MOBILE HIGH EFFICIENCY MULTIPLICITY COUNTER FOR URANIUM MEASUREMENTS

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ABSTRACT
To meet MPC&A contract terms and programmatic transparency objectives within the Cooperative Threat Reduction Treaty between the US and Russia, it is important to measure the material intended for down blending for its total uranium concentration and for its ^{235}U enrichment. A two-measurement technique provides the best combination of overall measurement accuracy, flexibility, ease of use, ready commercial availability, and economy of operation.

Gamma spectrometric systems that can measure the ^{235}U enrichment are readily available commercially. Active neutron counters for total uranium determinations are also readily available, but generally not in the size required for this program objective. Although there is no commercial off-the-shelf version of such a counter, neutron multiplicity counters are functionally very modular and it is common practice for the commercial companies that build them to produce them in an "as needed" form-factor (within sensitivity parameters). The counter that was found to meet the necessary design criteria is based on a high efficiency passive neutron counter which has been modified to include an excitation source, similar to the one in the Active Well Coincidence Counter (AWCC).

In this paper, we will report on the design and test results of a large scale Hexagonal High Efficiency Multiplicity Counter that was recently built for a mobile nuclear material characterization ISO container for deployment in Russia.

INTRODUCTION
Since 1994, the Department of Energy has undertaken the mission of upgrading the safeguards and security of Russian nuclear facilities under the Material Protection, Control and Accounting (MPC&A) program. In early 1999, the MPC&A program launched the Material Conversion and Consolidation (MCC) Project initiative with Russia. The mission of MCC Project was to assist in consolidating special nuclear material (SNM) to fewer locations, and to down blend the material with natural or depleted uranium to reduce its attractiveness as a diversion target. However, the process of moving the material from its point of origin to the down blending facility is currently vulnerable to undetected diversion. To eliminate this vulnerability, it is necessary that the material be assayed immediately prior to shipment from the point of origin, and then again immediately upon arrival at the down blending facility. Another reason for such a set of measurements is to assure the shipper facility that its material and its packaging meets the acceptance criteria of the receiver facility, which have been set in the MPC&A program contract terms and programmatic transparency objectives.

Such measurements, however, present significant procedural challenges arising from calibration, operation, and sensitivity variations between the assay instruments at each end of the transport cycle. One method to overcome these difficulties is to deploy a self-contained instrument van that is transported with the material shipment and is used to make the assays at both ends of the transport cycle. Because the same instruments are used at both ends, the calibration and sensitivity variances are canceled; because the operators of the instruments are different, the MPC&A program can have a high degree of confidence in the validity of the measurement results.

In order to determine the uranium weight percentage and its enrichment as required it is best to use two measuring techniques. A single measurement technique usually cannot do it alone with sufficient or desired accuracy. A gamma spectroscopic measurement is required to establish the uranium enrichment or the plutonium isotopics. However, a gamma spectrometric technique can not typically establish the weight percentage. Uranium is such a heavy material that the self-attenuation of the characteristic gamma rays prevents a gamma technique from seeing the entire sample. A neutron technique will obtain a measurement result that is directly proportional to an effective mass, which however, is dependent on the uranium enrichment. A neutron technique alone is sufficient if the uranium enrichment is known.

The most common approach, used extensively in various safeguards and waste measurements [1,2,3,4], and also the most sensible approach for this project, is not to assume that the uranium enrichment is known, but to use the two techniques in combination to measure the uranium mass in the sample. Combining the mass information with a weight obtained from a simple scale allows the instrument to report weight percentage.

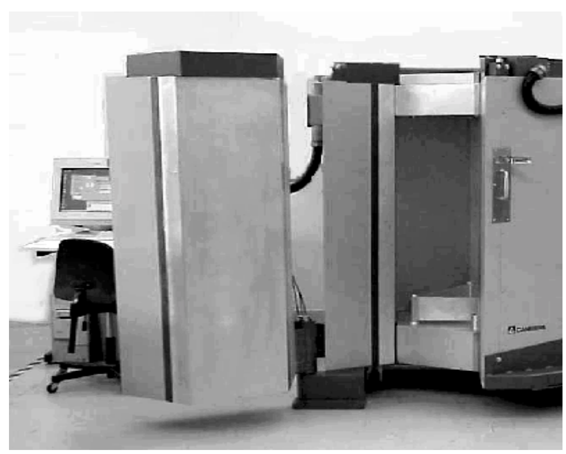
In order to meet the delivery schedule, it was also imperative that both the gamma and neutron equipment was chosen in such a way that it was either available from inventory or the components were available from inventory. For the gamma part of the system, we decided to use a simplified Segmented Gamma System (SGS) that only uses the MGAU[5] software for determining the uranium enrichment. The gamma system was also equipped with a scale to establish the weight of the item being measured. If necessary, the system can be converted to a traditional SGS for measuring canisters and drums[6,7] with the addition of a lift mechanism and, if desired, a transmission source and the appropriate software.

There are a number of neutron counting techniques that can be employed for verification of uranium material. Most neutron counters used for these types of applications use ^{3}He proportional counters as the detector element, although other detector technologies also exist. Both passive and active neutron measurement techniques are used. In the passive mode, neutrons from spontaneously fissioning isotopes, such as ^{240}Pu and ^{244}Cm, are measured. In the active mode, an external source of neutrons (from a neutron generator or isotopic source) first irradiates the sample. Then the neutron signal from fissionable isotopes, such as ^{239}Pu or ^{235}U, is measured. Both modes are in common usage in a variety of applications.

In this case, active neutron analysis is required for ^{235}U determination. A system favored by the IAEA for such measurements is the Active Well Coincidence Counter (AWCC)[8]. The AWCC uses small Americium-Lithium (Am(Li)) sources to interrogate the uranium material. Typically, AWCCs can also be operated in passive mode by removing the interrogating neutron source. However, standard AWCCs accommodate only smaller cans (typically less than 25 cm in diameter) and were not suited for this application as such. There are published designs of large multiplicity counters[9,10] that have been modified to include an excitation source, similar to the AWCC. Although there was no commercial off-the-shelf version of such a counter, neutron multiplicity counters are functionally very modular and it is common practice for the suppliers of neutron counters to produce them in an "as needed" form-factor (within sensitivity parameters). Therefore, we went ahead and designed a large multiplicity counter for this project using standard components and design techniques. This counter was based upon an existing 3-ring 55% drum multiplicity counter recently installed in the United States. The characteristics and performance of the resulting counter are described below.

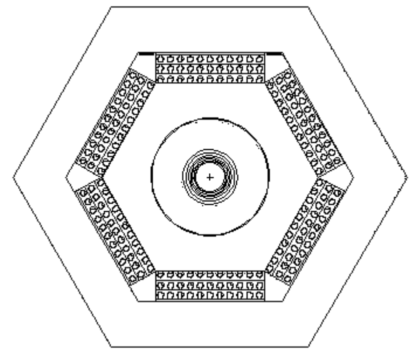
SYSTEM DESIGN

In order to accommodate the greatest possible range of expected sample sizes, the multiplicity counter was designed for 220 liter (55 gallon) drums. For reasons of neutron collection efficiency, structural integrity, and ease of manufacturing, the measurement cavity was designed to be of hexagonal shape. The resulting Hexagonal Neutron Multiplicity Counter (HNMC), shown in Figure 1, is a passive/active neutron coincidence counter intended for use with multiplicity counting. The assay cavity (Figure 2) has a nominal minor diameter of 78 cm (28") and measures 119 cm (47") tall. The counter utilizes 132 ^{3}He proportional tubes arranged in two concentric rings about the assay cavity. The basic configuration and detection properties of the six sides of the counter are identical. The counter utilizes graphite reflectors, located at the top and bottom of the cavity, to improve linearity in response and increased efficiency. Note that this counter was based on a three-ring design. MCNP studies indicated that the inner most tubes could be simply



- *Figure 1: Photograph of HNMC in testing* removed providing acceptable performance but at lower cost. Even though no ^{3}He tubes were installed for the third ring, the holes for the tubes were drilled into the polyethylene moderator blocks to facilitate future enhancement of the system.

The six detector modules each contain 22 ^{3}He tubes, arranged in two parallel rows of 11 tubes each. The 22 tubes are divided into six banks (3 JAB-01s per row); each connected to a JAB-01 pre-amp/amp/discriminator board. Each junction box contains a 20 MHz de-randomizer board. The de-randomizer takes the TTL signals from the six JAB-01 boards and provides two outputs from the detector module, one corresponding to each row of tubes. The signals from the six detector modules are input to a seventh de-randomizer board located in the panel mounted on the counter. This de-randomizer provides three outputs, one from each of the two rings plus a summed output from all 36 JAB-01 boards. This arrangement of JAB-01 boards and de-randomizer circuits results in a counter dead-time of about 25 nanoseconds.



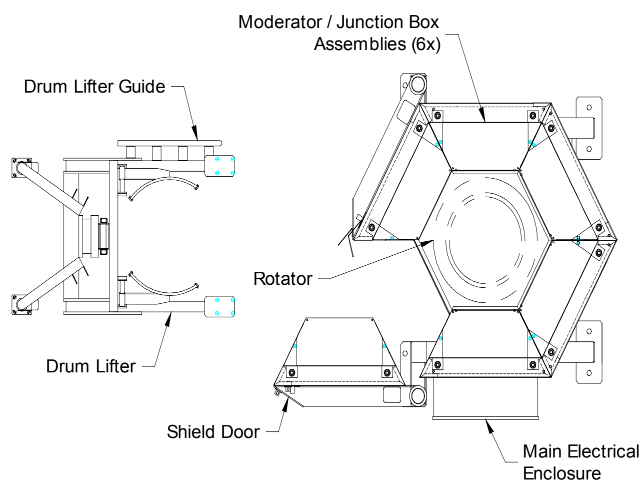
- *Figure 2: General Detector Bank Locations*

The counter is hexagonal shaped with two rows of 1” diameter ^{3}He tubes embedded in HDPE. The counter uses graphite top and bottom end-plugs to flatten the axial response and provide increased counting efficiency. The assay cavity is cadmium lined with a thin aluminum cover. The drum is placed in the assay cavity on a low Z metal (aluminum). An outer shield of HDPE is provided to minimize the effects of background and any nearby sample drums.

To assure an easy lateral loading of the specimen drum, each door assembly pivots about a vertical hinge pin. Both door counter assemblies may be latched shut for security purposes with the hasp provided.

Plain polymer journal bearings placed 12” apart provide the low friction resistance to motion required for manual door operation. A handle force between 7 and 13 pounds is required to open the 850-pound door.

The HNMC is intended to operate in both passive and active modes. The passive mode will be utilized to confirm the U-238 content of the sample, while the active mode will provide the U-235 content of the sample. The counter has been supplied with three Am(Li) source holders for active mode operation. A drum lifter has been provided to load the samples into the counter (Figure 3).



- *Figure 3: Top view of the counter with the drum lifter shown.*

SYSTEM PERFORMANCE

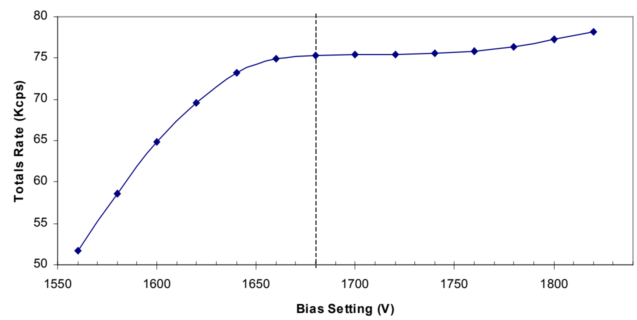
A neutron multiplicity counter is characterized by several basic operating parameters. These parameters are determined by the neutron response characteristics of the assay cavity and signal processing electronics. These parameters are differentiated from more typical calibration parameters, as they are determined without the need for actual plutonium or uranium samples. The required parameters are listed in Table 1, along with a brief explanation of each. The procedure to determine some of these important parameters is provided the sections that follow.

High Voltage	Optimal Setting of the bias supply
Pre-delay time	Short Coincidence Time Cut-off
Die-Away time	Typical time required for fast neutron to slow down and be detected.
Gate Width	Coincidence window width.
Dead time Parameters **a** and **b**	Dead-time Correction parameters for coincidence counting
Dead time parameter **τ**	Primary dead-time correction parameter for multiplicity counting.
Dead time parameters **c** and **d**	Dead-time Correction parameters for multiplicity counting, applied only to doubles and triples rates
Efficiency	Neutron Detection Efficiency for neutrons emitted in the center of the assay cavity.
Doubles Gate Fraction	Fraction of two neutron coincidence events which occur within the coincidence gate.
Triples Gate Fraction	Fraction of three neutron coincidence events which occur within the coincidence gate.
ρ_0	Reference ratio of reals to totals neutron events for use with the multiplication correction in coincidence counting.

- *Table 1: Calibration parameters for multiplicity counters*

High Voltage

The ^{3}He tubes are proportional counters and the optimal high voltage setting is determined in the traditional fashion for these types of detectors. With a small Cf-252 source placed within the counter, the voltage is slowly incremented and, at each step, the totals count rate is recorded. When the plateau region has been identified, the count rate versus voltage is plotted, and the correct setting is obtained by choosing a value approximately 40 volts above the knee (Figure 4).



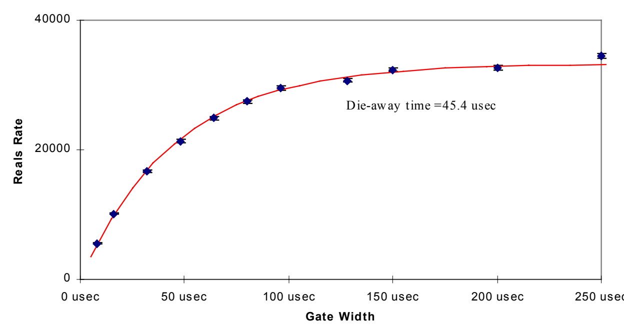
- *Figure 4: High voltage plateau for the HNMC. Optimal HV setting is 1740 volts.*

Pre-Delay

A discussion of the effects of the derandomizing buffer can be found in Reference [11]. A pre-delay setting of 4.5 µsec is suggested. The calibrations for this counter were determined using the 4.5 µsec setting.

Die-Away Time

The data was obtained by placing a small Cf-252 source in the detector and measuring the coincidence rate as the gate width is incremented. The data are fit to a function of the reals rate $R(t_d)$ as a function of the gate width to provide the value for τ. In this case, fitting the data by minimization of the chi-square yields R_0 = 33277 and τ = 45.37 µsec. See Figure 5.



- *Figure 5: Reals rate vs. gate width setting used to determine the die-away time of the counter.*

Gate Width

The detector gate width is determined from the same data as the die-away time. Since the real coincidence events fall off as a function of time, while the random events do not, there is an optimal window width that minimizes the uncertainty in the reals rate. The gate width is chosen such that the error in the reals rate is at a minimum or at a point where significant gains are no longer achieved. In this case the minimum occurs for a gate width setting of 64 µsec. This agrees well with the expected value of $1.257*\tau_{eff}$= 68 µsec. The recommended gate with setting is 64 µsec.

Dead Time Parameters

The dead time parameters for the standard NCC counting are determined by measurement of multiple Cf-252 sources. Ideally the sources should span a range in size such that the smallest introduces only negligible dead-time into the system and the largest causes a totals rate in excess of the largest expected totals rate, typically greater than several hundred thousand counts per second (kcps). For the HNMC, six Cf-252 sources were utilized, providing count rates from 3 kcps to 650 kcps. Each source was placed on the end of a thin aluminum source rod and assayed for at least 3600 seconds.

Historically, for counters containing greater than 6 detector banks utilizing the Amptek pre-amp/amp/disc boards, the ratio of parameter **a:b** is fixed at 3.1×10^6. Examination of the data indicates that a better fit is achieved if the parameter **b** is allowed to vary independently from **a**. Setting the parameter b=0 provides a reasonable fit and is chosen due the relative simplicity of the correction.

Because the count times were long, and the counting statistics were essentially negligible, the initial fits resulted in a large chi-square value. Assuming a 0.64 cm (0.25") positioning error introduces a 0.025% error into the R/T ratio. An additional error of 0.09% is introduced from door closing reproducibility. The errors in the R/T ratio have been increased 0.1% to account for these errors. The summary of the coincidence dead-time parameter determinations is provided in Table 2. (Multiplicity dead-time parameters are presented in Table 8)

Technique	**a (sec)**	**b (sec^2)**	**Standard Deviation of R/T Ratios**
b=0	102.3 E-9	----	0.12% (reduced χ^2 = 0.8)
b=a / 3.1E6	84.3 E-9	2.72 E-14	0.14% (reduced χ^2 = 1.0)
b independent of a	113.2 E-9	-41.65 E-14	0.11% (reduced χ^2 = 0.7)

- *Table 2: Summary of dead-time parameter determinations.*

For this counter setting b=0 was selected for simplicity.

Efficiency

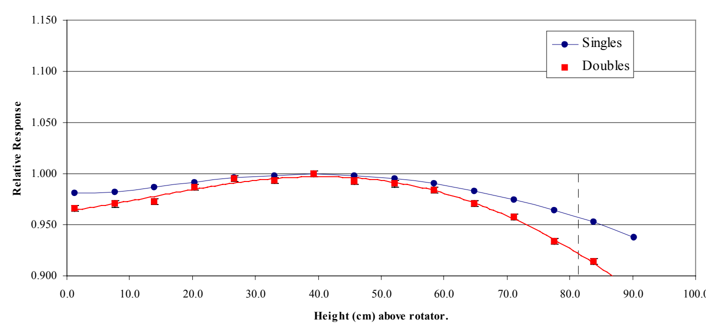
A Cf-252 source was placed in the assay cavity center and assayed for 3600 seconds. The measured singles rate is corrected for dead-time losses and for background. The counter efficiency is then simply determined by dividing the measured rate by the expected. The results for three configurations are given in Table 3.

	Empty Chamber
Date of Measurement	01-Nov-99
Source Strength as of date (nps)	222,432 +/- 3225
Corrected Totals Rate (cps)	75511.4 +/- 4.7
Efficiency	0.339 +/- 0.005
Pu-240 Efficiency Estimate	0.346 +/- 0.005

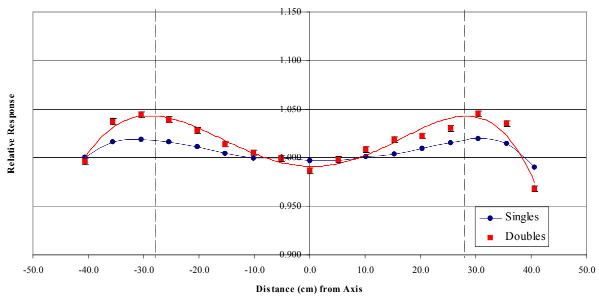
- *Table 3: Efficiency measurements for the HNMC.*

Axial Response Profiles

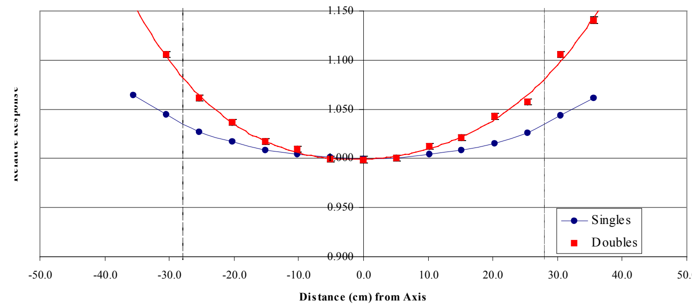
The response profiles for the three primary axes are shown in Figure 6, Figure 7, and Figure 8. The plots are given relative to rates measured at the center of the sample volume. The rotation of the drum reduces the axial variation exhibited in the Y-axis plot such that the variation in the coincidence rate is approximately +/- 5% over the drum volume (empty chamber). Introduction of moderating materials into the assay chamber further flattens this response.



- *Figure 6: Measured response as a function of height above the rotator.*



- *Figure 7: Measured response for the X-axis, illustrating along the major extent of the hexagon. The dips at the extreme edges of the counter are due to the dead effects of the corner.*



- *Figure 8: Measured response for the Y-axis, illustrating the response profile along the minor extent of the hexagon.*

Background Measurements

For reference purposes, a long background run was performed for the empty cavity. The cycle times of 3600 seconds were utilized. The statistical filters were turned off for this measurement. See Table 4.

	Average Rate	Standard Deviation
Totals:	12.32 cps	0.09 cps
Reals:	1.13 cps	0.06 cps

- *Table 4: Background measurements*

Active Measurements

To simulate the proposed operation of the system, 3 Am(Li) sources were selected from Canberra's inventory. The nominal source information is shown in Table 5.

Source ID	Nominal Activity	Neutron Emission Rate	Date
N-457	1.20Ci	55000 +/- 10%	2/12/98
N-469	1.20Ci	55000 +/- 10%	9/19/98
N-470	1.20Ci	55000 +/- 10%	9/19/98

- *Table 5: Nominal Am(Li) source information*

To determine the relative activities for each of these sources, the source was measured in the center of the empty counter with a count time of 600 seconds in multiplicity mode. The measured values for the sources are recorded in Table 6.

Source	Singles	Doubles	Triples
N-457	19184.223 +/- 5.663	3.215 +/- 8.884	-4.866 +/- 6.963
N-469	16645.461 +/- 5.276	4.489 +/- 7.711	7.386 +/- 5.640
N-470	19367.883 +/- 5.688	-20.526 +/- 8.966	-15.199 +/- 7.062

- *Table 6: Active measurements*

It can be seen from the values shown in Table 6 that the activity for these sources varies significantly from source to source. Three sources were chosen to provide a more uniform irradiation of the sample. Each of the sources was re-measured, this time with the source installed inside the HDPE wedge on the door rack. The lower activity was placed at the vertical center of the source rail to provide a more uniform sample irradiation. The rates are recorded in Table 7. As can be seen in the table the error in the coincident rate is proportional to the source strength (as expected). The count time for each measurement was 30 minutes.

Source	Position	Singles	Doubles	Triples
N-457	Bottom	14592.253 +/- 4.937	-5.232 +/- 6.752	-2.327 +/- 4.618
N-469	Middle	12663.544 +/- 4.607	14.250 +/- 5.881	4.899 +/- 3.746
N-470	Top	14556.931 +/- 4.931	-4.683 +/- 6.738	10.382 +/- 4.611
All 3	----	41530.969 +/- 8.338	11.579 +/- 19.263	-27.561 +/- 22.181

- *Table 7: Measurement results with source inside the HDPE wedge on the door rack*

Passive Uranium Measurements
The 5 uranium sources containing approximately 980 grams of U-238 were placed in the empty assay cavity and a passive assay was performed (i.e. no Am(Li) sources). The following net count rates were observed:

Singles	Doubles	Triples
4.424 +- 0.143	0.776 +- 0.048	0.142 +- 0.023

The net doubles rate above includes the spontaneous fission neutrons from U-238 and the increased background due to cosmic-ray induced neutron events. The increased background rates for uranium can be estimated from the measured background rates from lead. The cosmic ray induced neutron background rates due to lead are:

Singles	Doubles	Triples
0.034 cps/kg/sec	0.028 cps/kg/sec	0.019 cps/kg/sec

The rates for uranium will be somewhat higher due to its increased cross-section for cosmic-rays. However, the spontaneous fission events account for approximately 90% of the observed coincidence rate for the 980 gram Uranium sample.

Active Uranium Response
Due to the low U-235 mass, the two uranium highest enriched standards (15 grams U-235 total) were assayed for a 10-hour period in order to obtain useful statistics. The sources were position near the outer edge of the rotator with the motor off. The measured rates were:

Singles	Doubles	Triples
41209.941 +- 1.082	11.545 +- 2.482	6.932 +- 2.845

The resulting count rate in the empty chamber provides an approximate calibration coefficient of 0.77 cps/gram U-235. For a source in the center of the assay cavity or a rotating source, this value will be approximately, 0.1 cps/gram U-235. The LLD for the empty assay cavity will then be approximately 500 grams U-235. MCNP modeling indicates that the introduction of even modest quantities of moderator will increase this sensitivity. Measurements utilizing the expected shipping container will need to be performed to determine the calibration coefficient and detection level.

Note: It is anticipated that for smaller container sizes, a moderating insert for the Am(Li) sources will be used. This will improve the source coupling and resulting statistics.

Parameter Summary for HNMC
Table 8 presents a summary of the calibration parameters for the HNMC.

High Voltage	1740 V
Pre-delay time	4.5 usec
Die-Away time	45.3 usec
Gate Width	64 usec
Dead time Parameters (NCC)	a = $102.3 \times 10^{-9} \pm 2.2 \times 10^{-9}$ sec , b = 0 $usec^2$
Dead time parameters (multiplicity)	c = $22.61 \times 10^{-9} \pm 1.15 \times 10^{-9}$ sec, d = $39.05 \times 10^{-9} \pm 13.15 \times 10^{-9}$ sec
Dead time parameter (τ)	26.27 ± 1.29 nsec
Doubles Gate Fraction	0.6151 ± 0.0003
Triples Gate Fraction	0.3933 ± 0.0010
Efficiency (Cf-252 point source)	0.339 ± 0.005
Cf-252 ρ_0	0.3324± 0.0005
Cf-252 a	261.0 ± 1.6 cps/ nanogram Cf-252
Efficiency (Pu-240 estimated)	0.346 ± 0.005
Pu-240 ρ_0	0. 1837 ± 0.0017
Pu-240 a	66.2 ± 0.4 cps / g Pu-240 effective

Dead Time Parameters for INCC 4.03	
Dead time parameter (τ)	$\tau = 26.27 \pm 1.29$ nsec
Dead time Parameters (NCC)	$a = 102.3 \times 10^{-9} \pm 2.2 \times 10^{-9}$ sec , $b = 0$ $usec^2$
Triples Dead time parameter	$c = 18.22 \times 10^{-9} \pm 13 \times 10^{-9}$ sec

- *Table 8: Calibration parameters for HNMC empty chamber*

CONCLUSION

The Transportable Measurement Facility with the HNMC installed in it was completed on schedule and is now being deployed. The HNMC has been demonstrated to meet all of the known and contingent requirements of the MCC as they were known at the time the mobile measurement unit was designed.

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