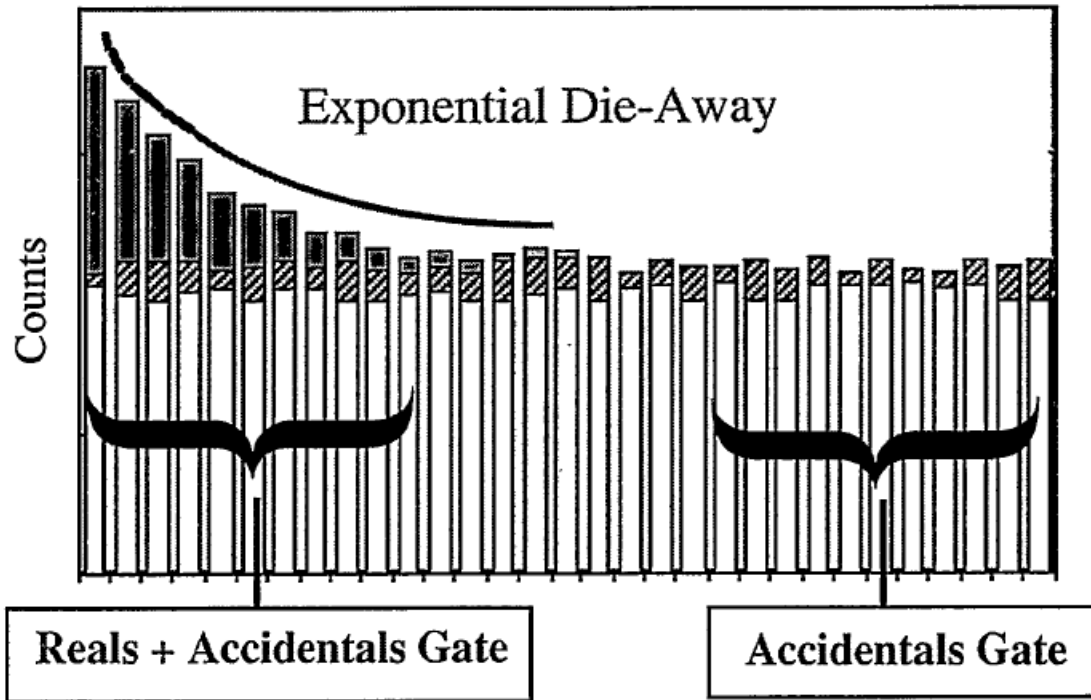


Figure 1-4: Histogram of a Rossi- α distribution associated with a neutron detection stream with a source of correlated fission neutrons [2].

Neutron multiplicity counters use a complex set of electronics called a shift register to read the neutron detection pulse stream and convert the stream into a usable set of measurements known as singles, doubles, and triples. The shift register utilized for all measurements was a Mirion JSR-15 Handheld Multiplicity Register (HHMR). Note that the doubles and triples are not a direct correlation to the number of neutrons emitted in coincidence i.e. a measurement of 20 doubles per second does not mean 20 fission events emitting 2 neutrons per event. The singles however are a direct correlation to the number of neutrons detected per second. An in-depth discussion on neutron multiplicity analytical derivations and electrical operations is beyond the scope of this document, however further information on neutron multiplicity counting can be found in the “Application Guide to Neutron Multiplicity Counting” located in the E-Notebook under experiment M5443-00686-08 [2]. For now, understanding that the singles, doubles, and triples rates are utilized to calculate the mass of plutonium in a sample is sufficient for this scope.

As previously mentioned, characterization of a neutron multiplicity counter requires extensive measurements to determine the proper operating parameters. The important operating parameters for a neutron multiplicity counter include the gate-width, pre-delay, dead-time coefficients, efficiency, doubles gate fraction, and triples gate fraction. The multiplication and α parameters are also important; however, these parameters are sample dependent not detector dependent.

The gate-width is the length of time the shift register is open for coincidence detection and is chosen based on the die-away time in the detector. The die-away time is the amount of time it takes for a neutron born from fission to moderate and be absorbed in the detector. It is detector specific and would need to be measured for both the FBLNMC and the LNMC. The die-away time is assumed to be a single exponential equation, which is a valid assumption for the ranges of source activities considered in this report. The optimal gate width is 1.27τ , where τ is the die-away time in microseconds. The gate width is a “very shallow

minimum” and is often rounded to the nearest multiple of 2 [2]. Additionally, the optimum gate width is divided in this report by a factor of 2 to reduce the effect of dead time on the triples rate, a common procedure in neutron multiplicity counting. Measurements to determine the die-away time will be discussed later in this report.

The pre-delay is the delay between time zero and the initial time gate for coincidence measurements. The pre-delay is required to reduce the bias from pulse pileup effects and amplifier recovery times. If the pre-delay was zero, the pulse-pileup effects would cause a bias and if the pre-delay is not long enough for the amplifier to recover there would also be a bias. It had previously been determined that for count rates <500,000 neutrons per second a pre-delay of 3 μ Sec is sufficient for the Amptek-111 amplifiers; the amplifiers utilized in the FBLNMC and the LNMC. This pre-delay time was confirmed and is discussed further in this report.

The dead-time coefficients are parameters that are utilized by the controlling software and shift register to correct for losses due to amplifier recovery from dead-time. For this report, the author decided to assume the dead-time parameters from the initial detector calibrations were correct due to the intensive process of calculating these values. As discussed later, this is a valid assumption, and the dead-time parameters successfully corrected for the loss in efficiency due to the dead-time effects.

The detector efficiency is a calculated value determined from a calibrated standard that is typically used to calculate the activity of unknown sources. The detector efficiency for neutron multiplicity counters is defined as the total number of detection events, the singles, divided by the output of the calibration standard. For this report Equation 1-1 was used to calculate the detection efficiency for the FBLNMC and the LNMC.

$$\epsilon = \frac{S}{A * (3.7E4 * 3.757 * 0.031028 * e^{(-0.000718*(t_m - t_c)})})} \quad \text{Equation 1-1}$$

Where ϵ is the efficiency of the detector, S is the singles rate in counts per second, and A is the source activity in uCi, t_m is the date of the measurement and t_c is the date of the calibration (11/15/2024). The constant values are detailed in the following: 3.7E4 is the conversion from uCi to Bq, 3.757 is the average number of neutrons per fission from a Cf-252 spontaneous fission decay event, 0.031028 is the branching ratio for Cf-252 spontaneous fission decay, and 7.18E-4 is the decay constant for Cf-252 in days⁻¹. The efficiency is used in the International Neutron Coincidence Counting (INCC) software to calculate the Pu content and is also used to calculate the doubles and triples gate fraction discussed further in this report.

The triples gate fraction is the probability that three neutrons emitted in coincidence from a fission event would be detected as a coincidence. Similarly, the doubles gate fraction is the probability that two neutrons emitted in coincidence from a fission event would be detected as a coincidence. The doubles and triples gate fractions can be determined using equations 1-2 and 1-3 respectively.

$$f_d = \frac{2 * \nu_{s1} * D}{\epsilon * \nu_{s2} * S} \quad \text{Equation 1-2}$$

$$f_t = \frac{3 * f_d * \nu_{s2} * T}{\epsilon * \nu_{s3} * D} \quad \text{Equation 1-3}$$

Where f_d is the doubles gate fraction, f_t is the triples gate fraction, ν_{s1} , ν_{s2} , ν_{s3} are the first, second and third moment of multiplicity for Cf-252 respectively, ϵ is the detector efficiency, and S, D, and T are the singles, doubles, and triples rate.

The neutron population in neutron multiplicity counters consists mainly of neutrons emanating from spontaneous fission, induced fission, and alpha,n reactions. The latter two reactions are sources of background in the system and are considered excess neutrons. Additionally, neutrons are absorbed in the sample or surrounding detector moderation material leading to a loss in the neutron population. These sources of excess and loss in the neutron population contribute to a term known as the leakage multiplication. The leakage multiplication is essentially, the overall change in neutron population due to the sample composition and geometry when placed inside the neutron multiplicity counter. The leakage multiplication will be used to calculate the fission rate of the sample later in this report. Furthermore, the ratio of alpha,n reactions to spontaneous fission reactions is known as the α parameter. Materials with large amounts of impurities and oxides have a large value, whereas materials such as Pu metal and Cf-252 have lower or near zero α value. The variability in the α value for samples can dramatically impact the final measurement results, however effects of the α parameter on the sources considered for this report are negligible.

2.0 Detector Characterization

2.1 High Voltage Plateau Measurements

Neutron multiplicity counters use He-3 detectors to detect thermal neutrons being absorbed in the $\text{He}^3(n,p)\text{H}^3$ reaction which releases 765 keV per reaction. The pulse height spectrum from this interaction in an He-3 detector is shown in Figure 2-1 [3].

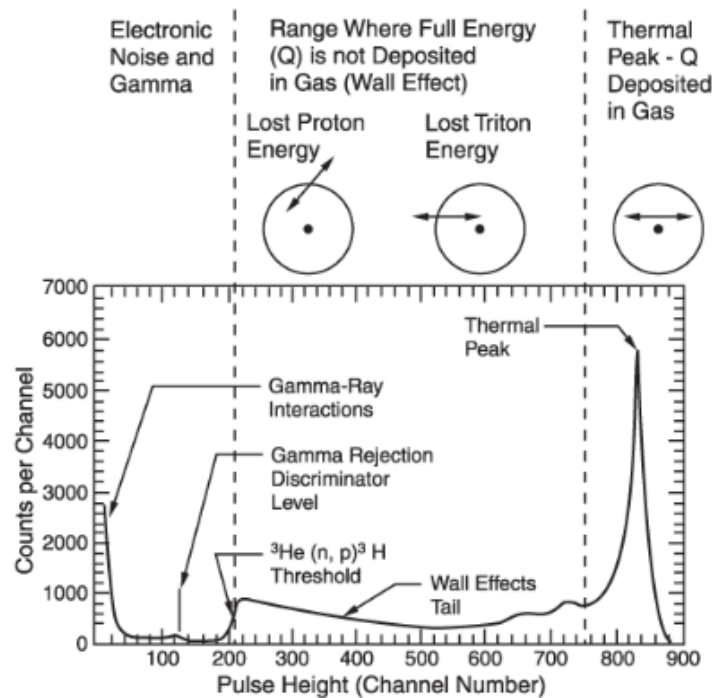


Figure 2-1: Pulse Height Spectrum for a Thermal Neutron Absorption Reaction in He-3.

He-3 detectors are gas proportional detectors and therefore require high voltage to operate. Given a constant amplifier gain, such as the case for the neutron multiplicity counters used in this report, the high voltage of the detector will shift the height and location of the thermal peak. Figure 2-2 shows the pulse height shift for a Reuter Stokes He-3 gas proportional detector with a Quaesta NPM3100E multi-channel analyzer at SRNL at varying high voltages.

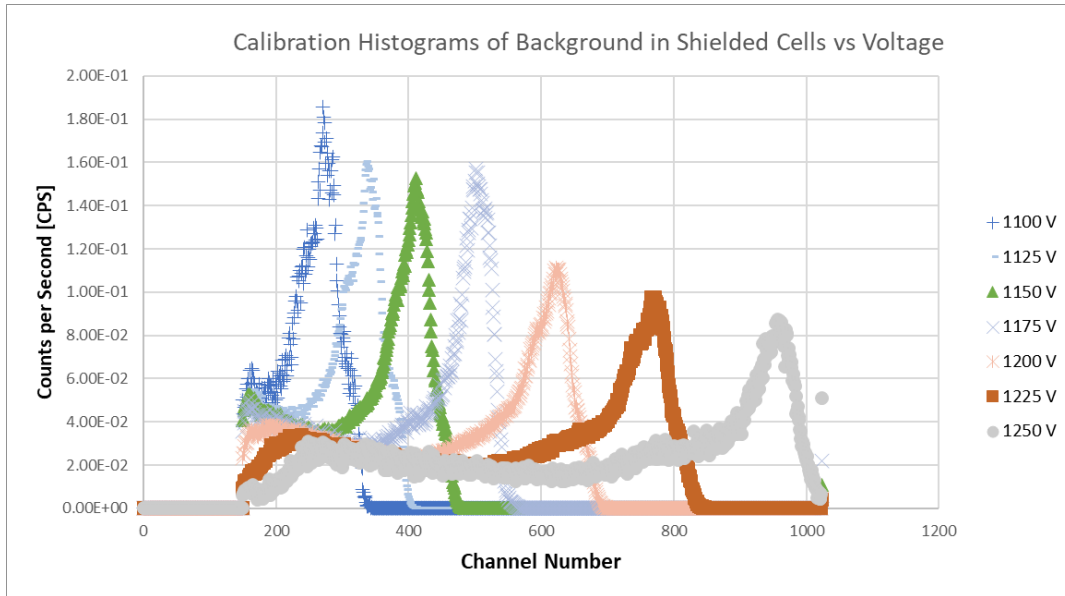


Figure 2-2: Pulse Height Distribution vs Voltage for He-3 Detector Placed in SRNL Shielded Cells

The height and location of the thermal peak shifts to the right as high voltage increases. Unfortunately, so does the gamma interaction peak. He-3 detectors can reject gamma radiation up to a certain exposure rate. However, beyond that threshold, pulse-pileup effects, or multiple gamma rays can deposit their energy within the detector's timing resolution. The use of a lower-level discriminator is required to mitigate or eliminate the signal from gammas interfering with the detector neutron pulse output stream. Neutron multiplicity counters only analyze the amplified pulses and do not process the pulse height through a multi-channel analyzer. Therefore, great care should be taken to ensure that the thermal peak is well above any lower-level discriminator in the system and the voltage is below the gamma interference voltage.

High voltage calibration measurements were performed in the FBLNMC and LMNC using a 40 uCi Cf-252 source. The Cf-252 source was screwed into a thin aluminum holder and placed into an aluminum "top hat" to restrict the movement of the holder in the detector and ensure precise placement between detectors. The location of the source in the FBLNMC and LMNC detectors was approximately the center of the open space in the chamber. After the source was placed inside, the detector lid was replaced, or the doors were closed for the FBLNMC and LMNC, respectively. The INCC was used to change the parameters for the high voltage. The high voltage was swept from 1400 to 1900 V in increments of 20 volts and a 2-minute measurement was taken at each high voltage. After the initial high voltage plateau sweep, the input of the HHMR was swapped from the sum signal output of the FBLNMC to an individual ring output and the high voltage plateau sweep was performed again. The output swapping and high voltage measurement was repeated until all 4 rings and the sum signal outputs of the FBLNMC were measured. Similarly, the input of the HHMR was swapped from the sum signal output of the LMNC to an individual ring for each junction box. The output swapping and high voltage measurement was repeated until all six junction boxes with three rings each were measured. Figure 2-3 shows the results of the high voltage calibration curve for the

FBLNMC. Figures 2-4 and 2-5 show the high voltage calibration curve for the LNMN sum signal output and individual junction box ring outputs respectively.

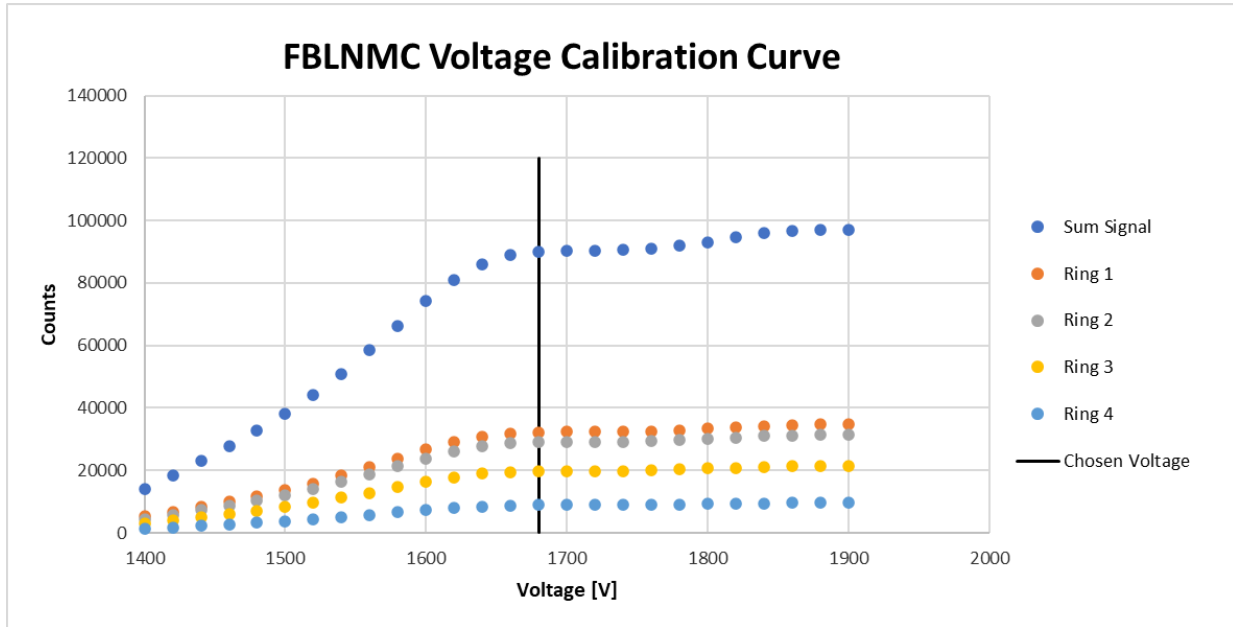


Figure 2-3:FBLNMC High Voltage Sum Signal and Individual Ring Calibration Curve.

The proper operating voltage for an He-3 detector is right after the first bend before the first plateau. The proper operating voltage for the FBLNMC occurs at 1680 V, the previously determined high voltage for the FBLNMC.

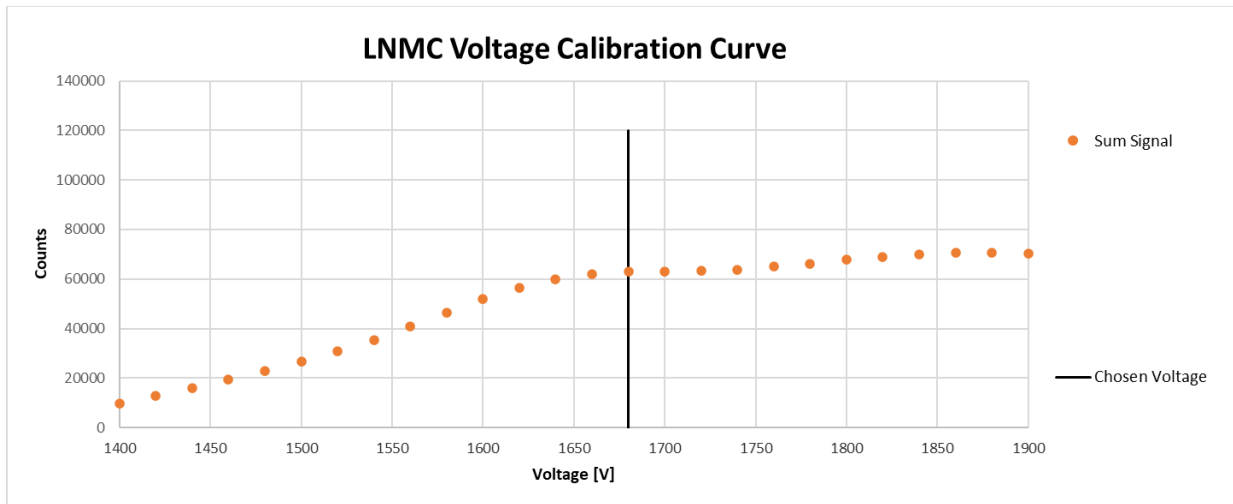


Figure 2-4: LNMN High Voltage Sum Signal Calibration Curve.

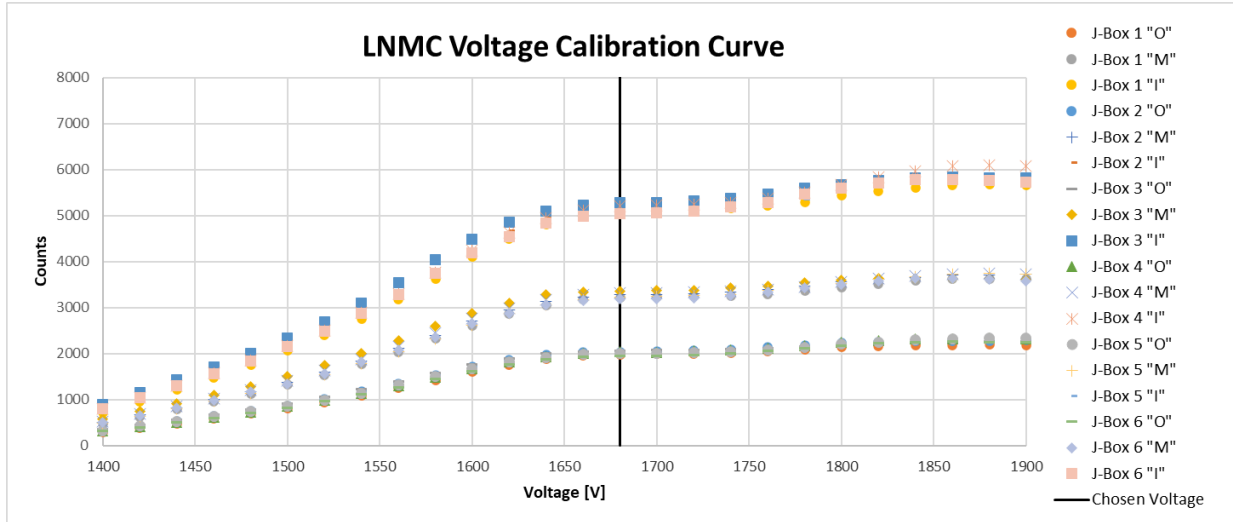


Figure 2-5: LNMC High Voltage Individual Ring Calibration Curve.

The proper operating voltage for the LNMC occurs at 1680 V, the previously determined high voltage for the LNMC. Overall, there was no significant deviation from any individual detector ring or junction box in the FBLNMC and LMNC therefore the He-3 detectors were determined to be functioning properly.

2.2 Background Measurements

A background measurement was performed in the FBLNMC and LNMC for 10 min (20 cycles of 30 seconds) prior to or after any set of calibration measurements. The background measurement was subtracted from the measurements for the respective detector’s characterization measurements discussed below. The verification samples were measured for 1000 minutes (2000 cycles 30 seconds), therefore a 1000-minute background was taken prior to each sample measurement to obtain better statistical uncertainty on the sample results.

2.3 Die-Away Time Measurements

Die-Away time measurements were performed in the FBLNMC and LMNC using a 40 uCi Cf-252 source. The Cf-252 source was screwed into a thin aluminum holder and placed into an aluminum “top hat” to restrict the movement of the holder in the detector and ensure precise placement between detectors. The location of the source in the FBLNMC and LMNC detectors was approximately the center of the open space in the chamber. After the source was placed inside, the detector lid was replaced, or the doors were closed for the FBLNMC and LNMC, respectively. The INCC was used to change the parameters for the gate-width at a fixed pre-delay for both detectors. The pre-delay was set at 3.0 μ Sec and a 10-minute measurement (20 cycles of 30 seconds) was started at each gate width listed: 8, 16, 24, 32, 48, 64, 80, 96, 128, 175, 250, 500 μ Sec. A linear regression using Microsoft Excel’s internal solver was performed with the doubles rate using an exponential fit in the form of equation 2-1 to minimize the difference between the fitted data and the measured data.

$$a * (1 - \exp\left(-\frac{GW}{\tau}\right)) \quad \text{Equation 2-1}$$

Where a is a scaler constant, τ is the die away time, and GW is the gate width in μSec . Figures 2-6 and 2-7 show the results of the linear regression fit and measured data for the FBLNMC and LNMC, respectively. Note: the LNMC has a metallic insert used to hold large drums in place while measuring, the effects of this insert were investigated by performing these measurements with and without the insert.

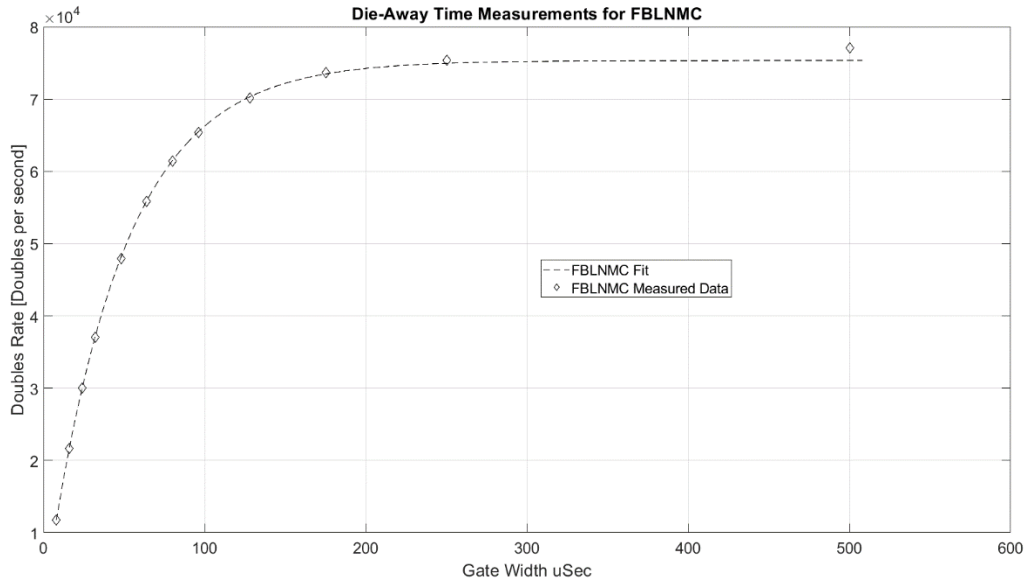


Figure 2-6: Die-Away Time Measurements for the FBLNMC.

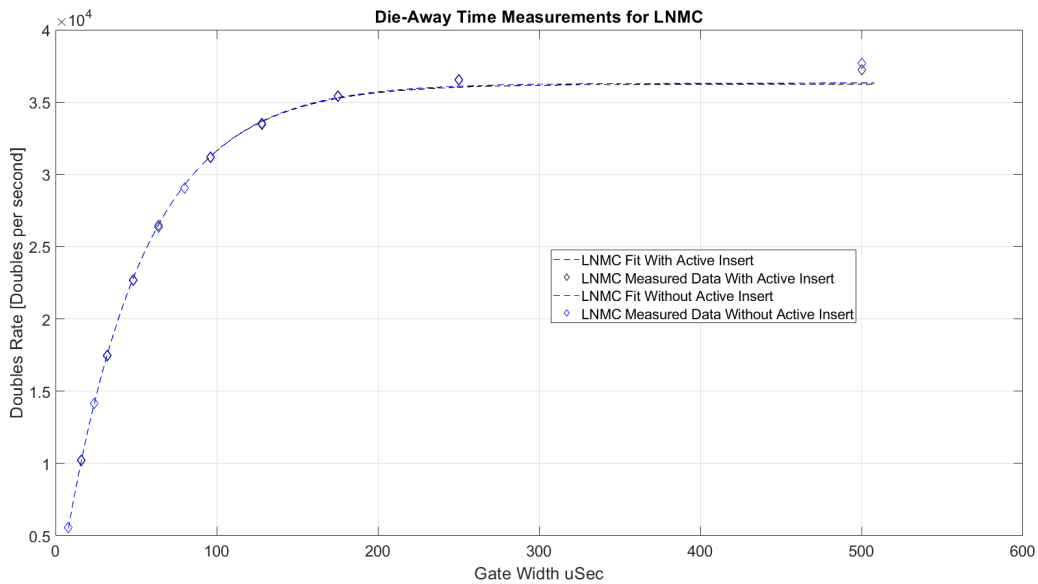


Figure 2-7: Die-Away Time Measurements for the LNMC.

The calculated exponential fits align well with the measured data up to the largest gate width. The slight deviation at a gate width of 500 μSec is an artifact of the one-term exponential fit assumption. A second term would be required to correct for this deviation, however in the typical region of operation ($<128 \mu\text{Sec}$) a one-term assumption is clearly valid.

The die-away time determined for the FBLNMC was 47.2 μSec and the LNMC with and without the active insert was 48.5 and 48.8 μSec , respectively. The die-away time for the LNMC with the active insert is slightly lower than the die-away time without. This is to be expected due to the additional moderation/absorption material in the chamber reducing the overall time to absorption. As mentioned previously, the optimal gate width is the measured die-away time multiplied by 1.27 and rounded to the nearest power of 2. For the FBLNMC and the LNMC the optimal gate width is therefore 64 μSec , the gate width was divided by a factor of 2 to reduce the effect of dead-time on the triples rate. The final selected gate width for both the FBLNMC and LNMC is 32 μSec .

2.4 Pre-Delay Justification

Pre-delay measurements were performed in the FBLNMC and LMNC using a 40 uCi Cf-252 source. The Cf-252 source was screwed into a thin aluminum holder and placed into an aluminum “top hat” to restrict the movement of the holder in the detector and ensure precise placement between detectors. The location of the source in the FBLNMC and LMNC detectors was approximately the center of the open space in the chamber. After the source was placed inside, the detector lid was replaced, or the doors were closed for the FBLNMC and LNMC, respectively. The INCC was used to change the parameters for the pre-delay at a fixed gate width for both detectors. The gate width was set at 8.0 μSec and a 10-minute measurement (20 cycles of 30 seconds) was started at each pre-delay listed: 0.5, 1, 1.5, 2, 2.5, 3, 4, 5, 6, 7, 8, 9, 10, μSec . The maximum normalized results of the pre-delay sweep for the FBLINM and LNMC are shown in Figure 2-8. The pre-delay for both the LNMC and FBLNMC was chosen to be 3.0 μSec . Neutron multiplicity counters in the industry typically operate with a pre-delay of either 3 or 4 μSec [4].

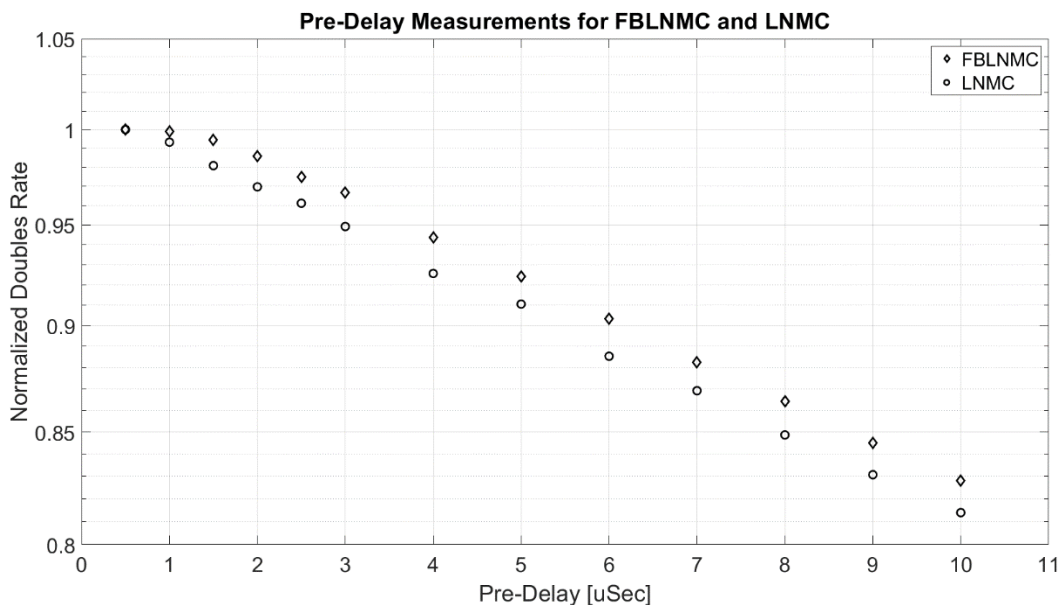


Figure 2-8: Pre-Delay Measurements for the FBLNMC and LNMC.

The decrease in the doubles rate versus pre-delay time is evident from Figure 2-8. Additionally, this trend is seen in the triples rate. Therefore, the pre-delay should be minimized to reduce the loss associated with

a longer gate time. Additionally, the uncertainties with the measurements should be minimized to increase overall detector performance. Table 2-1 shows the normalized uncertainties, and their average, associated with the doubles and triples rate for the FBLNMC and LNMC. Note: the uncertainty is normalized to the maximum value for triples and doubles rates of the 0.5 to 10 μ Sec data set and is used to compare between the doubles and triples uncertainty due to the scale difference in doubles and triples rates.

Table 2-1: Doubles, Triples, and Average Normalized Uncertainty Values for the FBLNMC and LNMC.

Pre-Delay μ Sec	FBLNMC			LNMC		
	Normalized Doubles Uncertainty	Normalized Triples Uncertainty	Average Normalized Uncertainty	Normalized Doubles Uncertainty	Normalized Triples Uncertainty	Average Normalized Uncertainty
3	0.722597	0.532431899	0.627515	0.799144	0.604775	0.70196
4	1	0.563998344	0.781999	0.869874	0.533927	0.701901

The FBLNMC has an uncertainty minimum at a pre-delay of 3.0 μ Sec but the LNMC has a slightly, near negligible, higher uncertainty at 3.0 μ Sec. Because the doubles and triples rates drop significantly between 3 and 4 μ Sec, it was decided that the negligible gain in uncertainty did not outweigh the doubles/triples measurement loss and therefore a pre-delay of 3 μ Sec was also chosen for the LNMC.

2.5 Dead-Time Coefficients and Detector Efficiency

The author noted that the pre-delay and gate-width did not change from the initial calibration so to save significant time in the characterization process it was decided to initially assume the dead-time coefficients would not change as well. To test this assumption, dead-time measurements were performed with and without the associated dead-time coefficient for each detector previously determined from the initial calibration.

Dead time measurements were performed with Cf-252 sources with overall sample activities ranging from 0.3 to 500 uCi. The Cf-252 sources were screwed into a thin aluminum holder and placed into an aluminum “top hat” to restrict the movement of the holder in the detector and ensure precise placement between sources and detectors. As mentioned previously, the Cf-252 sources ranged in activity from 0.3 – 100 uCi therefore to achieve higher activity samples, e.g. 200, 300, 400, 500 uCi, multiple 100 uCi Cf-252 sources were affixed to the aluminum holder. The aluminum holders with multiple Cf-252 sources were designed to hold the Cf-252 symmetrically to eliminate any bias associated with geometrical variability in the detector chambers. The location of the sources in the FBLNMC and LMNC detectors was approximately the center of the open space in the chamber. After the source was placed inside, the detector lid was replaced, or the doors were closed for the FBLNMC and LNMC, respectively. The pre-delay and gate width were set at 3.0 and 32 μ Sec, respectively, in the INCC and 10-minute measurements (20 cycles of 30 seconds) were performed with the following Cf-252 activities: 0.3, 1, 5, 10, 40, 100, 200, 300, 400, 500 uCi. The deadtime coefficients for the FBLNMC and LNMC are given in Table 2-2.

Table 2-2: INCC Deadtime Parameters used for FBLNMC and LNMC characterization.

INCC Deadtime Parameter	FBLNMC	LNMC
Multiplicity	50.0000	24.5000
Coefficient A	0.2102	0.0866
Coefficient B	0.0020	0.0429
Coefficient C	0.0000	0.0000

The triples/doubles and doubles/single ratios should be constant with increasing Cf-252 source strength, unless deadtime is not accurately corrected. Figures 2-9 and 2-10 depict the triples/doubles and doubles/singles ratio for the FBLNMC respectively.

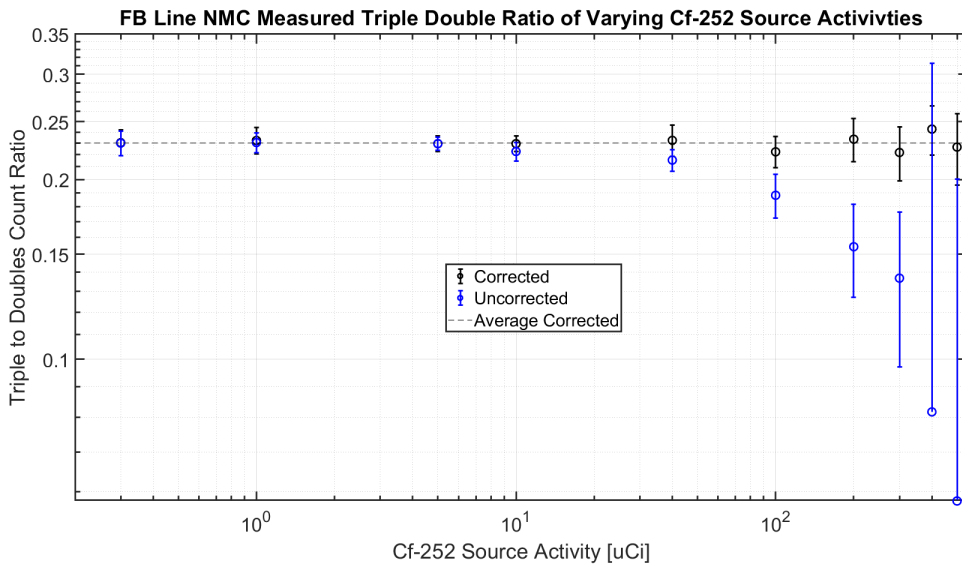


Figure 2-9: FBLNMC Measured Triples/Doubles Ratio for Varying Cf-252 Source Activity.

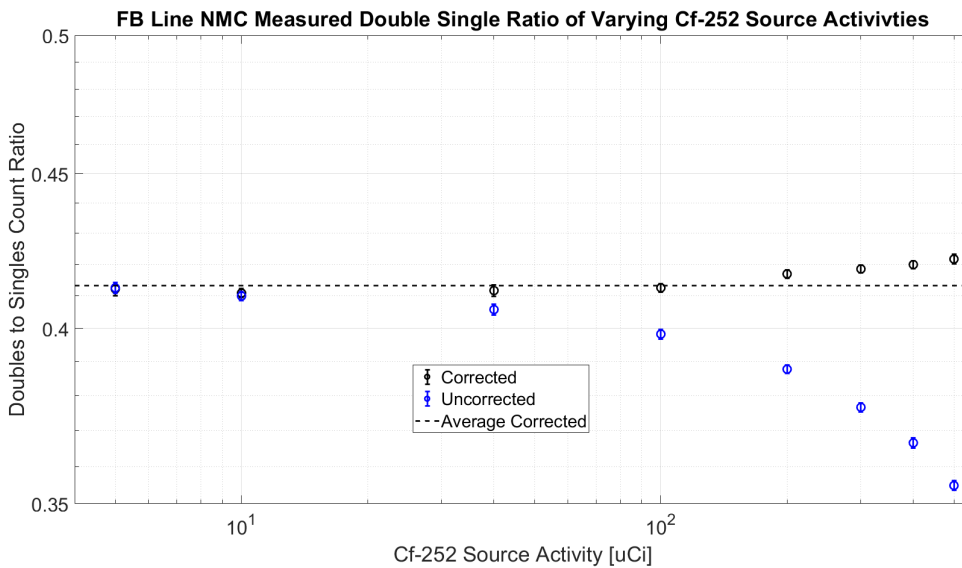


Figure 2-10: FBLNMC Measured Triples/Doubles Ratio for Varying Cf-252 Source Activity.

The corrected triples/doubles ratio of the FBLNMC is constant with source activity within uncertainties and the doubles/singles ratio is constant up to activities of around 100 uCi. This may be an overcorrection in the dead time for the doubles or singles, however neutron fluxes associated with Cf-252 source activities above 100 uCi (>400,000 neutrons per second in 4π) are rare in the neutron multiplicity measurement field and if necessary, a doubles/singles correction factor can be made from the data collected. Overall, the data in Figures 2-4 and 2-5 demonstrate that the deadtime coefficients for the FBLNMC are working correctly therefore the assumption that they have not changed is valid.

Similarly, Figures 2-11 and 2-12 depict the triples/doubles and doubles/singles ratio for the LNMC respectively.

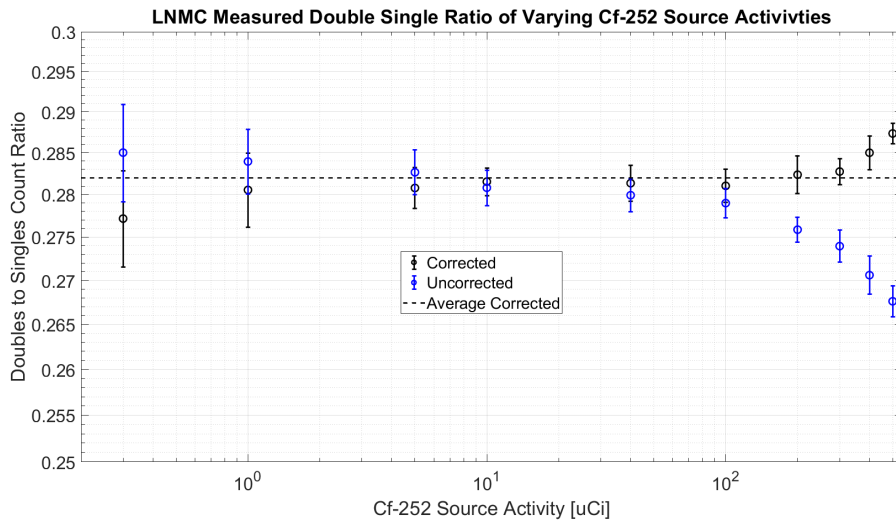


Figure 2-11: LNMC Measured Triples/Doubles Ratio for Varying Cf-252 Source Activity.

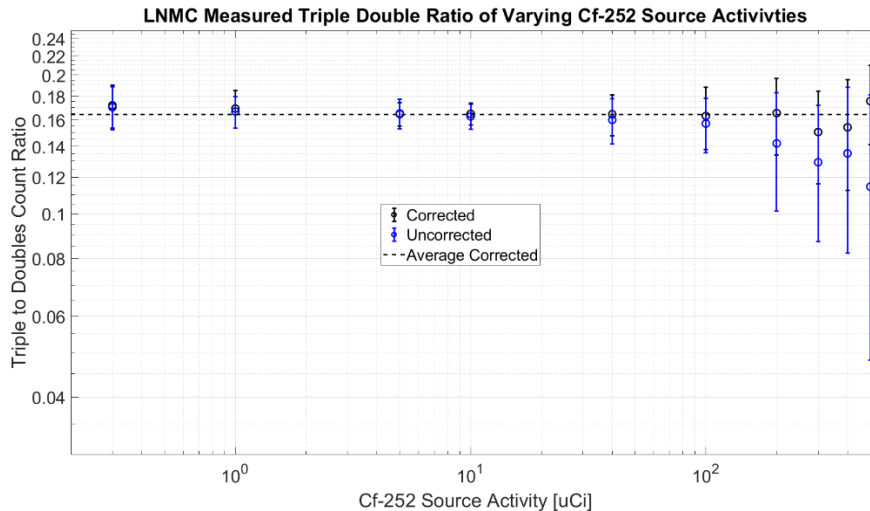


Figure 2-12: LNMC Measured Triples/Doubles Ratio for Varying Cf-252 Source Activity.

The corrected triples/doubles ratio of the FBLNMC is constant with source activity and the doubles/singles ratio is constant up to activities of around 300 uCi. As noted previously, this should not be an issue for traditional samples. Overall, the data in Figures 2-11 and 2-12 demonstrate that the deadtime coefficients for the LNMC are working correctly therefore the assumption that they have not changed is valid.

The efficiency of the detector can be easily calculated from the measurements performed for the deadtime coefficients validation. The data taken for the FBLNMC and LNMC were used with Equation 1-1 to calculate an efficiency at each activity. The wide range of source activities enables an average efficiency calculation that spans the entire range of expected measurements in the future. Table 2-3 gives the efficiency for each standard, the associated uncertainty and the average efficiency for the FBLNMC and LNMC.

Table 2-3: Measured Efficiency for Each Cf-252 Sample Activity Measured in the FBLNMC and LNMC.

Cf-252 Activity [uCi]	FBLNMC		LNMC	
	Efficiency	Efficiency 1 σ Uncertainty	Efficiency	Efficiency 1 σ Uncertainty
0.3	62.8%	1.3%	44.8%	0.91%
1	55.1%	1.1%	39.4%	0.79%
5	56.2%	1.1%	40.4%	0.81%
10	62.0%	1.2%	44.5%	0.89%
40	56.9%	1.1%	40.7%	0.81%
100	58.0%	1.2%	41.4%	0.83%
200	58.4%	1.2%	41.5%	0.83%
300	58.0%	1.2%	41.2%	0.82%
400	58.1%	1.2%	41.4%	0.83%
500	58.2%	1.2%	41.6%	0.83%
Average	58.4%	0.37%	41.7%	0.26%

The previous calibration for the FBLNMC and the LNMC measured an efficiency of 57.8% and 41.9% respectively. The current calibration measured an efficiency of 58.4% \pm 0.37% and 41.7% \pm 0.26% for the FBLNMC and LNMC respectively [5,6]. The currently measured FBLNMC efficiency and the currently measured LNMC efficiency were within 2 σ uncertainty of the previous calibration. Note: uncertainties for the previous calibrations could not be identified.

2.6 Doubles and Triples Gate Fraction

The doubles and triples gate fractions can be easily calculated once the efficiency of the detector is known. The Cf-252 measurement results from the deadtime coefficients measurements and Equations 1-2 and 1-3 were utilized to calculate the doubles and triples gate fraction for the FBLNMC and the LNMC. Table 2-4 gives the doubles and triples gate fraction for the FBLNMC and LNMC.

Table 2-4: Doubles and Triples Gate Fraction for the FBLNMC and LNMC.

Cf-252 Activity	FBLNMC		LNMC	
	Doubles Gate Fraction	Triples Gate Fraction	Doubles Gate Fraction	Triples Gate Fraction
0.3	0.4301	0.1981	0.4176	0.1975
1	0.4394	0.1997	0.4227	0.1944
5	0.4436	0.1973	0.4231	0.1891
10	0.4422	0.1973	0.4242	0.1893
40	0.4431	0.1998	0.4239	0.1890
100	0.4440	0.1912	0.4235	0.1874
200	0.4487	0.2007	0.4255	0.1900
300	0.4504	0.1907	0.4260	0.1729
400	0.4519	0.2087	0.4295	0.1770
500	0.4538	0.1947	0.4330	0.2018
Average	0.4447	0.1978	0.4249	0.1888

2.7 Comparison to Previous Calibration

The FBLNMC and LNMC have undergone extensive calibration and characterization. Comparisons of the results for the FBLNMC operational parameters and the LNMC operational parameters with the previous and SRNL calibration are shown in Tables 2-5 and 2-6 respectively [5,6].

Table 2-5: Comparison of Previous and Current Calibration Parameters for the FBLNMC.

FBLNMC Calibration Parameter Comparison		
	Previous Calibration	SRNL Calibration
Efficiency	57.8%	58.4% ± 0.37%
Die-Away Time [μSec]	50.4	47.3
Pre-Delay [μSec]	3	3
Gate Width [μSec]	32	32
High Voltage [V]	1680	1680
Deadtime Coefficient A	0.2102	0.2102
Deadtime Coefficient B	0.002	0.002
Multiplicity Deadtime	50	50
Doubles Gate Fraction	0.4426	0.4447
Triples Gate Fraction	0.1919	0.1978

Table 2-6: Comparison of Previous and Current Calibration Parameters for the LNMC.

LNMC Calibration Parameter Comparison		
	Previous Calibration	SRNL Calibration
Efficiency	41.9%	41.7% ± 0.26%
Die-Away Time [μSec]	54.9	48.5
Pre-Delay [μSec]	3	3
Gate Width [μSec]	32	32
High Voltage [V]	1680	1680
Deadtime Coefficient A	0.0866	0.0866
Deadtime Coefficient B	0.0429	0.0429
Multiplicity Deadtime	24.5	24.5
Doubles Gate Fraction	0.4130	0.4249
Triples Gate Fraction	0.1820	0.1888

Overall, the calibration parameters for the FBLNMC and the LNMC have not dramatically changed since the previous calibrations.

2.8 Verification of Operational Parameters

Verification of the operational parameters for the FBLNMC and LNMC was performed by measuring a mixed U/Pu sample. The sample mass distribution is listed in Table 2-7.

Table 2-7: Mass Distribution of Mixed U/Pu Sample for Detector Parameter Verification.

Isotope	Mass [g]
Pu-239	6.92
Pu-240	0.26
U-235	1.04
U-238	40.75

Mixed oxide samples are notoriously difficult to measure via traditional neutron multiplicity measurements due to the additional U-238/U-235 content that causes an increase in the coincidence signals due to the induced fission of U-238 and U-235. At the time of characterization, the only Pu material available to the author was the previous sample due to a hold placed on all internal transfers on radioactive material in SRNL. The sample was measured in the LNMC and FBLNMC for 10 min (20 cycles of 30 seconds).

The fission rate of the sample can be determined from Equations 2-2 through 2-7 below. The derivations of the analytical solutions for the equations are beyond the scope of this report but can be found in Reference 1.

$$a = \frac{-6 * T * \nu_{s2} * (\nu_{i1} - 1)}{\epsilon^2 * f_t * S * (\nu_{s2} * \nu_{i3} - \nu_{s3} * \nu_{i2})}$$

Equation 2-2

$$b = \frac{2 * D * [v_{s3} * (v_{i1} - 1) - 3 * v_{s2} * v_{i2}]}{\epsilon * f_d * S * (v_{s2} * v_{i3} - v_{s3} * v_{i2})}$$

Equation 2-3

$$c = \frac{6 * D * v_{s2} * v_{i2}}{\epsilon * f_d * S * (v_{s2} * v_{i3} - v_{s3} * v_{i2})} - 1$$

Equation 2-4

$$a + b * M + c * M^2 + M^3 = 0$$

Equation 2-5

$$F = \frac{\left[\frac{2 * D}{\epsilon * f_d} - \frac{M * (M - 1) * v_{i2} * S}{v_{i1} - 1} \right]}{\epsilon * M^2 * v_{s2}}$$

Equation 2-6

$$m_{240_{eff}} = \frac{F}{473.5 \frac{\text{fissions}}{\frac{\text{S}}{\text{g}}}}$$

Equation 2-7

Where T is the triples rates in triples per second, v_{s2} and v_{s3} are the second and third moment of multiplicity for spontaneous fission of Pu-240, respectively, v_{i1} , v_{i2} and v_{i3} are the first, second, and third moment of multiplicity for fast neutron induced fission of Pu-240, ϵ is the detector efficiency, f_t is the triples gate fraction, S is the singles rate in singles per second, D is the doubles rate in doubles per second, f_d is the doubles gate fraction, a, b, and c are coefficients used to calculate M, M is the leakage multiplication coefficient, F is the calculated number of fissions per second of the measured sample, and $m_{240_{eff}}$ is the effective mass of Pu-240 measured by the detector in grams. The measured M for the mixed Pu sample for the FBLNMC and LNMC was 1.0148 and 1.0150, respectively.

The previously determined detector parameters and the results for the singles, doubles, and triples rates for the sample measured in the FBLNMC and LNMC were utilized with Equations 2-2 through 2-7 to calculate an effective Pu-240 mass in grams of $0.2782 \pm 1.32\%$ and $0.2801 \pm 3.02\%$ respectively. The uncertainties quoted are the random 1 sigma relative counting uncertainty and do not include systematic uncertainty. The Pu-240 mass in the sample was expected to be 0.26 g, however due to the relatively large U-235/U-238 content the sample has a higher induced neutron signature and therefore the measured effective Pu-240 mass is slightly higher (~8% average deviation) than the expected Pu-240 content. However, the measurements agree with each other within ~1% and the leakage multiplication of the sample measured by both detectors also agrees lending further evidence that the detectors are operating correctly.

After the hold was released on sample movements, a glass B-vial filled with plutonium was transferred to the detector laboratory and measured in the FBLNMC. The same B-vial and a Pu bearing SAVY-4000 container were measured in the LNMC. The SAVY was too large to fit inside the FBLNMC, therefore only the B-vial was measured. The mass distribution of the SAVY sample and the B-vial sample are shown in Tables 2-8 and 2-9, respectively.

Table 2-8: Isotopic Distribution of the SAVY Sample Measured.

Isotope	Mass (g)
Pu-238	0.0117
Pu-239	58.299
Pu-240	3.676

Table 2-9: Isotopic Distribution of the B-vial Measured.

Isotope	Mass (g)
Pu-238	0.0003
Pu-239	1.5103
Pu-240	0.1944

The calculation procedure described previously was repeated using the singles, doubles, and triples rates for each measured sample in each detector. Table 2-10 shows the results of the calculated Pu-240 mass for each sample and detector combination measured. Uncertainty in the measured mass is the absolute 1 sigma uncertainty.

Table 2-10: Results of Expected vs Measured Pu-240 Mass for all samples measured.

Detector	Sample	Expected Pu-240 Mass [g]	Measured Pu-240 Mass [g]	Deviation [%]
FBLNMC	Mixed Oxide	0.26	0.2782 ± 0.0192	7.00
LNMC	Mixed Oxide	0.26	0.2801 ± 0.0037	7.73
FBLNMC	B-Vial	0.1944	0.1954 ± 0.0024	0.5
LNMC	B-Vial	0.1944	0.1935 ± 0.008	0.5
LNMC	SAVY	3.676	3.6172 ± 0.0113	1.6

The results for the B-vial measured in both detectors show a very low deviation, indicating that both detectors are calibrated and operating as intended. The result for the SAVY in the LNMC further confirm the detector is calibrated. As mentioned previously, container shape, container composition, sample shape and sample composition all have an effect on the measured Pu-240 mass which can increase the deviation due to these potential unknowns. It is recommended to calibrate the system with a known standard mass in a similar configuration to the unknown or verification samples that will be measured.

2.9 Uncertainty Calculation

As mentioned previously, an assumed 2% error was added to the Cf-252 source activities. The uncertainty was added because the certificate from the vendor did not display an uncertainty for the procured Cf-252 sources. The 2% uncertainty was determined from previous certificates of Cf-252 from the vendor [4]. The uncertainty for singles, doubles, and triples, was propagated via the general error propagation equation, shown below, to obtain the uncertainty in the Pu-240 effective mass. The uncertainty for the singles, doubles, and triples, is provided by the INCC software as raw data. The uncertainties quoted previously for the effective mass are 1σ uncertainties.

given $f(x_1, x_2, \dots, x_n)$

$$E_f = \sqrt{\left(\frac{\delta f}{\delta x_1}\right)^2 * E_{x_1}^2 + \left(\frac{\delta f}{\delta x_2}\right)^2 * E_{x_2}^2 + \dots + \left(\frac{\delta f}{\delta x_n}\right)^2 * E_{x_n}^2}$$

Where E_f is the error associated with function $f(x_1, x_2, \dots, x_n)$ and E_{x_i} is the error associated with the i th element x_i .

2.10 Quality Assurance

Measurements were performed in accordance with the statement of work to evaluate and characterize neutron multiplicity counters, both LNMC and FBLNMC and associated electronics [1].

The Cf-252 sources are ISO/ANSI 77C66535 classification procured from Eckert & Ziegler Isotope Products. The sources were taken from Lot #6050731. The isotopic composition of the Cf-252 sources is given below in Table 2-11. The Cm-248 decay product was last separated on 04/19/2018. After 7 years of ingrowth, the Cm-248 activity is expected to be 6.21E-01 nCi, which is a negligible contribution to the neutron emission.

Table 2-11: Isotopic Composition of Cf Sources Used for the Calibration.

Nuclide	Mass %	Activity %
Cf-249	25.608	0.4612
Cf-250	25.206	12.0745
Cf-251	12.112	0.0845
Cf-252	37.074	87.380

The calibration measurements assumed that the neutron emission was stemming from Cf-252. Cf-250 has a small spontaneous fission branching ratio, therefore some portion of the neutron emission measured will be from Cf-250. Using the isotopes listed in Table 2-11 and the reference values from the Cf-252 certificates, the calculated value for the Cf-250 contamination is 0.35% which is within the assumed 2% deviation from the certificate. Cf-250 has a longer half life than C-252 so this contamination percentage will increase over time, however the isotopic composition was measured on 09/16/2024 and the contamination percentage increase is unlikely to affect the overall measurements.

Requirements for performing technical reviews, and the required extent of review are established in manual E7 2.60. The Environmental and Legacy Management (ELM) directorate documents the extent and type of review using the SRNL Technical Report Design Checklist contained in WSRC-IM-2002-00011, Rev. 2. The review for this data and report was documented by email from the technical reviewer and can be found in the Electronic Laboratory Notebook (ELN) under experiment number M5443-00686-08. The information, data, and calculations associated with the results reported herein are recorded in the SRNL ELN system as notebook/experiment number M5443-00686-08.

3.0 Conclusions

Savannah River National Laboratory's (SRNL) Nuclear Measurements group was tasked with characterizing the performance of two neutron multiplicity counters located at SRNL. Characterization measurements were made to determine the gate width, pre-delay, deadtime parameters, triples and doubles gate fractions, detector efficiency, and operating high voltage for the LNMC and the FBLNMC. The parameters were determined and were, as to be expected, slightly different than the previous calibrations, which were performed over 20 years ago. Several Pu samples were measured to validate the characterizations of the FBLNMC and LNMC. The measurements determined the sample Pu-240 mass within <2% deviation for the pure plutonium samples and ~8% for the mixed oxide sample. The pure Pu samples had significantly better accuracy compared with the impure mixed oxide sample due to the lack of induced fission or alpha,n

neutrons from impurities. Overall, the characterization of the neutron multiplicity counters and the determination of their operability has been completed successfully.

4.0 References

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