Neutron Multiplicity Measurements with ^{3}He Alternative–Straw Neutron Detectors

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Abstract

Counting neutrons emitted by special nuclear material (SNM) and differentiating them from the background neutrons of various origins is the most effective passive means of detecting SNM. Unfortunately, neutron detection, counting, and partitioning in a maritime environment is complex due to the presence of high-multiplicity spallation neutrons (commonly known as “ship effect”) and to the complicated nature of the neutron scattering in that environment. A prototype neutron detector was built using boron-10 (^{10}B) as the converter in a special form factor called “straws” that would address the above problems by looking into the details of multiplicity distributions of neutrons originating from a fissioning source. This article describes the straw neutron multiplicity counter (NMC) and assesses the performance with those of a commercially available fission meter, manufactured by Ortec Corporation. The prototype straw neutron detector provides a large-area, efficient, light-weight, more granular (than fission meter) neutron-responsive detection surface (to facilitate imaging) to enhance the ease of application of fission meters. Presented here are the results of preliminary investigations, modeling, and engineering considerations leading to the construction of this prototype. This design is capable of multiplicity and Feynman variance measurements. This prototype may lead to a near-term solution to the crisis that has arisen from the global scarcity of ^{3}He by offering a viable alternative to the fission meters.

This article describes the work performed during a 2-year Site-Directed Research and Development (SDRD) project that incorporated straw detectors for neutron multiplicity counting.

The two-panel detector system is called the NMC. We used ^{10}B (in the form of enriched boron carbide—$^{10}B_4C$) for neutron detection instead of ^3He (helium-3). In the first year, the project worked with a panel of straw neutron detectors manufactured by Proportional Technologies, Inc., investigated its characteristics, and developed a data acquisition system to collect neutron multiplicity information from spontaneous fission sources using a single panel consisting of 60 straws equally distributed over three rows in high density polyethylene moderator. In the following year, we developed the field-programmable gate array and associated data acquisition software. This SDRD effort successfully produced a prototype NMC with approximately 33% detection efficiency compared to a commercial fission meter.

Keywords: Neutron multiplicity, fission meter, straws, Poisson distribution, field-programmable gate array (FPGA)

I. Background

A straw neutron detector is a narrow copper tube (4 mm in diameter and 5.08×10^{-2} mm thick) coated inside with a very thin (1 μm) layer of boron-10 (^{10}B) that works as a proportional counter when biased with a high voltage. A collection of closely packed straws embedded in a high-density polyethylene moderator form a panel detector that provides high detection efficiency, large solid angle, and enough spatial resolution for it to be useful as a neutron imaging system. A schematic diagram of a panel detector is shown in Fig 1.

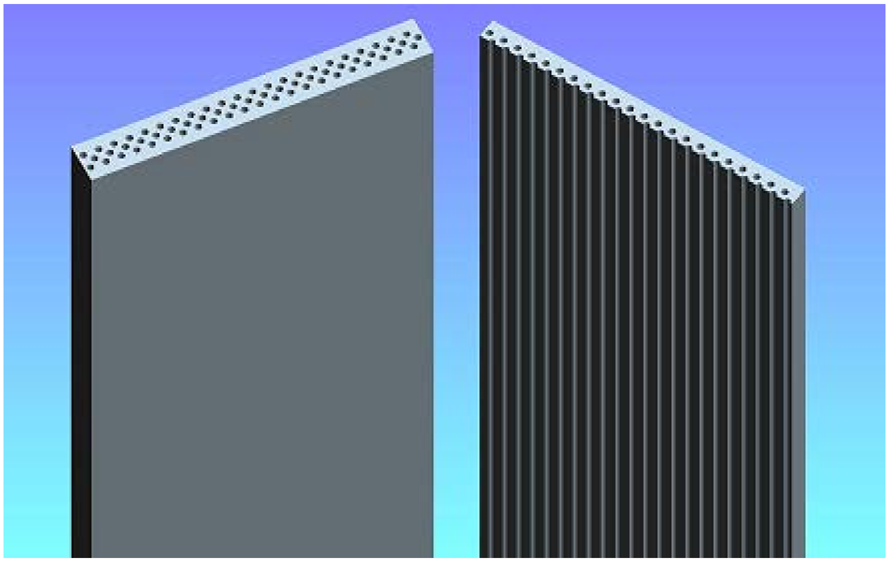

Fig. 1. On the left one set of two paddles with 3 staggered rows, 20 ea. in a row, 50 cm long 4 mm diameter embedded in 5 cm high density polyethylene. On the right is shown a single layer of 20 straws in a row.

Proportional Technologies, Inc. manufactured both the panels used in this research. The scientific value of this effort is that the prototype neutron multiplicity counter (NMC) seeks to provide an effective way to meet two contradictory physics requirements in detecting neutrons via the exothermic neutron absorption on ^{10}B. The mean free path of thermal neutrons in solid

^{10}B is about 18 μm. It takes about a 90 μm layer of ^{10}B to completely stop thermal neutrons. The α-particles generated in the reaction have ranges of about 2 to 3 μm, hence the thin layer of boron coating on the copper tube. The thinness of this layer reduces the intrinsic neutron detection efficiency (the intrinsic detection efficiency of any device operated in pulse mode is defined as the probability that a neutron incident on the detector will produce a recorded pulse). One solution is to make the coating in the shape of narrow tubes (straws) to increase the total ^{10}B surface area.

The project leveraged two core technology bases: (1) the ability to provide uniform thin (approximately a few microns) large surfaces of ^{10}B to interact with neutrons to produce α-particles in a proportional counter configuration, and (2) a high-speed field-programmable gate array (FPGA) to exploit the time correlations between neutrons from different sources. For example, cosmic or background neutrons in a maritime environment are only mildly correlated, primarily because of the high rates of spallation neutrons created by cosmic interactions. Neutrons from (α, n) channels are completely uncorrelated, but neutrons from fission, particularly when multiplications are taking place following spontaneous or induced fission, are very highly correlated. The key neutron signature for SNM results from the fact that the spontaneous fission process emits multiple neutrons in closely spaced temporal groupings. The number of neutrons emitted in spontaneous fission can vary from zero to six or more. The process is random, or statistical, in nature, and the probability distribution of the number neutrons is referred to as the "neutron multiplicity distribution." Total neutron counting counts all the emitted neutrons without further analysis. Neutron coincidence counting looks for pairs of neutrons within a small time window. Multiplicity counting counts separately the number of neutrons detected within a time gate (e.g., none, 1, 2, 3, 4, 5, 6, 7…). The average spontaneous fission multiplicity associated with ^{240}Pu is reported to be about 2.16 and is characteristic of the material. The neutron multiplicity from spontaneous fission of ^{235}U is 1.86. On the other hand a single 1.6 GeV proton would cause as many as 30 neutrons at a time through the spallation mechanism in steel. In an actual ^{240}Pu source, there is also the possibility of induced fission which changes the observed multiplicity. The observed multiplicity depends on the efficiency of detection. A simple way to characterize a count distribution is the variance-to mean ratio[1], R, given by

$$R = \frac{\overline{C^2}}{\bar{C}} - \frac{\bar{C}^2}{\bar{C}}, \tag{1}$$

where is the first moment and is the second moment of the count distribution. For a random (Poisson) distribution the ratio is unity. If correlation is present, the ratio, *R*, is not 1. (The Feynman Variance (Y2F), is defined as (*R* – 1)/2.) To determine the presence of an SNM source, therefore, it is required that we determine if *R* can be distinguished from random (Poisson) background or from correlated, but not SNM, sources such as are produced in cosmic ray showers. By following the detected neutron counting distributions with narrow time gates ranging from 1 to 512 μs, one can distinguish between fission and cosmic neutrons within a short time, on the order of 10 minutes. This approach serves dual purpose—it provides a unique solution to discriminate against cosmic neutrons in a maritime search environment and enables effective measurement of a neutron source on the ground from a large standoff distance (~10 meters). Neutron signals are very important when using passive methods to detect special nuclear materials from large standoff distances; particularly from aerial platform or in maritime environments because neutrons penetrate through common construction materials like steel, lead, tin, concrete and aluminum and have a high probability of reaching the detectors. Fast neutrons from a plutonium source can be detected at a long distance from an aerial platform because the air scattering mean free path is long (>100 meters for 2 MeV neutrons).[2] The fundamental detection limit is always determined by the neutron production by cosmic-ray interactions. Precise neutron counting becomes complicated because of varying, high cosmic neutron background (for example the average cosmic background neutron flux is 120 neutrons/m^2-sec in New York City). The neutron background varies on geomagnetic latitude, atmospheric pressure, solar activity, and altitude of the observation location. In maritime environments neutron counting is made difficult because of the "ship effect," the excess production of neutrons by secondary cosmic-induced interaction (spallation) of high-Z materials (for example steel structures, containers, large engine blocks on board a ship). Spallation by neutron causes as many as 30 neutrons simultaneously and triggers a sporadic high multiplicity event that disturbs the neutron multiplicity background in a way different from neutrons caused by fission process. The ship effect can create a change in measured local backgrounds as much as 2-5 times within a short time interval, which makes instantaneous neutron count rate measurement as a search procedure for finding neutron source useless. The minimization of ship effect for onboard neutron counting and measurement is a difficult challenge in maritime safeguard operations. However, by careful study of the higher-order moments of the multiplicity distribution, one can estimate the source strength and generate intelligent inferences on the surrounding materials around the neutron source.[3]

II. Technical Description

Manufacturing of straws requires ^{10}B (in the form of boron carbide (B_4C) be uniformly and very thinly sputtered (approximately 1 μm thick) on large, thin (5.08×10^{-2} mm thick) copper sheets. With the boron enriched to 96%, the effective percentile weight proportion of ^{10}B in the boron carbide is ~76.9%.[4] Low-energy neutrons incident on a ^{10}B-coated surface will go through the following nuclear reactions

$$^{10}\text{B}(\text{n}, \alpha)\ ^{7}\text{Li} + 2.792\ \text{MeV (ground state)} - 6\%, \qquad (2)$$

and

$$^{10}\text{B}(\text{n}, \alpha)\ ^{*7}\text{Li} + 2.310\ \text{MeV (excited state)} - 94\%. \qquad (3)$$

The respective kinetic energies of the positive ions generated from reaction (Eq. 3) are E_{*Li} = 0.84 MeV and Eα = 1.47 MeV; this restricts the α-particles to have a mean free path of approximately 2 μm in bulk ^{10}B. This limitation on the kinetic energy available to the α-particles requires that the ^{10}B layer be thin so that the α-particles can punch through the ^{10}B surface. The resulting ions from the reaction ^{10}B(n, α)^{7}Li ionize the P-10 gas inside the tubes. The electrons and positive ions generated from this ionization process get drawn towards the anode and cathode (the grounded wall), respectively. The thermal neutron cross section for the ^{10}B(n, α) ^{7}Li reaction is 3840 barns. This cross section drops rapidly with increasing neutron energy and is proportional to 1/v (the reciprocal of the neutron velocity). The corresponding value for ^{3}He is 5330 barns, 40% higher than that for the boron reaction, which makes ^{3}He a better intrinsic thermal neutron detector than ^{10}B. It is the walls of the tubes that contribute Compton and photoelectrons to the gas, which means that it is the geometry and materials of the tubes' construction and the signal induced by neutrons (in case of ^{10}B it generates 477 keV gamma rays) that are critical to the gamma sensitivity. The gamma-ray responses can be separated completely by simple pulse height separation. A lower-level discriminator at 100 keV separates out the gamma-ray responses from the neutrons. Only neutron pulse height is used by the current electronics for n/γ discrimination. Kouzes et al. report a gamma rejection ratio better than ~10^{-9} in an environment of 1 mG/h gamma dose rates.[5]

II. Technical Description

A second detector panel with 100 straws was added to provide a two-panel straw neutron detector with 160 straws.[6] Each NMC straw has an inner radius of 2.2×10^{-1} cm and an average active length of 50 cm. With a layer of 1 μm thick ^{10}B coating, 160 straws contain 1.3×10^{23} atoms of ^{10}B. At thermal neutron energy, the absorption cross section being 3.84×10^{3} barns (1 barn = 10^{-24} cm^2). The available total macroscopic ^{10}B-surface area for neutron interaction within NMC in cm^2 for ^{10}B would be 4.98×10^{2}. A commercial fission meter contains 46 liters of ^{3}He under one atmospheric pressure, and with the thermal neutron absorption cross section of 5.33×10^{3} barns for ^{3}He, the total thermal neutron absorption cross section would be 6.6×10^{3}.[7] These physical constants provide some important useful ratios; for example, the ratios of absorption probabilities (^{10}B:^{3}He) = 7.56%, the number of atoms (^{10}B:^{3}He) = 10.5%, and the thermal neutron absorption cross section (^{10}B:^{3}He) = 72%.

III. System Descriptions

III.A. Data Acquisition System

A data acquisition (DAQ) system was developed using a single-board FPGA (SBRIO-9602 manufactured by Nuclear Instruments) and the LabVIEW software platform. Using the data acquisition system and the NMC, benchmark measurements of the response functions in terms of the neutron counting efficiency for spontaneous fission neutrons from a modest ^{252}Cf neutron source (8.6×10^{3} neutrons per second) were compared to an existing fission meter. Fundamentally the data acquisition system consisted of four components:

1. System Initialization (SI)

2. Microsecond Counter (MC)
3. Real-Time Data Acquisition (RDA)
4. Characterization Data Acquisition (CDA)

Functionally, the SI component has to ensure that the system and memory are reset prior to turning on concurrently the rest of the system modules: the MC, RDA, and CDA.

The MC is a very fast master clock with a 40 MHz pulse rate with hold time (defined as the minimum time following the clock transition from LOW to HIGH that the logic level must be maintained at the input in order to ensure continued recognition) of 3 ns. This provides a self-consistent internal clock (master clock) that tracks neutron counts per microsecond. It also provides the shortest length of time characteristic for the data acquisition. The MC examines the state of a pin for indication of a new pulse. If a new pulse is found, then the previous count data are incremented by one; otherwise, the count data remain the same. After the pulse detection algorithm, the MC examines a counter to determine if 1 μs has elapsed. Once the MC has elapsed 1 μs, the MC examines the operating states of the RDA and CDA. If they are active, the MC will pass the collected count data to their respective first-in first-out (FIFO) buffers, before resetting the count data to zero. Neutron arrival-time statistics are correlated in a histogram of the numbers of neutrons arriving within a time gate for a range of gate widths (1 μs through 512 μs in steps of 1 μs).

The RDA keeps track of multiplet statistics (singles, doubles, triples, etc.), calculates instantaneous value of the excess of variance above the true random Poisson distribution (called the Feynman variance [Y2F]), and determines trends of Y2F to perform partitioning of neutrons according to their origin: cosmic or man-made. The RDA operates within a loop, which has two phases, Get Parameters and Acquire Data. In the first phase, the RDA continually gathers parameters that are input by users (items like bin width, time gate width, maximum number of time bins etc. After the Acquire switch is pressed, the RDA passes the last gathered parameters and begins the second phase to acquire data. The RDA gathers microsecond count data from its FIFO, from which it builds the real-time log file. The RDA sums data from the FIFO (bin width/1 μs) times. It then increments the summed position in the Multiplet Array and resets the summed value to zero. After repeating the previous process, Bin Count Times, the Multiplet Array is passed to the host computer via a second FIFO, and then the Multiplet Array is reset to zero. The host is responsible for generating the log file. The RDA's second phase can be stopped and reconfigured at any point during operation. However, the second phase will only halt after the completion of a host FIFO transfer. The RDA will then reenter the first phase.

The CDA builds statistics of the higher moments Y2F, Y3F, Y4F and generates asymptotic values of R2F, R3F, and R4F, respectively, at the end of all cycles. Fitting of R1F, R2F, R3F, and R4F yields the neutron fission source term S, the neutron die-away time (λ^{-1}), multiplication (M), efficiency (ε), and the Rossi alpha (α) ratio, which can be defined as the functions of the (α, n) channel strength to the spontaneous fission channel strength. Similar to the RDA, the CDA operates in two phases of Get Parameters and Acquire Data. The CDA continually gathers parameters until the Acquire switch is pressed. After the switch is pressed, the CDA passes the last gathered parameters and gathers the microsecond count data from its FIFO, from which it constructs the 2A log file. The CDA sums data from the FIFO (bin width/1 μs) times. It then increments the Multiplet Matrix (MM) based on the summed count and the active gate. After incrementing the MM, the CDA returns to the previous step and collects data for the next gate. The summed count data is reset to zero after the data for the last gate/bin were incremented in the MM (also known as the completion of a cycle). A cycle's duration can be calculated as Bin Width × Bin Count. The above steps are repeated the number of times specified by the cycle parameter. The MM is transferred to the host when all cycles are completed. When data acquisition is complete, the CDA reenters the Get Parameters phase. The host is responsible for generating the log file.

The CDA has the ability to append data to previously acquired data by not clearing the memory for the MM. The previous data file will not be overwritten. The CDA Acquire Data routine can be stopped at any time; however, the routine will only halt after a full cycle is completed, at which point it will transfer the MM to the host computer and reenter the first phase. Flow charts for real-time data acquisition (Fig. 2) and characterization data acquisition (Fig. 3) are included for clarification of the flow of the electronic logic.

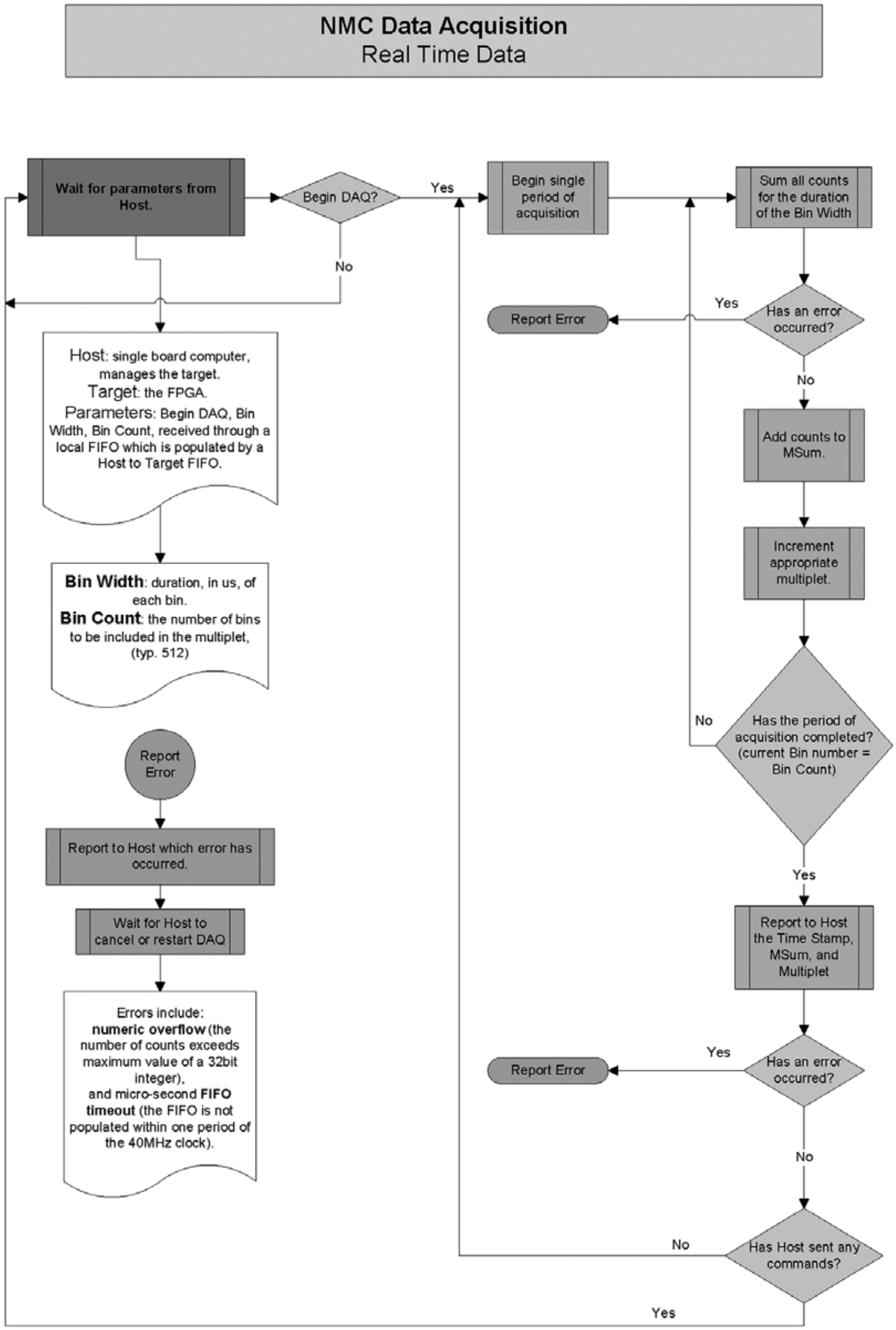

Fig. 2. Electronic flow chart for NMC real-time data acquisition (RDA) system.

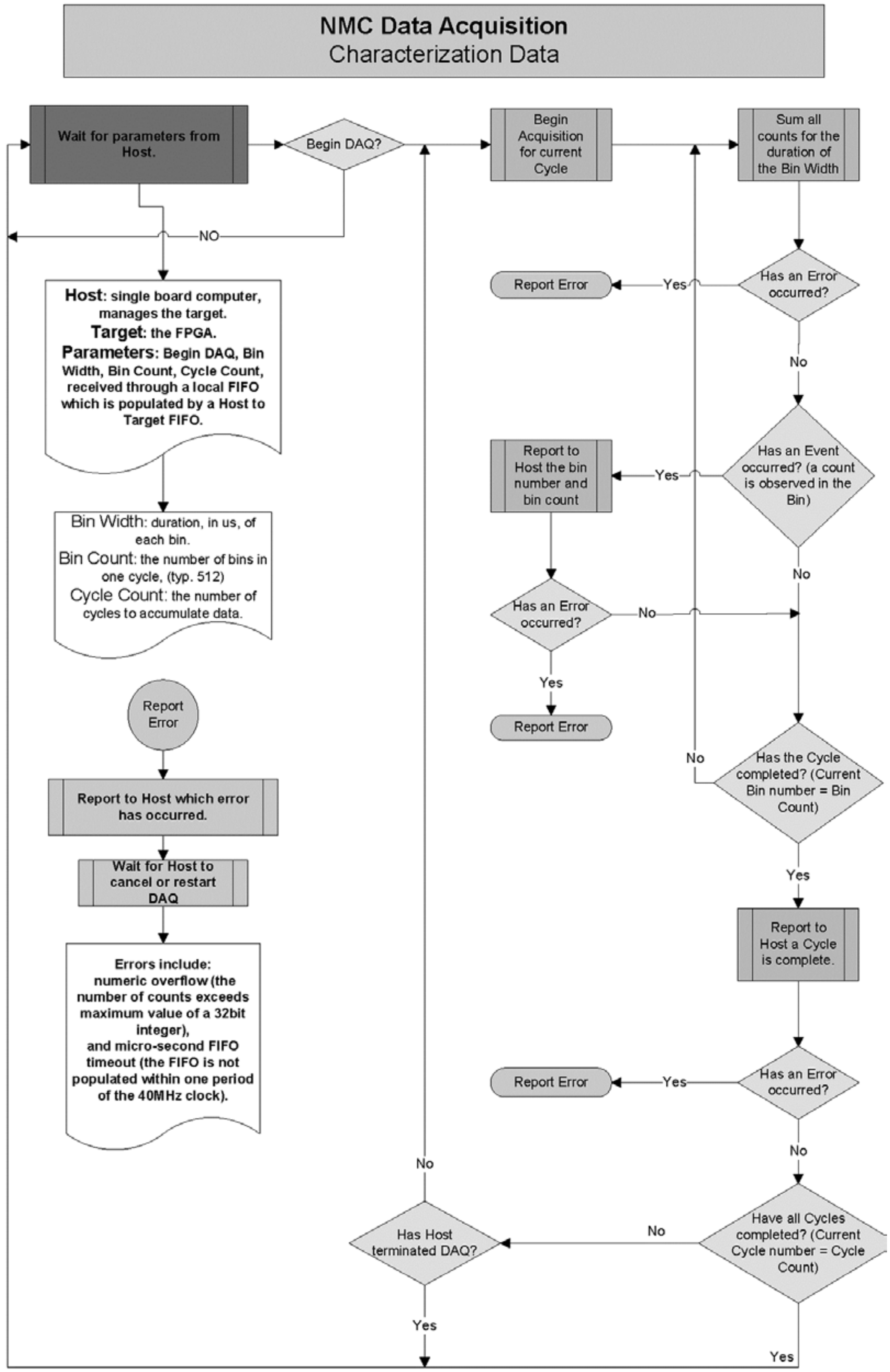

Fig. 3. Electronic flow chart for NMC characterization data acquisition (CDA) system.

III.B. Physical and System Descriptions

The physical characteristics of the two-panel straw NMCs are listed in Table I. The two panels have a different number of straws, but they have the same effective length (48.3 cm), width (40 cm), and thickness (6.25 cm).

Table I

Dimensions and Physical Characteristics of NMC Panels

Physical dimensions	Panel 1 (60 straws)	Panel 2 (100 straws)
Panel length	70 cm	70 cm
Straw length	50 cm	50 cm
Effective straw length	48.3 cm	48.3 cm
Straw inner diameter	4.43 mm	4.43 mm
Panel weight	14.5 kg	14.9 kg
Separation between tubes	6.5 mm	4.0 mm
Solid angle at 1 meter	0.015 str	0.015 str
Power input	+12 VDC at 0.83 A	+12 VDC at 0.25 A (lower power, newer battery supply; battery life is1^{6} hours)

With a 12 VDC power input, each of the NMC panels individually generates one each of an analog pulse and one transistor-to-transistor (TTL) pulse. The TTL output is about 500 ns wide and 3.3 VDC in amplitude. The two TTL outputs could be summed up electronically so that the NMC works as a single unit for neutron counting purposes. The TTL pulses were fed to the FPGA input, which is a single board reconfigurable input/output system printed circuit board. The data acquisition software to control the FPGA sorts by time value counts neutrons in list mode. The NMC is shown in Fig. 4.

Fig. 4. Two-panel NMC. The left panel has 100 straws and the right one has 60 straws. Each panel's external dimension is 70 × 40 × 6.25 cm and the combined weight is 29.4 kg (64.8 lb).

IV. Experimental Results with Straw NMC

IV.A. Sensitivity Analysis

A 8.6×10^3 neutrons/sec ^{252}Cf source was used to compare the neutron counting efficiency of the two-panel straw NMC against a fission meter. Fifteen-minute average counts from the source were measured with the two systems, for the source placements at different distances from the detector surface centers. The moderated side of the fission meter was facing the source to maximize the count rates. Measurements with distance up to 2 meters between the source and the system were also performed. The neutron count rates per second as a function of the distance between the bare source and the detector are plotted in Fig. 5. As seen from the figure, the NMC is ~33% efficient with ^{252}Cf as compared to the fission meter for counting neutrons. Similar measurements were carried out with the same ^{252}Cf source but with ~2.5 cm of polyethylene moderator surrounding the neutron source. The detected neutron count rates are shown in Fig. 6.

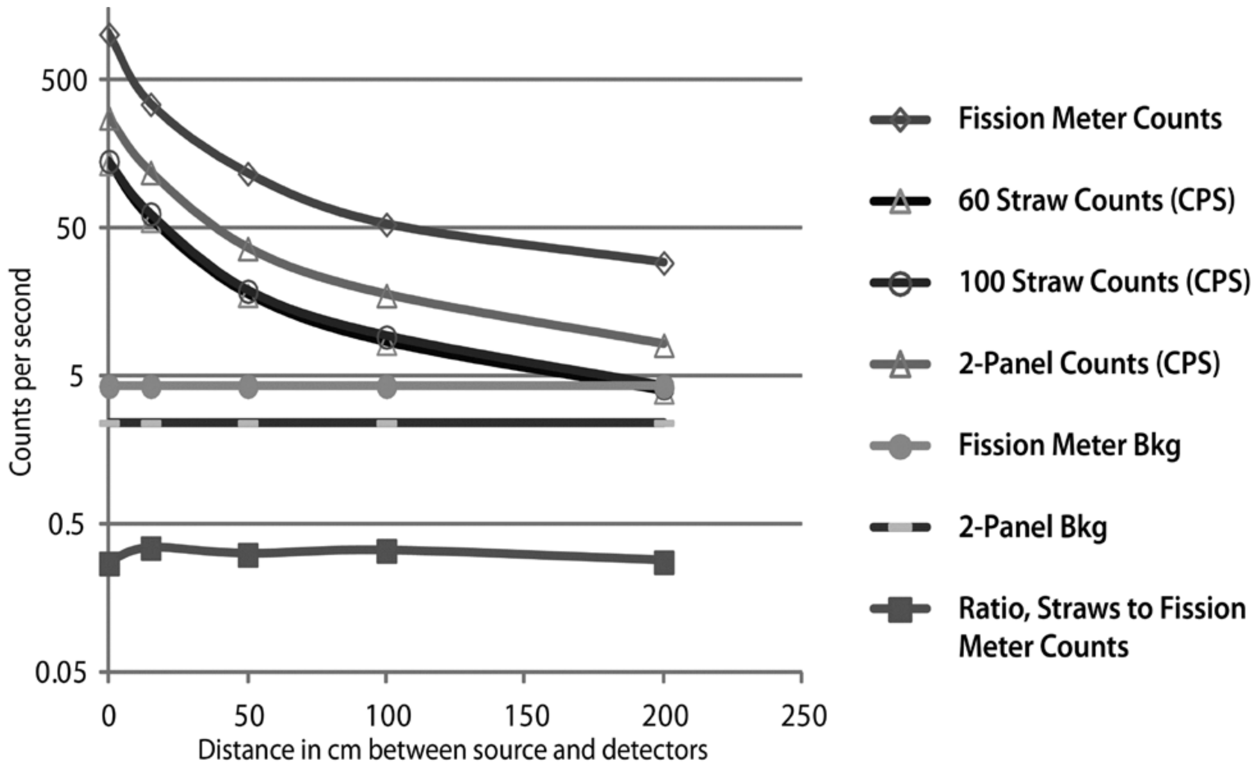

Fig. 5. Separate count rates from two panels were tracked from a bare 8.6×10^3 neutrons/sec ^{252}Cf source. The 2-panel count rate is the algebraic sum of the two 1-panel count rates. The ratio of the NMC neutron counts per second to the fission meter neutron counts is shown at the bottom part of the plot; the ratio is flat across the distance between the source and the detectors. Bkg stands for the average background neutron counts for each of the detectors.

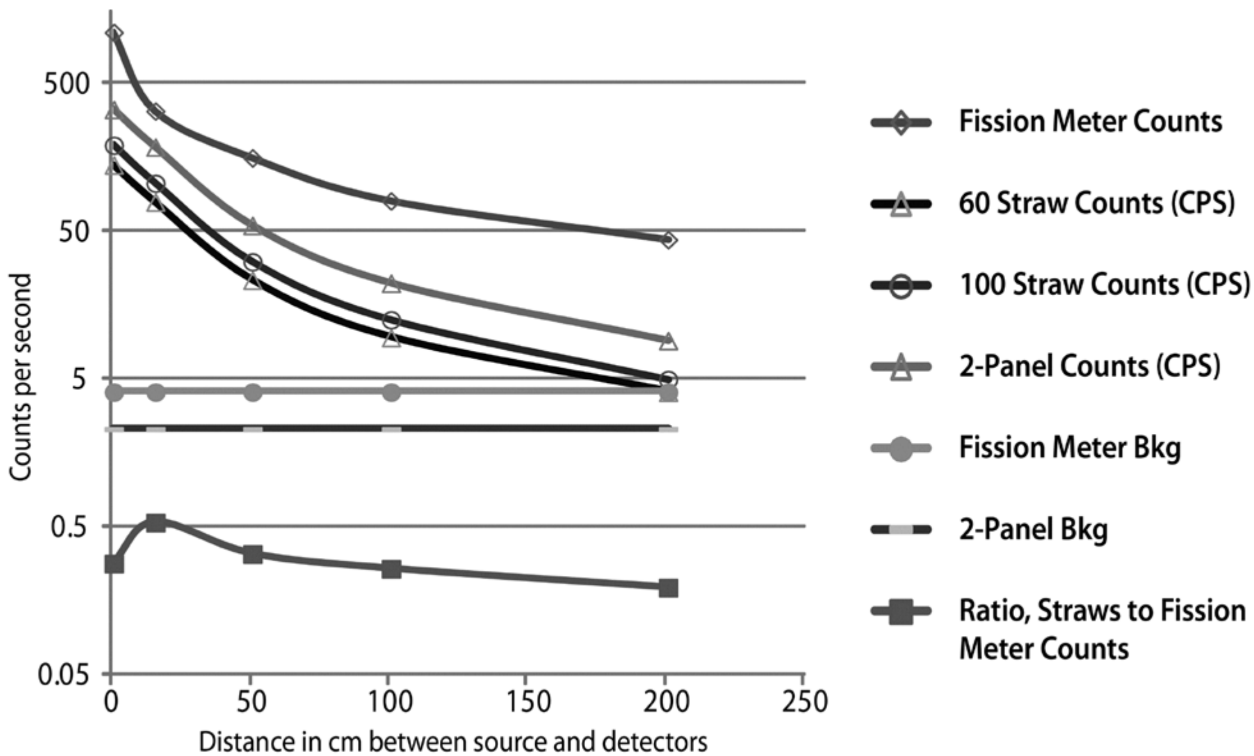

Fig. 6. Separate count rates from two panels were tracked from a moderated 8.6×10^3 neutrons/sec ^{252}Cf source. The 2-panel count rate is the algebraic sum of the two 1-panel count rates. The ratio of the NMC neutron counts per second to the fission meter neutron counts is shown at the bottom part of the plot.

IV.B. Neutron Partitioning

The RDA creates a data log file that tallies the number of zeros, singles, doubles, triples, and up to septuplets (seven sets of neutrons that arrive at the same time within a preset time bin). The Feynman variance Y2F can be determined from these data tables to obtain the multiplicity distribution. Depending on the initial value of the Y2F in the background, one can calculate the relative percentile contributions of the (cosmic) background and the correlated neutrons. Table II shows the results of neutron partitioning obtained with the total average neutron counts in the counter. The averages vary from 1.7 to 3.0 times the average background counts. In this regime of low neutron counts, the partitioning is not exact, but the introduction of a time-correlated neutron source always shifts the cosmic contribution substantially (100% to start with for the background).

Table II

Real-Time Data (Partitioning of Neutrons by Origin)

Total Source + Bkg Strength (Length of Run)	Mean Count Rate Total Cps ±Error	Y2F ±Error	Calculated Cosmic Percentile	Calculated Non-Cosmic Percentile	Actual Cosmic/ Non-Cosmic Percentile
1.7 × Bkg (60 min)	6.9 ±0.6	0.00153 ±0.00034	74 ±7	26 ±3	58.8/41.2
1.8 × Bkg (71.4 min)	7.5 ±0.8	0.0015 ±0.0003	74 ±8	26 ±3	55.6/44.4
2.0 × Bkg (63.8 min)	8.0 ±0.9	0.00145 ±0.0003	70 ±8	30 ±4	50/50
2.2 × Bkg (24 min)	8.8 ±0.4	0.000888 ±0.0005	46 ±14	54 ±15	45.5/54.5
3.0 × Bkg (30 min)	11.4 ±0.8	0.00095 ±0.0004	46 ±5	54 ±5	33.3/66.7

IV.C. Characterization Data

Characterization data contain the counts of a progressive 40 × 512 elements matrix (up to 40 multiplets are kept for each time gate width, ranging from 1 μs to 512 μs) from which the statistics for Y2F, Y3F, and Y4F are built. Their corresponding asymptotic values, namely R2F, R3F, and R4F, respectively, are determined at the end of the 512 μs gate. The algebraic operations involved in calculating the Feynman variances are lengthy and cumbersome.[7] The fission parameters calculated from the characterization data (obtained from the two-panel NMC using a 2 μCi ^{252}Cf source in close proximity) are tabulated in Table III.

Table III

Measured Spontaneous Fission Parameters for the Prompt Neutrons

Fission Properties	Bare Source	Moderated Source	Comments
Spontaneous fission source strength	4.06×10^{-9} g	3.71×10^{-9} g	Actual mass of 2 μCi ^{252}Cf (with specific activity of 540 Ci/gm) is 3.7×10^{-9} g.
Neutron die-away time, λ^{-1}	52.62 μs	97.28 μs	The bare source die away value is true neutron die away value; the excess of about 45 μs die-away time for moderated source accounts for neutrons traveling through the polyethylene moderator.
Total multiplication	1	1	There is no other medium to undergo induced fission; therefore, there is no multiplication for ^{252}Cf.
Absolute efficiency (neutrons detected/neutrons emitted)	1.49%	2.2%	Moderation lowers the neutron energy and makes the sensor more efficient because the source configuration is under moderated.
α-parameters (ratio of (α, n) channel to spontaneous fission channel	0.01	0.01	The prompt neutrons emitted from the ^{252}Cf source are mostly due to spontaneous fission.

Figs. 7 and 8 show the multiplicity plots for bare and moderated ^{252}Cf sources, respectively. The fact that the tails of the multiplicity plot deviate from the random Poisson distribution shows that the neutron source is a fission material. From the moderated source one obtains an approximate thickness of the hydrogenous moderator.

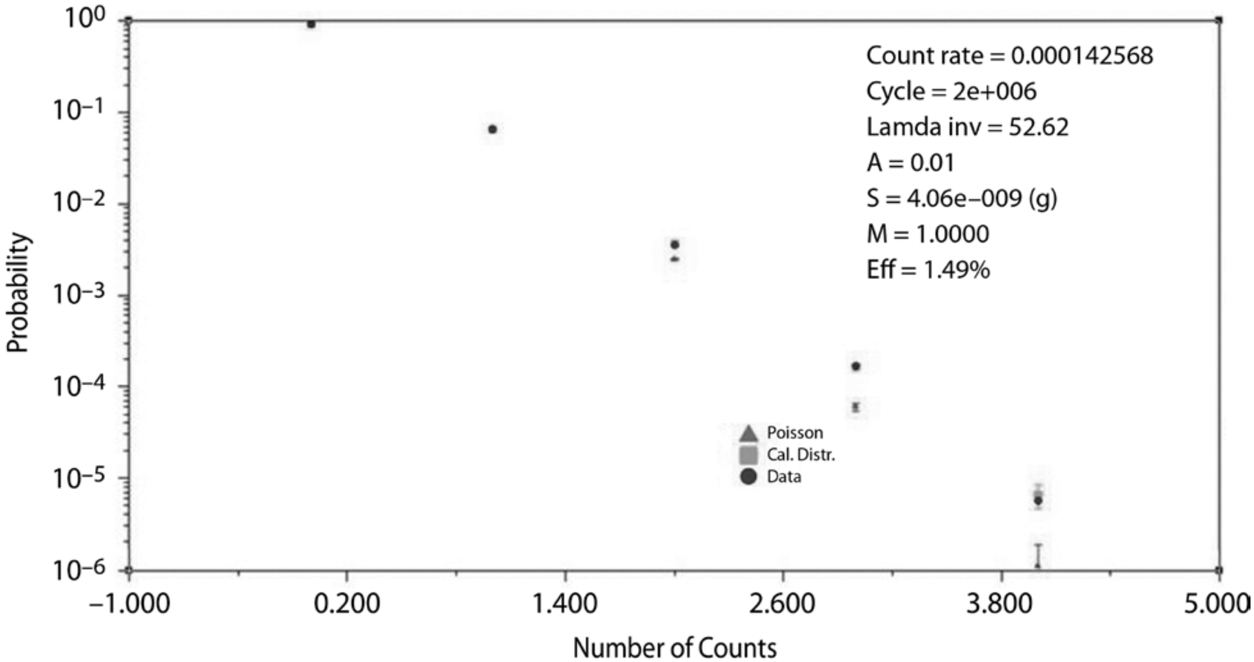

Fig. 7. The measured probability distribution of neutron count rates is shown from a bare ^{252}Cf source. The theoretical fit estimates an absolute neutron detection efficiency of about 1.5% and source strength of approximately 4×10^{-9} g. In Fig. 7 the estimated source strength is about the same.

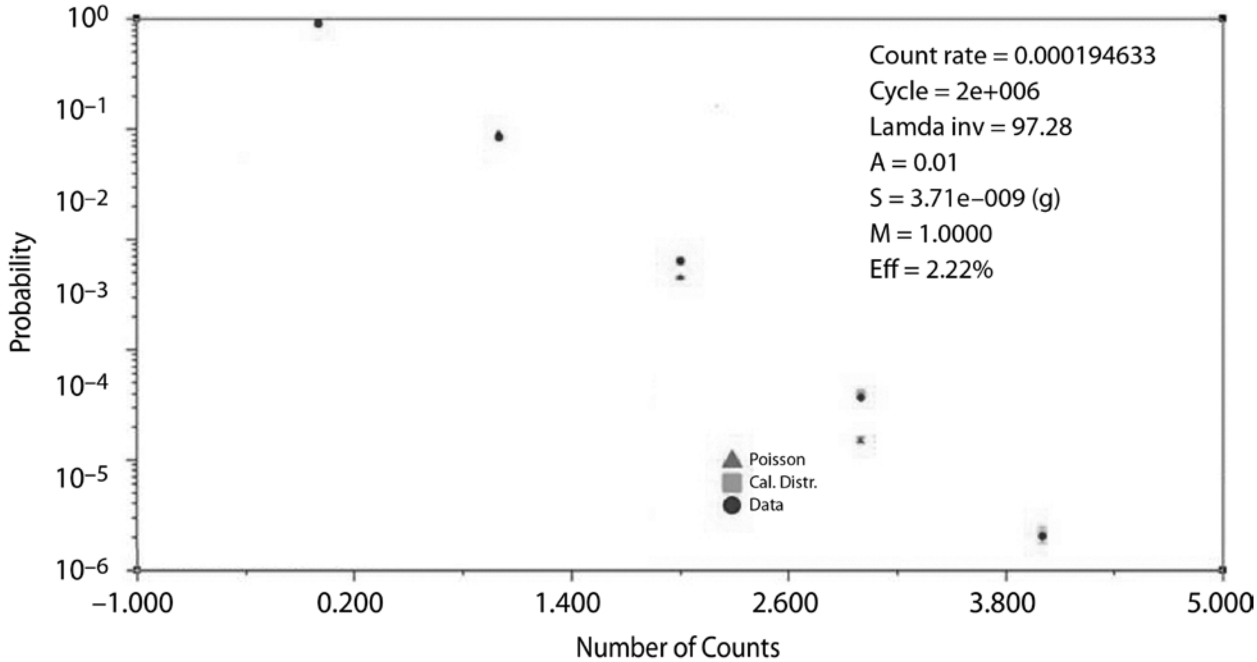

Fig. 8. The measured probability distribution of neutron count rates is shown from a moderated ^{252}Cf source. The theoretical fit estimates an absolute neutron detection efficiency of about 2.2% and source strength of approximately 3.7×10^{-9} g. In Fig. 7 the estimated bare source strength is about the same. The efficiency for the moderated source is as expected, somewhat higher than that of a bare source. The estimated thickness of the moderator is slightly above 2.5 cm.

V. Conclusion

This project produced and demonstrated a prototype neutron multiplicity detector using ^{10}B-lined straw neutron detectors with approximately 33% detection efficiency when compared to a commercial fission meter. Successful experiments were carried out to characterize a 2 μCi ^{252}Cf source in close proximity for 15 minutes. The NMC successfully discriminated between cosmic neutrons and time-correlated neutrons within 5 minutes at a distance of 2 meters. The NMC determined the average neutron die away time (λ^{-1}) to the order of ~50 μs. Using the same source (once bare and once moderated), the NMC was able to determine the thickness of the polyethylene in terms of additional neutron delay time (~18 μs/cm or 45 μs/inch). Unless bundled together to produce more ^{10}B surface, or without having a complex deposition pattern, the NMC will not have the neutron detection efficiency of a commercial fission meter. Neutron detection efficiency of 160 straws is about one-third of that of a fission meter up to a distance of 2 meters from both a bare and a moderated ^{252}Cf source. The NMC data acquisition system has the unique advantage over the fission meter because it can simultaneously parse data in real time and perform characterization analysis. In addition, the current FPGA can sort neutron data like a shift register.

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