LA-UR-15-29225
Approved for public release; distribution is unlimited.

Title: Implementation and Initial Testing of Advanced Processing and Analysis Algorithms for Correlated Neutron Counting

Author(s):
Santi, Peter Angelo
Cutler, Theresa Elizabeth
Favalli, Andrea
Koehler, Katrina Elizabeth
Henzl, Vladimir
Henzlova, Daniela
Parker, Robert Francis
Croft, Stephen

Intended for: Report

Issued: 2015-12-01

LA-UR-15-?????

Implementation and Initial Testing of Advanced Processing and Analysis Algorithms for Correlated Neutron Counting

Peter Santi, Theresa Cutler, Andrea Favalli, Katrina Koehler, Vlad Henzl,
Daniela Henzlova, and Robert Parker
Los Alamos National Laboratory

Stephen Croft
Oak Ridge National Laboratory

Abstract

In order to improve the accuracy and capabilities of neutron multiplicity counting, additional quantifiable information is needed in order to address the assumptions that are present in the point model. Extracting and utilizing higher order moments (Quads and Pents) from the neutron pulse train represents the most direct way of extracting additional information from the measurement data to allow for an improved determination of the physical properties of the item of interest. The extraction of higher order moments from a neutron pulse train required the development of advanced dead time correction algorithms which could correct for dead time effects in all of the measurement moments in a self-consistent manner. In addition, advanced analysis algorithms have been developed to address specific assumptions that are made within the current analysis model, namely that all neutrons are created at a single point within the item of interest, and that all neutrons that are produced within an item are created with the same energy distribution. This report will discuss the current status of implementation and initial testing of the advanced dead time correction and analysis algorithms that have been developed in an attempt to utilize higher order moments to improve the capabilities of correlated neutron measurement techniques.

Table of Contents

1. Introduction

One of the most distinctive and informative signatures of special nuclear materials (SNM) is the emission of correlated neutrons from either spontaneous or induced fission. The detection of correlated neutrons is currently associated with a variety of nondestructive assay (NDA) measurement techniques that detect, identify, characterize, and assay plutonium. Because the emission of correlated neutrons is considered a unique and unmistakable signature of plutonium and uranium, correlated neutron counting is frequently used in a variety of safeguards measurements across the DOE complex (and world) including materials accountability, material verification and weapons material inspections. While these measurement techniques have been effective over the past several decades for a wide range of measurement needs, a number of technical and practical limitations exist, especially within the algorithms that are used to process and analyze the detected neutron signals. Maximizing the amount and quality of information that is determined from detecting correlated neutrons is important for improving the capability of quantifying SNM.

The observables that are extracted from the neuron pulse train that is produced by a neutron counting system are the autocorrelation reduced factorial multiplet rates. These rates only have meaning for quantitative analysis of special nuclear materials if they can be interpreted in terms of known properties of the detection system and the physical properties of the item that are of interest. The most widely used method for interpreting the rates into physical properties of the item is to use some form of the point model [Hage and Cifarelli (1985); Bohnel (1985)] which possesses a number of assumptions concerning the conditions associated with measurement. The point model consists of analytical equations which can be inverted (or solved) to determine specific physical properties of an item based on the measured multiplet rates. Depending on the specific neutron detection system that is being used, up to three measured rates are currently used to determine the properties of the item of interest. For these three rates (known as Singles (S), Doubles (D), and Triples (T) when determined using the Multiplicity Shift Register (MSR) technique), the point model expressions are well defined. In order to improve the accuracy and capabilities of neutron multiplicity counting, additional quantifiable information is needed in order to address the assumptions that are present in the point model. Extracting and utilizing higher order moments (Quads (Q) and Pents (P)) from the neutron pulse train represents the most direct way of extracting additional information from the measurement data to allow for an improved determination of the physical properties of the item of interest. This report will discuss an initial evaluation of the feasibility of extracting those higher order moments from neutron pulse trains. In addition, progress on implementing novel algorithms that utilize these higher order moments into a dedicated software analysis tool will be presented along with initial evaluation results.

2. Relative Merits of Higher Order Moments (Quads and Pents)

As discussed in a previous report [Santi (2014)], equations to extract multiplet rates from a neutron pulse train were derived up to 5^{th} order and were extended to include additional pulse train analysis methods. The additional analysis methods allowed neutron multiplicity counting algorithms to be extended beyond the traditional MSR type analysis in order to evaluate the relative merits and potential advantages/disadvantages of the different approaches. In addition to standard MSR type analysis that utilizes a combination of Reals + Accidentals (R+A)-gates and A-gates, i.e. MIXED analysis, the information contained solely in the random A-gate was used to extract neutron multiplet rates, i.e. Randomly Triggered Inspection (RTI) formalism, and the correlated information from (R+A)-gate was used to develop a Signal Triggered Inspection (STI) formalism.

The equations for multiplet rates up to 5^{th} order for these analysis approaches were implemented into a dedicated software tool (Multiplicity Advanced AlGorithm Implementation Code (MAGIC)) under development within this project in order to evaluate the feasibility of extracting higher order multiplet rates (Quads and Pents) from realistic measured pulse trains. In this initial evaluation, a ^{252}Cf data set was measured in three medium-to-high efficiency thermal neutron well counters within a laboratory setting. The ^{252}Cf sources with a range of activities were used to evaluate the anticipated measurement uncertainties with increasing measured count rate. The well counters included the high efficiency Epithermal Neutron Multiplicity Counter (ENMC) [Menlove (2004)], 3-Ring Neutron Multiplicity Counter (3RNMC) [Ensslin (1998)] and medium efficiency Active Well Coincidence Counter (AWCC) [Menlove (1979)]. The efficiencies of these counters correspond to 65%, 42% and 33% for the ENMC, 3RNMC and AWCC, respectively. The AWCC was selected to evaluate the limits of higher order multiplet rate measurements with decreasing efficiency.

Neutron emission rates of the ^{252}Cf sources measured in each counter are summarized in Table 1 (as of 01/01/2010). Measurement times were all 2 hours to provide direct comparison of measurement uncertainties. The uncertainties were extracted from statistical scatter of individual measurements that were acquired in a series of short (10 s – 50 s long) cycles. The rates presented in this section are not corrected for dead time to provide an overview of measurement uncertainties for raw data. Implementation and effects of dead time correction for these higher order multiplet rates will be discussed in the following section.

Table 1: Overview of neutron emission rates of ^{252}Cf sources measured in ENMC, 3RNMC and AWCC (as of 01/01/2010).

Source ID	Emission rate [n/s]	Well counter
Cf2	1.5E+02	ENMC
Cf3	3.3E+02	ENMC
Cf4	6.7E+02	ENMC/3RNMC
Cf5	1.3E+03	ENMC
Cf9	1.9E+04	AWCC
Cf10	3.8E+04	3RNMC
Cf11	6.8E+04	ENMC
Cf12	1.3E+05	ENMC/3RNMC
A7-866	2.6E+05	ENMC/3RNMC
A7-867	5.1E+05	ENMC/3RNMC
A7-868	8.3E+05	AWCC
A7-869	1.1E+06	ENMC/AWCC
FTC-CF135	3.8E+06	ENMC/AWCC
FTC-CF1992	4.2E+06	ENMC/AWCC
SRCF335	1.0E+07	AWCC

The extracted multiplet rates up to Pents with corresponding uncertainties for 2 hour-long measurement times are summarized in Tables 2-4. Measurements of different ^{252}Cf sources listed in Table 1 span a period of several years and therefore the measured results presented in Tables 2-4 were decay corrected to a common reference date of 01/01/2010 to allow for a direct comparison among the different sources strengths. Results for the MIXED and RTI approaches are presented for comparison of their relative merits. Both analyses were performed using high frequency sampling of A-gate corresponding to 1 MHz, i.e. fast accidentals. The RTI analysis results include corresponding gate fractions as described in [Santi (2014)] so that the rates can be compared to the MIXED approach results.

The results presented in Tables 2-4 illustrate the complexity and challenges associated with reliable measurement of Quads and Pents. As can be seen from the presented results, a very good uncertainty (of the order of 1% or less) can be achieved in 2 hours for Quads for sources with low to medium count rates and when measurements are performed in high efficiency counters such as ENMC or 3RNMC (65% and 42% efficiency, respectively). Uncertainties of Pents for the same measurement time are noticeably larger; greater than 4% for all the measured ^{252}Cf sources. It can also be seen that the uncertainties for both Quads and Pents deteriorate dramatically with increasing measured count rates and decreasing efficiency. The measurement results from the AWCC illustrate the limits of higher order multiplet rate extraction and highlight the need for high efficiency in order to obtain statistically meaningful results for these rates. Comparison of RTI and MIXED approaches reveals slightly better uncertainties in the case of the RTI analysis, an observation in agreement with previous findings documented in [Henzlova (2012)].

Tables 2-4 also provide an important experimental demonstration of the effects of dead time on the measured multiplet rates. As can be seen in the case of all three well counters, Quads and Pents exhibit saturation trends or even turn to negative values with increasing count rate; an effect that was theoretically predicted in [Hauck (2012)]. Saturation effects are observed also for Doubles and Triples rates albeit at reduced level. Negative Triples values are observed for the lowest efficiency counter in this comparison. A closer look at the presented results also reveals saturation differences between the RTI and MIXED approaches. Namely, the RTI results tend to saturate or turn negative sooner (i.e. for lower Singles rates) than MIXED results. Also this effect supports predictions in [Hauck (2012)].

Table 2: Correlated multiplet rates up to Pents from RTI (top) and MIXED (bottom) approaches with corresponding measurement uncertainties for range of ^{252}Cf sources measured in the ENMC. The rates presented in this table have not been corrected for dead time and were decay-corrected to a reference date of 01/01/2010 for direct comparison.

Source	S_{RTI}	σ [%]	D_{RTI}	σ [%]	T_{RTI}	σ [%]	Q_{RTI}	σ [%]	P_{RTI}	σ [%]
Cf2	96.5	0.226	62.0	0.35	23.1	0.6	5.1	1.7	0.3	14.3
Cf3	217.9	0.150	139.1	0.23	51.7	0.4	11.7	1.2	1.5	7.4
Cf4	425.4	0.106	280.8	0.16	104.5	0.3	23.7	0.9	3.1	4.2
Cf5	858.5	0.075	526.3	0.12	196.5	0.2	45.0	0.8	6.6	5.8
Cf11	43,736.2	0.012	28,675.1	0.02	10,430.3	0.1	2,183.5	0.6	217.7	9.5
Cf12	83,748.6	0.008	54,606.7	0.02	19,446.2	0.1	3,708.1	0.8	133.1	36.8
A7866	167,063.3	0.005	107,570.3	0.02	36,535.9	0.1	5,544.8	1.6	-604.9	27.6
A7867	322,197.3	0.004	201,672.4	0.02	61,187.5	0.2	3,114.7	7.6	-5,493.0	9.9
A7869	647,756.2	0.005	404,855.0	0.02	130,066.3	0.1	15,333.2	2.6	-4,282.8	17.5
FTC-CF135	2,483,229.7	0.002	1,380,217.9	0.02	257,824.5	0.4	-95,611.9	3.4	-49,247.6	20.5
FTC-CF1992	2,760,499.2	0.002	1,529,095.7	0.02	262,408.7	0.4	-122,935.9	3.2	-63,630.2	18.1

Source	S_{MIXED}	σ [%]	D_{MIXED}	σ [%]	T_{MIXED}	σ [%]	Q_{MIXED}	σ [%]	P_{MIXED}	σ [%]
Cf2	96.7	0.226	59.7	0.35	21.5	0.7	4.7	1.6	0.6	7.8
Cf3	218.1	0.150	134.0	0.22	48.4	0.4	10.9	1.3	2.6	22.1
Cf4	425.5	0.106	270.4	0.15	97.4	0.3	21.6	0.8	4.1	17.2
Cf5	858.6	0.075	507.0	0.11	182.8	0.2	41.0	0.7	7.0	6.2
Cf11	43,736.1	0.012	27,694.3	0.03	9,801.2	0.1	2,035.0	0.8	243.7	10.4
Cf12	83,748.6	0.008	52,875.3	0.02	18,441.7	0.1	3,530.7	0.6	144.2	43.0
A7866	167,063.2	0.005	104,742.8	0.02	35,307.3	0.1	5,689.6	1.9	-393.5	54.8
A7867	322,195.2	0.004	198,772.1	0.02	61,980.1	0.2	5,331.4	5.6	-4,654.1	15.8
A7869	647,755.6	0.005	397,610.5	0.02	128,849.7	0.2	17,372.5	2.9	-3,032.0	32.8
FTC-CF135	2,483,227.2	0.002	1,428,759.2	0.02	336,396.8	0.4	-56,570.1	7.4	-47,748.3	29.1
FTC-CF1992	2,760,499.2	0.002	1,593,788.2	0.02	363,137.4	0.4	-69,169.9	7.6	-64,646.6	25.9

Table 3: Correlated multiplet rates up to Pents from RTI (top) and MIXED (bottom) approaches with corresponding measurement uncertainties for a range of ^{252}Cf sources measured in the 3RNMC. The rates presented in this table have not been corrected for dead time and were decay-corrected to a reference date of 01/01/2010 for direct comparison.

Source	S_{RTI}	σ [%]	D_{RTI}	σ [%]	T_{RTI}	σ [%]	Q_{RTI}	σ [%]	P_{RTI}	σ [%]
Cf4	324.1	0.163	128.4	0.28	32.7	0.7	5.3	1.8	0.6	4.2
Cf10	16,717.7	0.020	6,983.0	0.05	1,703.3	0.2	253.3	1.4	23.3	12.9
Cf12	57,595.9	0.012	24,525.3	0.04	5,780.3	0.2	759.0	2.4	41.8	52.6
A7866	113,642.5	0.008	48,128.7	0.04	10,806.8	0.3	1,087.9	4.5	-104.2	68.4
A7867	219,398.1	0.006	90,758.4	0.04	18,357.8	0.4	896.4	16.6	-138.5	190.3

Source	S_{MIXED}	σ [%]	D_{MIXED}	σ [%]	T_{MIXED}	σ [%]	Q_{MIXED}	σ [%]	P_{MIXED}	σ [%]
Cf4	324.4	0.163	125.7	0.29	31.2	0.6	4.9	2.0	0.6	4.3
Cf10	16,717.7	0.020	6,911.1	0.06	1,673.2	0.3	246.6	1.7	22.6	16.8
Cf12	57,594.7	0.012	24,419.1	0.04	5,780.7	0.3	767.2	3.0	54.5	54.3
A7866	113,641.1	0.008	48,255.6	0.04	11,072.6	0.3	1,206.9	5.4	-78.5	133.6
A7867	219,398.4	0.006	92,550.9	0.04	20,105.8	0.5	1,671.8	12.0	348.8	111.9

Table 4: Correlated multiplet rates up to Pents from RTI (top) and MIXED (bottom) approaches with corresponding measurement uncertainties for a range of ^{252}Cf sources measured in the AWCC. The rates presented in this table have not been corrected for dead time and were decay-corrected to a reference date of 01/01/2010 for direct comparison.

Source	S_{RTI}	σ [%]	D_{RTI}	σ [%]	T_{RTI}	σ [%]	Q_{RTI}	σ [%]	P_{RTI}	σ [%]
Cf9	4,993.1	0.038	1,278.8	0.10	180.8	0.4	15.5	2.4	1.2	21.6
A7868	209,429.2	0.006	47,443.5	0.05	3,307.1	1.2	-627.3	8.9	-115.2	61.7
A7869	256,590.2	0.005	56,381.2	0.05	3,020.6	1.7	-1,125.3	6.8	-274.1	37.4
FTC-CF135	960,500.6	0.003	112,900.7	0.08	-34,134.3	0.8	-3,072.9	24.9	3,240.3	45.8
FTC-CF1992	1,044,757.4	0.002	109,091.1	0.10	-40,843.5	0.8	-1,516.6	52.1	4,570.9	38.0
SRCF335	3,354,135.5	0.001	-651,981.1	0.03	47,316.2	2.0	77,305.0	4.8	-78,770.3	15.8

Source	S_{MIXED}	σ [%]	D_{MIXED}	σ [%]	T_{MIXED}	σ [%]	Q_{MIXED}	σ [%]	P_{MIXED}	σ [%]
Cf9	4,992.8	0.038	1,296.1	0.09	185.5	0.4	15.9	2.6	0.9	26.7
A7868	209,428.3	0.006	52,216.4	0.06	5,376.2	1.0	-287.3	27.3	-100.2	111.5
A7869	256,589.4	0.005	63,371.2	0.05	5,940.6	1.1	-622.2	17.9	-214.0	75.5
FTC-CF135	960,496.6	0.003	202,528.4	0.05	-9,911.9	3.7	-6,299.4	16.2	1,532.0	161.2
FTC-CF1992	1,044,756.6	0.002	215,888.5	0.06	-13,513.9	3.1	-7,441.1	16.5	5,938.7	48.0
SRCF335	3,354,135.5	0.001	353,723.5	0.07	-157,436.9	0.8	66,792.4	8.0	-37,170.0	51.8

A separate set of ^{252}Cf measurements had been taken within an operating facility in 2004 with the ARIES Multiplicity Counter, which has a detection efficiency of 52% [Cremers (1998)]. As these measurements were performed within an operating nuclear facility, background Singles rates ranged between 1,500 and 2,000 cps. The measurements were 60 s cycles, for a total of 1 hour per source. Extracted multiplet rates up to Pents for RTI and MIXED gates are summarized in Table 5.

Table 5: Correlated multiplet rates up to Pents from RTI (top) and MIXED (bottom) approaches with corresponding measurement uncertainties for a range of ^{252}Cf sources measured in the ARIES Multiplicity Counter in a 1 hour count time. The rates presented in this table have not been corrected for dead time.

Source	S_{RTI}	σ [%]	D_{RTI}	σ [%]	T_{RTI}	σ [%]	Q_{RTI}	σ [%]	P_{RTI}	σ [%]
Cf06	6,855.7	0.054	2,776.5	0.169	838.2	0.635	150.2	3.47	14.7	32.74
Cf08	21,184.2	0.026	10,247.2	0.065	3,080.7	0.371	527.7	3.68	25.8	102.7
Cf10	83,984.5	0.011	43,349.7	0.049	12,311.5	0.446	1,629.0	7.85	-309.7	72.31
Cf11	153,478.1	0.008	78,956.7	0.057	20,796.5	0.578	1,918.5	14.94	-29.6	2065.1
Cf12	290,686.3	0.005	144,638.9	0.047	32,137.1	0.832	-1,045.4	94.21	-1059.4	259.9

Source	S_{MIXED}	σ [%]	D_{MIXED}	σ [%]	T_{MIXED}	σ [%]	Q_{MIXED}	σ [%]	P_{MIXED}	σ [%]
Cf06	6,855.7	0.054	2,753.6	0.068	825.3	0.276	148.2	1.62	13.9	16.8
Cf08	21,184.2	0.026	10,181.8	0.067	3,028.0	0.391	516.3	2.90	34.1	57.7
Cf10	83,984.5	0.011	43,430.7	0.062	12,496.6	0.422	1,693.6	8.72	-528.8	55.8
Cf11	153,478.1	0.008	79,765.0	0.075	21,662.9	0.772	1,974.2	27.18	-864.1	152.2
Cf12	290,686.3	0.005	148,830.8	0.063	36,477.7	1.148	910.0	182.92	2,907.1	167.7

Overall, the current results provide an initial, yet quite comprehensive assessment of the feasibility of experimentally extracting Quads and Pents. In particular, the results confirm the expected relationship between high neutron detection efficiency and uncertainty in the higher order moments counting rates. The results also demonstrate that for sufficiently high efficiency counters, uncertainties of 1% or less can be achieved in measurements of Quads for feasible measurement times of the order of 2 hours. Pents present a more challenging scenario due to their 5^{th} order dependence on the neutron detection efficiency. Although Pents suffer from noticeably worse uncertainty over ~2 hour measurement period, the full feasibility assessment can only be made once the actual requirements on uncertainty and its propagation into the final physics quantities are taken into account. Both Quads and Pents exhibit significant deterioration of uncertainty with increasing count rate. This may impact the feasibility of using the higher order rates for items which produce extremely large count rates within the counter.

Presented results provide a very useful initial step in the overall feasibility assessment. They will be further explored in conjunction with advanced analysis models developed within this project to fully assess the Quads and Pents feasibility and their uncertainty limits for practical use.

3. Implementation and Initial Testing of Advanced Dead Time Correction Methodologies

To create a common framework with which to implement and test the different combinations of dead time correction algorithms and advanced analysis algorithms that have been developed within this project, the Multiplicity Advanced AlGorithm Implementation Code (MAGIC) is currently under development which when completed will allow the user to process either list-mode data or data collected with a multiplicity shift register with either a MIXED or a RTI gating scheme and apply a choice of dead time correction algorithms (Dytlewski-Croft-Favalli algorithm [Croft 2014] or the Correlated Neutron Dead time model [Hauck (2013)]) to the data. In addition, the code will also have the capability of analyzing the dead time corrected data using one of three analysis models (Standard Point Model [Cifarelli (1986), Böhnel (1985)], Spatial Multiplication Model [Hauck (2014)], and the Two-Energy Point model) to determine the physical properties of the item being measured. The MAGIC software tool is being developed in a modular manner so that if one or more of the advanced algorithms are determined to produce improved results over the currently available analysis capabilities, the specific module associated with those algorithms could be readily integrated into existing neutron analysis packages such as the International Neutron Coincidence Code (INCC) [Krick 2010].

A. Implementation of Dytlewski-Croft-Favalli Dead Time Corrections

As a means to test the implementation of the dead time corrections that have been developed in this project, previously acquired data taken with the ARIES multiplicity counter were analyzed. The data come from an operating facility and thus is representative of scenarios where these algorithms will be used in the future. This data included various ^{252}Cf sources, Plutonium Oxide (PuO_x) samples, and Pu metal samples. Background rates were also fully representative of an operating facility—Singles rates varied from approx. 180 counts per second (cps) for the small Pu metal samples to 2,000 cps for the ^{252}Cf data. The Dytlewski-Croft-Favalli (D-C-F) implementation was the primary focus for this data analysis using the MAGIC tool.

The ^{252}Cf sources ranged in source strength from ~9,000 cps to ~300,000 cps. The data was originally collected from 2004 to 2007. Note that all the results presented in this section were extracted using MIXED gates analysis technique in order to be directly comparable to INCC that utilizes this approach. To determine the appropriate value of the dead time parameter for use in the D-C-F correction, the dead time parameter was varied in the MAGIC software tool. As a starting point, the value of INCC multiplicity dead time parameter was chosen, because it corresponds to initial Dytlewski implementation incorporated within INCC [Dytlewski 1991]. The value of the parameter was then varied from the INCC value (50.7 µs) by +/- 20 µs. It was found that the D-C-F corrected rates yielded the most consistent results with the INCC dead time parameter (i.e. 50.7 µs). The D-C-F corrected results using this dead time parameter also resulted in the best match of the source strengths known from the source certificates. These results provided confirmation that the MAGIC software tool was functioning correctly with experimental data.

For all of the measurements performed with the various sized ^{252}Cf sources, the ratios of Triples, Quads, and Pents to Doubles were computed. Because ^{252}Cf sources predominately produce correlated neutrons via spontaneous fission and not induced fissions, the ratio of higher order counting rates to lower order counting rates is expected to be constant as a function of source strength and subsequently the measured Singles counting rate. Figure 1 shows the counting rate ratios as a function of the Singles counting rate within the ARIES counter. As can be seen in the top portion of Figure 1, which shows the Triples/Doubles counting ratio, the D-C-F dead time correction appears to produce consistent results with respect to the dead time corrections that are applied within INCC. For the ratio of Quads to Doubles shown in the bottom portion of Figure 1, the D-C-F corrections appear to be working reasonably well up to a Singles counting rate of 200,000 cps, given that the trend of the data went from having a distinct downward slope to being reasonable flat as a function of the Singles counting rate. The cause of the deviation that occurs above 200,000 cps is currently under investigation. While the D-C-F dead time corrected Quads rates appear to be quite stable as a function of Singles counting rate, the D-C-F dead time corrected Pents to Doubles ratio is inconclusive. While the dead time corrected Pents to Doubles ratio is more level as a function of the Singles counting rate relative to the dead time uncorrected (i.e. measured) ratio, the dead time corrected rate approaches zero for the second largest ^{252}Cf source that was measured with this counter. The statistical uncertainties for the Pents/Doubles ratio for these measurements have been estimated at 1%. Further work is currently ongoing to fully evaluate this trend.

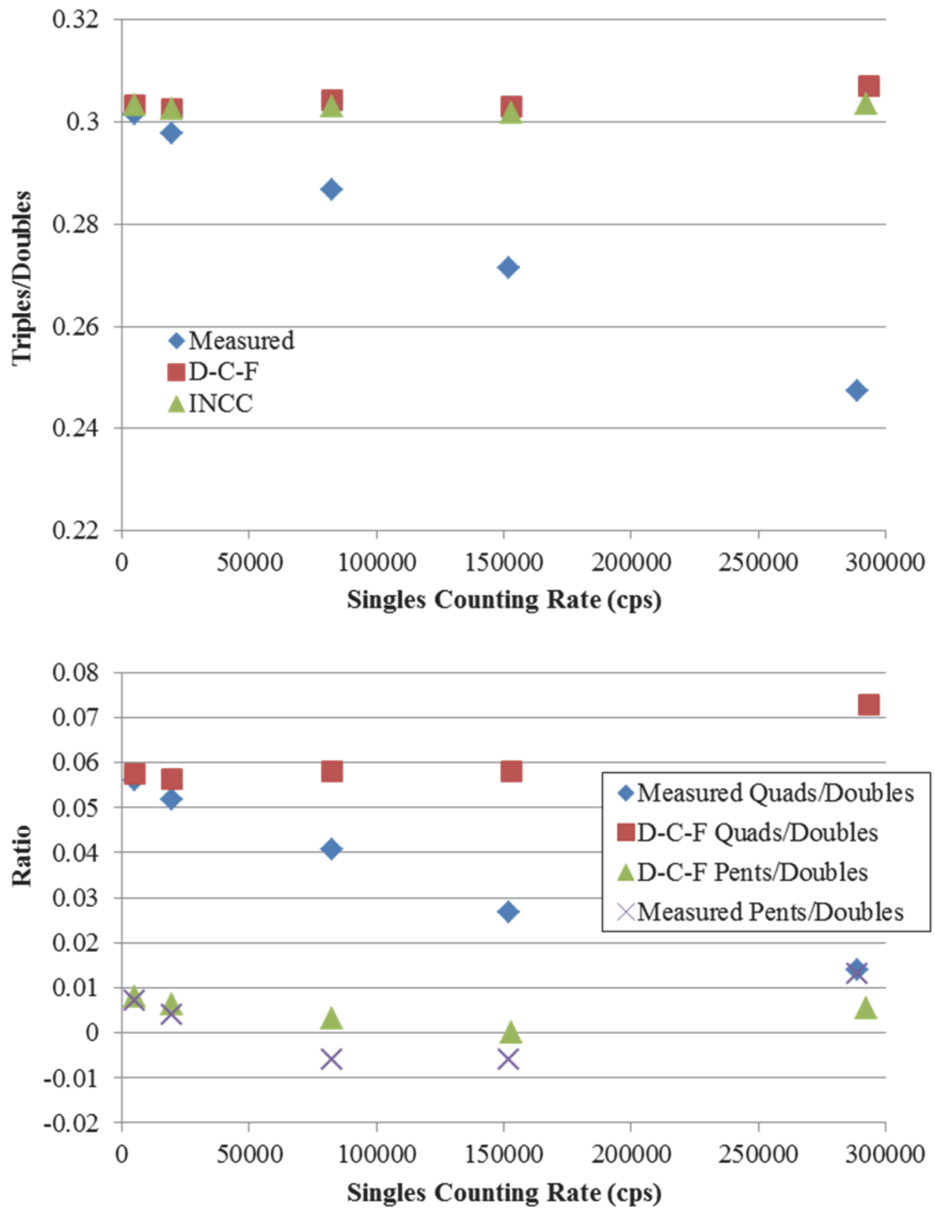

Figure 1. Ratio of counting rates as a function of the Singles counting rate for various ^{252}Cf sources as measured by the ARIES multiplicity counter and analyzed using MIXED gates. The top figure shows the measured Triples/Doubles count rate ratios (blue diamonds), and the dead time corrected ratios using the D-C-F correction (red squares) and from the INCC code (green triangles). The bottom figure shows the measured Quads/Doubles ratio (blue diamonds), the D-C-F dead time corrected Quads/Doubles ratio (red squares) and measured Pents/Doubles ratio (purple x) and dead time corrected Pents/Doubles ratio (green triangles).

Plutonium Oxide data of varying masses were also analyzed using the MAGIC software tool. The D-C-F dead time correction parameter of 50.7 µs as discussed above was used. The item properties calculated using INCC typically match the declarations within a few percent for PuOx samples. As such, benchmarking the D-C-F dead time corrected counting rates for PuOx data was done relative to the dead time corrected rates from INCC. The Singles, Doubles, and Triples rates from INCC were compared to the net (background subtracted) D-C-F MIXED gates from the MAGIC tool. The results for these values are shown in Figure 2 below. All samples match the INCC rates within one percent.

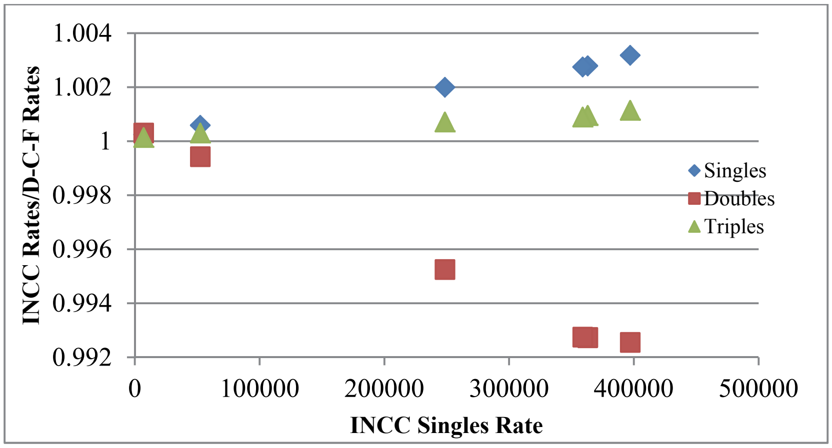

Figure 2. PuOx S,D,T ratios INCC to D-C-F Mixed Gates

The Quads and Pents rates were also evaluated with the MAGIC tool. INCC only calculates dead time uncorrected Quads and does not include calculation of Pents and therefore no direct reference for these dead time corrected rates is available. The comparison below should provide an illustration of the overall effect of the D-C-F dead time correction on the dead time uncorrected (i.e. measured) Quads and Pents rates for PuOx samples. Figure 3 below shows the comparison of measured and D-C-F corrected Quads and Pents rates from the MAGIC software tool. As can be seen, the effect using the D-C-F dead time correction is significant at samples with Singles rates greater than 200,000 cps. The dead time corrected rates will serve as input into the advanced analysis algorithms developed within the project to further evaluate the merit of higher order rates and use of advanced algorithms.

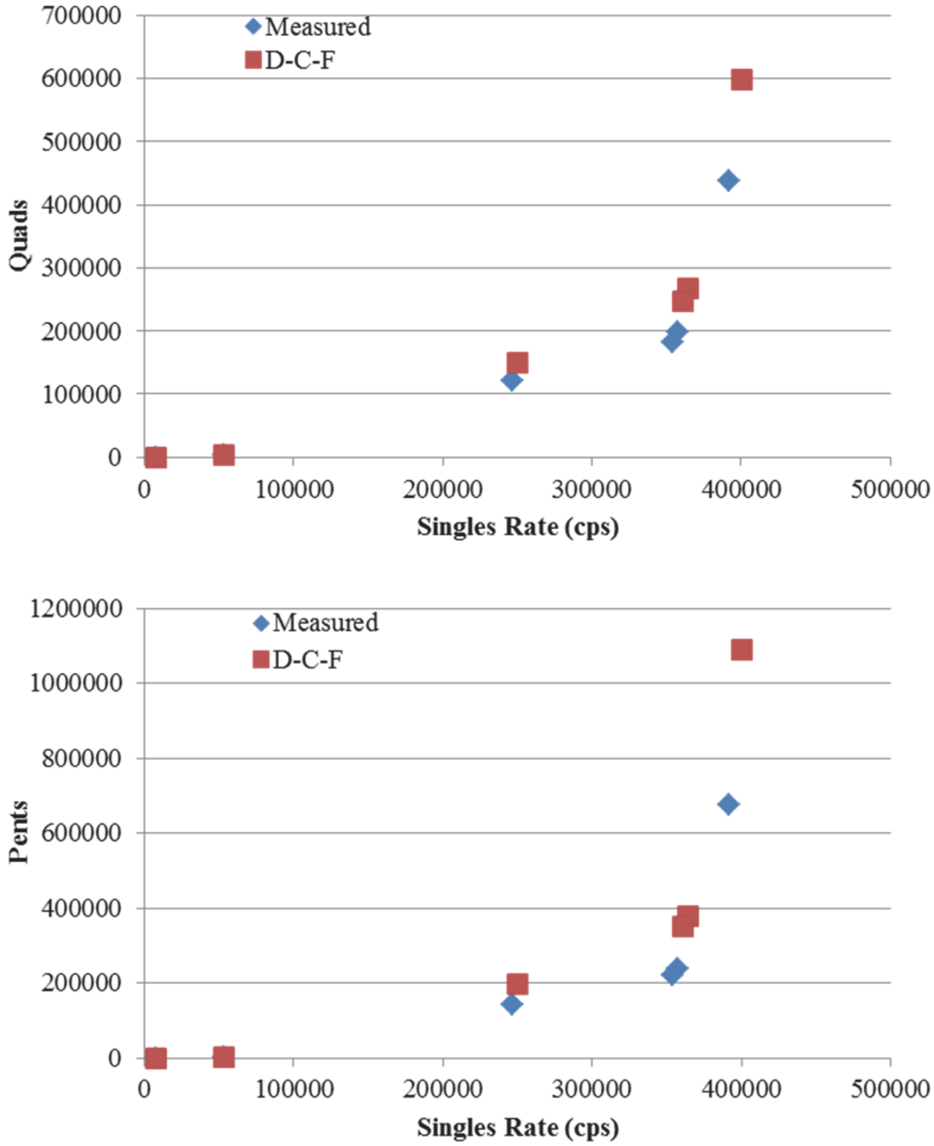

Figure 3. Quads and Pents rates for PuOx samples, measured and D-C-F dead time corrected.

The plutonium metal sample data was analyzed with the MAGIC software tool with a known total mass of 400 g (Singles rate of 27,600 cps). As with the ^{252}Cf and PuOx data, the Singles, Doubles, and Triples matched the INCC reported results. The Quads and Pents were also evaluated and will be used in the advanced analysis algorithms developed within the project. The mass, multiplication, and alpha of this data is explicitly known and the data will provide an invaluable resource for comparisons of the MAGIC reported values analyzed with the Standard Point Model to both the values determined using advanced algorithms as well as the declared values.

B. Status of Implementing the Correlated Neutron Dead Time Model

The implementation of the Correlated Neutron Dead Time Model (CNDTM) (previously referred to as the Exact Dead Time Model, EDTM) up to Triples was tested with ^{252}Cf pulse trains simulated for an AWCC using MCNPX [Pelowitz (2009)] PTRACs and the in-house developed software packages FastTrain and FastTap [Koehler (2015)]. MCNPX PTRACs contain neutron fission histories and relative capture times in the ^{3}He tubes of a simulated multiplicity counter. FastTrain applies a spontaneous fission rate to the neutron pulse train, resulting in a pulse train of absolute neutron capture times, similar to list mode data. This pulse train can be analyzed with FastTap. FastTap can extract standard shift register multiplicity rates (using a MIXED approach) from the list mode data and create Rossi-alpha distributions and time interval distributions. FastTap can read both simulated and experimental list mode data. For the simulated data, FastTap has extra features that allow for high fidelity simulations. FastTap is able to implement dead time structures on the list mode data before analyzing. This feature allows the user to analyze the same pulse train with and without dead time.

Multiplicity distributions were created from the original pulse train and from the pulse train with user-added dead time. The dead time applied to the pulse train was an updating dead time of 120 ns. Both multiplicity distributions were analyzed with the MAGIC software tool, and the multiplicity distribution with dead time was corrected using a dead time parameter δ=120 ns. This initial test was done to determine if the implementation of the dead time correction algorithms were done properly. Comparisons of Doubles/Singles with a CNDTM dead time correction and the Doubles/Singles values for the same data without dead time show agreement to within 0.2% when the uncorrected ratio diverged by as much as 7%. The same comparisons with a D-C-F correction show agreement to within 2.5%. Comparisons of Triples/Singles with a CNDTM dead time correction and the Triples/Singles values for the same data without dead time show agreement to within 23% when the uncorrected ratio diverged by as much as 60%. Discounting the last data point at 200 kHz, agreement is found to within 9%. The same comparisons with a D-C-F correction show agreement to within 9%.

The CNDTM has limitations in its current implementation within the MAGIC tool. The CNDTM code is unable to return a dead time correction when correcting for 120 ns updating dead time with neutron source strengths above 200 kHz. The cause of this problems and its correction are under investigation. The CNDTM as currently implemented in the MAGIC tool only includes corrections up to Triples and thus is truncated at the third order. The issues associated with large counting rates could be an indicator that corrections associated with higher orders may be needed and will be investigated in the next level of implementation that should extend CNDTM up to Quads.

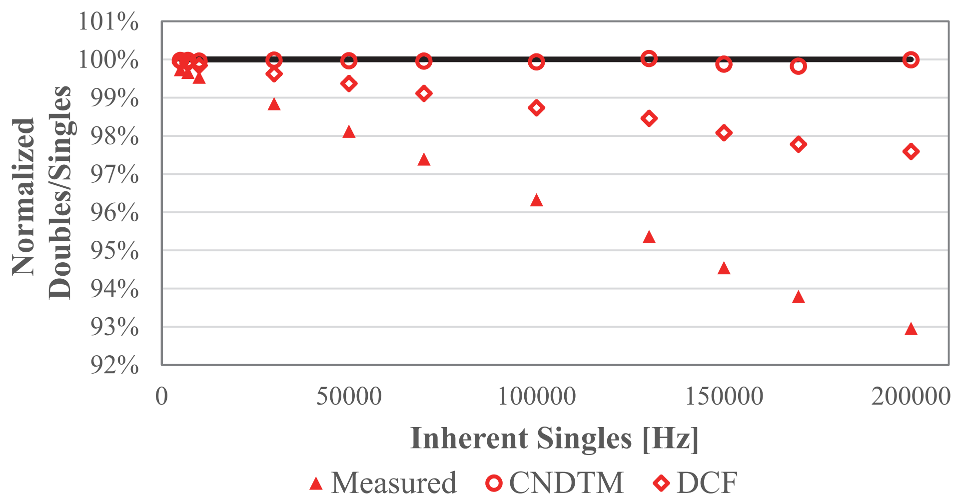

Figure 4. A plot of dead-time corrected Doubles/Singles normalized by the inherent Doubles/Singles as a function of inherent Singles, where inherent rates are multiplicity rates from a pulse train with no dead time. Using δ=120 ns for both the CNDTM and D-C-F dead time corrections yields dead time corrections to within 0.2% and 2.5% respectively.

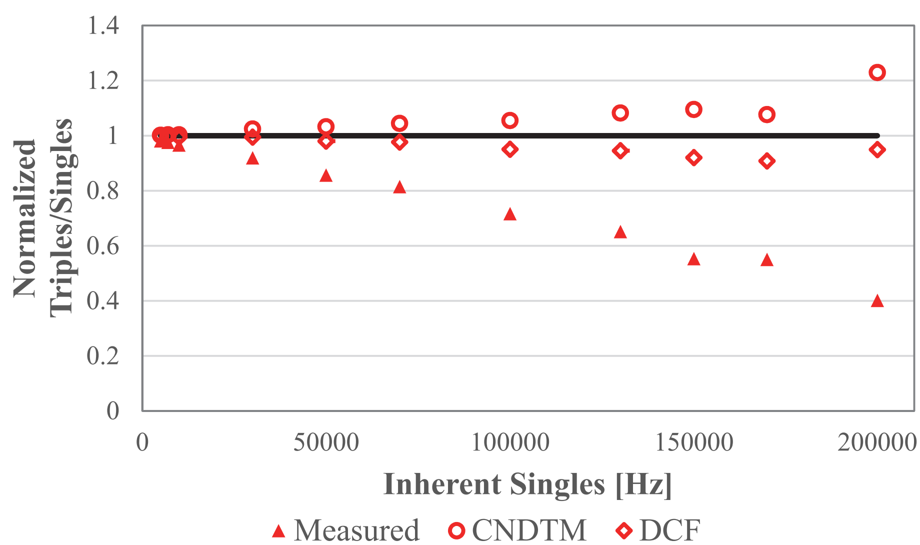

Figure 5. A plot of dead-time corrected Triples/Singles normalized by the inherent Triples/Singles as a function of inherent Singles, where inherent rates are multiplicity rates from a pulse train with no dead time. Using δ=120 ns for both the CNDTM and D-C-F dead time corrections yields dead time corrections to within 23% and 9% respectively.

4. Current Status of Implementing Advanced Analysis Models

The extraction of higher order moments from the emitted multiplicity distribution in a self-consistent manner provides a means for developing and implementing advanced analysis models. These advanced analysis models have the potential to improve the accuracy and capabilities of the Standard Point Model [Cifarelli (1986), Böhnel (1985)] which is currently used to determine the physical properties of the items being measured. Although the point model works well for the measurement scenarios for which it was developed, applying the technique to measurement scenarios which deviate significantly from the model assumptions results in inaccurate interpretations of useful data. The accuracy of the point model can be affected by several bulk material parameters such as composition, geometry (e.g. shape, dimensions), spatial distribution, reflectors, and chemical form of materials. Each of these sample characteristics causes the point model assumptions to fail to different degrees and therefore introduces bias in the assay results. While the material properties do not change the actual mass of Pu, they do affect the neutron multiplication (leakage and total) in the item, the neutron production by (α,n) reactions, and the efficiency by which the neutrons can be detected within a counter.

The two main assumptions that are made within the point model that have shown a demonstrative impact on the measurement results are the following: all of the fissions within an item occur at a single point in space [Langner (1993)], and that all of the neutrons that are produced within the item have the same energy distribution [Krick (1997)]. To determine if measuring higher order moments can remove the dependency of the analysis of neutron multiplicity counting on these assumptions, two advanced analysis models have been developed to address these assumptions: the Spatial Multiplication Model and the Two-Energy Point Model.

A. Spatial Multiplication Model

As a result of the assumption that all fissions occur at a single point within the item, the leakage multiplication (M_L) of an item is treated within the point model as a single average value. Treating M_L in this manner can have a major impact on the interpretation of higher order counting rates such as Doubles and Triples due to the fact that these counting rates are dependent on M_L^2 and M_L^3, respectively [Hauck (2014)]. Because the multiplication within an item is dependent on the location at which the initial neutron is produced, M_L is best described as a spatial distribution and not by a single value as is done in the point model. The discrepancy between treating M_L as a single value rather than a distribution affects the higher order counting rates and becomes larger as the multiplication increases within an item, which leads to an observable bias when measuring large mass or large multiplication items. A previous approach to correct this effect led to the development of the Weighted Point Model [Geist (2004), Krick (2005)] (WPM) which utilized Monte Carlo calculations to determine weighting factors that were used to correct the Doubles and Triples counting rates based on the type of detector and items that were being measured. To avoid the need for Monte Carlo calculations of each detector system, the Spatial Multiplication Model (SMM) has been developed in which the leakage multiplication of the item is assumed to follow a spatial distribution. To characterize the nature of the spatial distribution, an additional parameter, namely the Quads counting rate, is needed to solve for the item properties based on the selection of a specific spatial distribution to describe the leakage multiplication of the item. Initial testing of the SMM utilized MCNPX simulations of a wide range of Pu metal cylinders within the AWCC to illustrate the improvements that can be achieved using the SMM over the Standard Point Model. [Hauck (2014)]. The results of that testing showed that application of the SMM extended the capabilities of the AWCC to accurately measure the mass of Pu bearing items by expanding the range of items that could be accurately assayed the detector system to larger mass items. While the SMM showed an improved accuracy in measuring items with a larger leakage multiplication over the Standard Point Model, the SMM displayed the same trend as the point model in systematically failing for items with a leakage multiplication that was larger than 1.6. By contrast the point model begins to fail at a leakage multiplication of 1.2. In addition, the selection of the specific spatial distribution (cosine or Bessel function) within the SMM had no significant impact on the results of the measurements. As the SMM needs Quads to determine the properties of the measured item, the implementation of the SMM requires the implementation of the advanced dead time corrections to accurately extract the Quads counting rates from the measured multiplicity distribution. This effort is currently ongoing.

B. Two-Energy Point Model for Neutron Correlation Counting

Krick et al [1997] considered in some detail the energy dependent bias in the assay of plutonium dioxide by neutron multiplicity counting in moderated ^{3}He proportional counter wells. Their thrust was the quantification of the problem by Monte Carlo simulation. They looked at a variety of thermal-well designs from one to five full rings with markedly different efficiency vs. energy characteristics. They suggest that the ring ratio might be used as a spectral index from which the effective energy of the (α,n) component might be estimated. Within that paper, expressions for the Singles, Doubles, and Triples rates are presented without explanation as to their derivation.

As the previous work did not investigate the utilization of higher order moments to better describe the measurement scenario with two separate neutron energy groups, the derivation of the two-energy point-model is presented below and extended to include equations for Quads and Pents. One distinct difference between the current derivation and the prior work is that the probability of parasitic capture in the measurement item is finite is retained within the current model. By setting the probability of capture to zero for both neutron energy groups, the expressions for Singles, Doubles, and Triples rates in the current model agree with those of Krick et al [1997] although they are expressed differently.

Santi and Geist [2005] build on the work of Krick et al [1997] to develop an energy-dependent variant of the WPM for when the model parameters, especially self-leakage multiplication, exhibit spatial dependence across the measurement item. In deriving the energy-dependent WPM Singles, Doubles, and Triples equations Santi and Geist make two simplifying assumptions. First that the detection efficiency is the same for spontaneous fission neutrons as for (α,n) neutrons, and second that the probability for an (α,n) neutron to induce fission is the same as the probability for a spontaneous fission spectrum neutron. Currently, the Two-Energy Point Model has been restricted to a point item rather than the geometrically extended item model. However in the current model, both the efficiency and the probability of fission are treated as different for (α,n) neutrons than for fission neutrons.

Neutrons are created through four processes, spontaneous fission, (α,n) reactions, induced fission, and delayed neutrons. For the present discussion it is assumed that both spontaneous and induced fission neutrons have a sufficiently similar energy spectrum that they may be treated as one energy group. Delayed neutrons are weak and usually ignored in the conventional point-model expressions and so they are ignored here too. Logically they may be treated as an additional source of (α,n)-like neutrons because they are emitted long after the fission event, and collectively, with the character of being random in time compared to the time scales over which the correlated neutron groups exit in the system. The second energy group in our refined model is for (α,n) neutrons (including delayed neutrons). The (α,n) spectrum can be quite different, and significantly harder or softer than the fission spectrum depending on the relative abundance of target materials. Examples include the light atom fraction of Li, Be, B, C, and F with respect to Pu.

It is noted that given that the (α,n) neutrons can have a different energy spectrum than neutrons from fission, the detection efficiency from (α,n) neutrons leaking from the items will be different than for fission neutrons. Also the probability to induce fission inside the item is different. However, it is important to realize that these differences only affect the outcomes of the first generation of neutrons that are produced by (α,n) reactions and not the next generations of neutrons that have been subsequently generated by induced fission due to the fact that the later generation of neutrons will be emitted at fission energies and can therefore be described with the fission neutron energy parameters. Neutrons from (α,n) reactions directly contribute to the Singles rates but only to the higher order rates through subsequent induced fission. The induced fission from (α,n) neutrons can thereby be treated as a fission source for the subsequent generation of neutrons that may be produced.

The way to generate the two energy expression for a given rate from the corresponding one energy version is to sum the two contributions. The first contribution is the expression for a pure spontaneous fission source (with α, the ratio of (α,n) tospontaneous fission neutron production, set to zero), and the second is that for the (α,n) induced fission source, treated as if it were a pure spontaneous fission source, but with factorial moments of $\nu_{\alpha 1}$, $\nu_{\alpha 2}$, $\nu_{\alpha 3}$, etc. chosen appropriately for (α,n) induced fission.

Using the standard single energy point model as derived for all orders as a foundation [Favalli 2015], the following extra terms are being introduced in the two energy model:

> a_ε the ratio of the detection efficiency, count per neutron, for neutrons created in (α,n) reactions that emerge from the item to the detection efficiency from neutrons created in fission processes that emerge from the item. $\varepsilon_\alpha = a_\varepsilon \varepsilon$ is the detection efficiency of (α,n) neutrons leaking from the surface of the measurement item. In the one energy case it is common for ε, the detection efficiency for fission neutrons, to be known. This introduces the additional assumption that scattering in the measurement item does not change the energy spectrum significantly from item to item so that ε can be calibrated empirically.
>
> a_p the ratio of the probability that a neutrons created in an (α,n) reaction will induce fission in the item the corresponding probability for neutrons created in fission. $p_\alpha = a_p p$ is the probability that (α,n) neutrons will induce fission.
>
> $\nu_{\alpha 1}$, $\nu_{\alpha 2}$, $\nu_{\alpha 3}$, etc. the first and second and higher factorial moments of the induced-fission prompt-neutron multiplicity distribution resulting from (α,n) neutron interactions. In treating (α,n) induced fission as a steady state fission source term, $\nu_{\alpha 1}$, $\nu_{\alpha 2}$, $\nu_{\alpha 3}$, etc. replace the spontaneous fission factorial moments. Assuming that scattering in the matrix does not perturb the launch spectrum, one may calculate the factorial moments from basic nuclear data in the first level of approximation, by folding the energy dependence over the induced fission production channel, to create single energy (group) parameters, as follows:
>
> $$\nu_{\alpha j} \approx \int_{E=0}^{\infty} \chi_{\alpha n}(E)\sigma_f(E)\nu_{\alpha j}(E)dE \bigg/ \int_{E=0}^{\infty}\int_{E=0}^{\infty} \chi_{\alpha n}(E)\sigma_f(E)dE$$
>
> where $\chi_{\alpha n}(E)$ is the (α,n) emission spectrum and $\sigma_f(E)$ is the microscopic fission cross section. This expression explicitly shows the two energy model may more accurately be thought of as a ‘two effective energy spectrum or group’ description. Additional knowledge about the (α,n) spectrum is also needed– perhaps based on an understanding of the item generation process or prior study of similar items drawn from the same measurement stream.

Under the assumption that the probability of parasitic neutron capture in the measurement item is small compared to the probability of fission, given that a neutron can only be captured, cause fission, or leak from the item, one can approximate the leakage probability for (α,n) neutrons as $(1 - p_\alpha)$. In terms of the probability of fission by fission neutrons p, this quantity can be written as:

$$(1 - p_\alpha) = 1 - a_p p = 1 - a_p\left(\frac{M - 1}{M\nu_{I1} - 1}\right) = \frac{M(\nu_{I1} - a_p) + (a_p - 1)M/M}{M\nu_{I1} - 1}$$

where M is the leakage self-multiplication factor of the item for fission energy neutrons.

The rate at which (α,n) neutrons induce fission, giving rise to prompt fission neutron factorial moments of $\nu_{\alpha 1}$, $\nu_{\alpha 2}$, $\nu_{\alpha 3}$ etc., is $p_\alpha F_\alpha$ where F_α is the (α,n) production rate. This source term can be expressed in terms of fission neutron quantities as follows:

$$p_\alpha F_\alpha = a_p p F_s \nu_{S1} \alpha = F_s \alpha a_p \nu_{S1} \left(\frac{M-1}{\nu_{I1}-1}\right)\left(\frac{\nu_{I1}-1}{M\nu_{I1}-1}\right)$$

where the definition of $\alpha = F_\alpha/(F_S \nu_{S1})$ has been used as the ratio of (α,n) to spontaneous fission neutron production. This particular form has been chosen because the factor $\left(\frac{M-1}{\nu_{I1}-1}\right)$, which is the usual point model surrogate for "$p_f M_T$ in the limit that "$(p_c/p_f)/(\nu_{I1}-1) \ll 1$" , appears in the higher order point model expressions , and it will be useful to gather terms in powers of $\left(\frac{M-1}{\nu_{I1}-1}\right)$ as will be seen later.

With these few concepts and substitutions the two energy point model equations are as follows.

$$S = \varepsilon \mathrm{F}_S \nu_{S1} M \left[1 + \alpha \left(\frac{a_p(M-1)\nu_{\alpha 1} + a_\varepsilon\left[(\nu_{I1} - a_p) + (a_p - 1)/M\right]}{M\nu_{I1} - 1}\right)\right]$$

$$D = \varepsilon^2 f_d \mathrm{F}_S M^2 \left[\frac{\nu_{S2}}{2} + \left(1 + \alpha a_p \left(\frac{(M-1)\nu_{\alpha 1} + (\nu_{I1} - 1)\,\nu_{\alpha 2}/\nu_{I2}}{M\nu_{I1} - 1}\right)\right)\nu_{S1}\left(\frac{M-1}{\nu_{I1}-1}\right)\frac{\nu_{I2}}{2}\right]$$

Where f_d is the double gate fraction.

Derivation of expressions for Triples, Quads and Pents is based on the extension of the equations for the one-energy point model reported in Favalli et al [Favalli, 2015], developed in the frame of this project. In writing the expressions, it is convenient to introduce two additional quantities:

$$Y = \frac{(\alpha, n)induced\ fission\ rate}{SF\ rate} = a_p p_f \alpha \nu_{S1}$$

and

$$Z = "p_f M_T" = \frac{(M_L - 1)}{\nu_{I1} - \left[1 + (p_c/p_f)\right]}$$

as before, the equations work exclusively in terms of leakage self-multiplication for fission spectrum neutrons, i.e. $M \equiv M_L$.

Here is the expression for Triples:

$$T = f_t \frac{F_S(\varepsilon M)^3}{6}[\Omega_T(\nu_{S1},\nu_{S2},\nu_{S3}) + Y\Omega_T(\nu_{\alpha1},\nu_{\alpha2},\nu_{\alpha3})]$$

Where f_t, is the gate fraction, and

$$\Omega_T(\nu_1,\nu_2,\nu_3) = \nu_3 + (\nu_1\nu_{I3} + 3\nu_2\nu_{I2})Z + 3\nu_1\nu_{I2}^2 Z^2$$

The expression for Quads is given by:

$$Q = f_q \frac{F_S(\varepsilon M)^4}{24}\left[\Omega_Q(\nu_{S1},\nu_{S2},\nu_{S3},\nu_{S4}) + Y\Omega_Q(\nu_{\alpha1},\nu_{\alpha2},\nu_{\alpha3},\nu_{\alpha4})\right]$$

Where f_q is the gate fraction for Quads, and

$\Omega_Q(\nu_1,\nu_2,\nu_3,\nu_4) = \nu_4 + (\nu_1\nu_{I4} + 4\nu_2\nu_{I3} + 6\nu_3\nu_{I2})Z + (10\nu_1\nu_{I2}\nu_{I3} + 15\nu_2\nu_{I2}^2)Z^2 + 15\nu_1\nu_{I2}^3 Z^3$For Pents:

$$P = f_p \frac{F_S(\varepsilon M)^5}{120}[\Omega_P(\nu_{S1},\nu_{S2},\nu_{S3},\nu_{S4},\nu_{S5}) + Y\Omega_P(\nu_{\alpha1},\nu_{\alpha2},\nu_{\alpha3},\nu_{\alpha4},\nu_{\alpha5})]$$

Where f_p is the gate fraction for Pents, and

$\Omega_P(\nu_1,\nu_2,\nu_3,\nu_4,\nu_5) = \nu_5 + (\nu_1\nu_{I5} + 5\nu_2\nu_{I4} + 10\nu_3\nu_{I3} + 10\nu_4\nu_{I2})Z + (\nu_1(15\nu_{I2}\nu_{I4} + 10\nu_{I2}^3) + 60\nu_2\nu_{I2}\nu_{I3} + 45\nu_3\nu_{I2}^2)Z^2 + (105\nu_1\nu_{I2}^2\nu_{I3} + 105\nu_2\nu_{I2}^3)Z^3 + 105\nu_1\nu_{I2}^4 Z^4$

In general, for an order N, the expression is given by:

$$N = f_N \frac{F_S(\varepsilon M)^N}{N!}[\Omega_N(\nu_{S1},\nu_{S2},\dots,\nu_{Sn}) + Y\Omega_N(\nu_{\alpha1},\nu_{\alpha2},\dots,\nu_{\alpha N})]$$

And the $\Omega_N(\nu_{S1},\nu_{S2},\dots,\nu_{Sn})$ is calculated following algorithms introduced in [Favalli 2015].

The new equations reported may help to improve the accuracy of Pu mass assay for items with largeα ratios.

5. Summary

A series of ^{252}Cf measurements using different multiplicity counting systems have been performed to determine practical feasibility of extracting and accurately measuring higher order moments (Quads and Pents) from a neutron pulse train. The initial results indicated that precisions on the order of a few percent can be achieved for Quad counting rates for high efficiency (40% or greater) counting systems within a 2 hour measurement time. The tendency of the measured Quads and Pents values to produce negative values as the Singles counting rate increased within the counters illustrated the strong reliance on a dead time correction to extract accurate values for these counting rates.

The initial implementation of the D-C-F dead time correction for higher order rates was tested in the MAGIC software tool using previously recorded data from ^{252}Cf and Pu measurements that were performed using the ARIES multiplicity counter within an operating nuclear facility. The results of that testing indicated that implementation of the D-C-F dead time correction for Singles, Doubles and Triples produced dead time corrected rates that were in agreement with the rates produced by INCC. For the Quads counting rates, the application of the D-C-F dead time correction appeared to work appropriately based on the fact that the dead time corrected Quads/Doubles ratio was relatively flat as a function of the Singles counting rate which is what would be expected when measuring a pure spontaneous fission neutron source. Further evaluation will be performed to fully understand and if possible resolve this feature.

Implementation of the CNDTM dead time correction is currently in progress. Initial testing of the implementation of the CNDTM up to Triples within the MAGIC software tool was performed using high-fidelity simulations of AWCC measurements of ^{252}Cf sources. The results indicate that while the correction on the Doubles appear to be implemented quite well, the dead time corrected Triples rate deviates from the known rates that were produced in the simulations for high Singles rates. As the current implementation for Triples does not include the correction terms for higher order moments, this may indicate the effect of truncation on the correction.

To address the shortcomings of the point model in assuming that neutrons are produced within an item of interest with the same energy distribution, a Two-Energy Point Model has been developed which treats the neutrons from (α,n) reactions as having a different energy distribution from neutrons that are produced by either spontaneous or induced fission. The Two-Energy Point Model, along with the Spatial Multiplication Model that had been previously developed within this project, will be implemented into the MAGIC software tool once the dead time correction algorithms have been implemented.

6. Acknowledgements

This work was sponsored by the U.S. Department of Energy (DOE), National Nuclear Security Administration (NNSA), Office of Defense Nuclear Nonproliferation Research and Development (NA-22).

7. References

Böhnel, K. (1985) The effect of multiplication on the quantitative determination of spontaneously fissioning isotopes by neutron correlation analysis. *Nuclear Science and Engineering* (90) 75-82.

Cremers, T. L., T. Sampson, N. L. Scheer, and M. K. McDonald (1998) ARIES Nondestructive Assay System Description. Los Alamos National Laboratory Technical Report LA-UR-98-5337.

Croft, S., and A. Favalli (2014) Variations on and Extensions to Dytlewski's Dead Time Correction for Passive Multiplicity Counting. *Los Alamos National Laboratory Technical Report* LA-UR-14-29141.

Dytlewski, N. (1991) Dead-time corrections for multiplicity counters. *Nuclear Instruments and Methods in Physics Research A* (305) 492-494.

Ensslin, N., W.C. Harker, M.S. Krick, D.G. Langner, M.M. Pickrell, J.E. Stewart (1998) Application guide to neutron multiplicity counting, *Los Alamos National Laboratory Manual LA-13422-M*.

Favalli, A. and S. Croft (2014) Point Model Equations for Neutron Correlation Counting: Extension of Böhnel's Equations to Any Order, *Nuclear Instruments and Methods in Physics Research A* (795) 370-375.

Geist, W.H., M.S. Krick, and D.R. Mayo (2004) Reduction of Bias in Neutron Multiplicity Assay Using a Weighted Point Model. *Los Alamos National Laboratory Technical Report LA-UR-04-1149*.

Hage, W. and D.M. Cifarelli (1985) On the factorial moments of the neutron multiplicity distribution of fission cascades, *Nucl. Instr. Meth. A* (236) 165-177.

Hage, W. and D.M. Cifarelli (1985) Correlation Analysis with Neutron Count Distributions in Randomly or Signal Triggered Time Intervals for Assay of Special Fissile Materials, *Nuclear Science and Engineering* (89) 159-176.

Hauck, D. K. (2012) An Exact Dead Time Model for Correlated Neutron Counting based on the Joint Probability Function. *Los Alamos National Laboratory Technical Report* LA-UR-12-24194.

Hauck, D. K., S. Croft, L. G. Evans, A. Favalli, P. A. Santi, and J. Dowell (2013) Study of a theoretical model for the measured gate moments resulting from correlated detection evens and an extending dead time. *Nuclear Instruments and Methods in Physics Research A* (719) 57-69.

Hauck, D. K. and V. Henzl (2014) Spatial Multiplication Model as an alternative to the Point Model in Neutron Multiplicity Counting. *Los Alamos National Laboratory Technical Report* LA-UR-14-21991.

Koehler, K.E., V. Henzl, D. Hauck, D. Henzlova, and P.A. Santi, (2015) Characterizing Dead Time of Neutron Multiplicity Counters Using Rossi-Alpha Distributions. *Proceedings of the 56th Annual INMM Meeting*, Indian Wells, CA, July 12-16, 2015. Los Alamos National Laboratory Technical Report LA-UR-15-25187.

Krick, M.S., D.G. Langner, and J.E. Stewart (1997) Energy-dependent bias in plutonium verification measurements using thermal neutron multiplicity counters. *Los Alamos National Laboratory Technical Report* LA-UR-97-3427.

Krick, M.S., W.H. Geist, and D.R. Mayo (2005) A Weighted Point Model for the Thermal Neutron Multiplicity Assay of High-Mass Plutonium Samples. *Los Alamos National Laboratory Report* LA-14157.

Krick, M. B. Harker, W. Geist, and J. Longo (2010) INCC Software Users Manual. *Los Alamos National Laboratory Technical Report LA-UR-10-06277*.

Langner, D.G., M.S. Krick, D.R. Parks, and K.S. Hooper (1993) Thermal Neutron Multiplicity Measurements Using the Pyrochemical Multiplicity Counter at Lawrence Livermore National Laboratory. *Proceedings of the 34th Annual INMM Meeting*, Scottsdale, AZ, July 18-21, 1993. Los Alamos National Laboratory Technical Report LA-UR-93-2610.

Menlove, H.O (1979) Description and Operation Manual for the Active Well Coincidence Counter. *Los Alamos National Laboratory Report* LA-7823-M.

Menlove, H.O., C.D. Rael, K.E. Kroncke, K.J. DeAguero (2004) Manual for the Epithermal Neutron Multiplicity Detector (ENMC) for Measurement of Impure MOX and Plutonium Samples, *Los Alamos National Laboratory Technical Report, LA-14088*.

Pelowitz, D.B., M.R. James, G.W. McKinney, J.W. Durkee, M.L. Fensin, J.S. Hendricks, S.G. Mashnik and L.S. Waters (2009) MCNPX 2.7.B Extensions, *Los Alamos National Laboratory Technical Report LA-UR-09-04150.*

Reilly D., N. Ensslin, H. Smith, Jr., and S. Kreiner (1991) Passive nondestructive assay of nuclear materials, *Los Alamos National Laboratory Technical Report LA-UR-90732*.

Santi, P.A. and W.H. Geist (2005) Energy Dependent Bias in the Weighted Point Model, *Proceedings of the 46th Institute of Nuclear Materials Management Annual Meeting, July 10-14 2005, Los Alamos National Laboratory Technical Report* LA-UR-05-4287.

Santi, P., A. Favalli, D. K. Hauck, V. Henzl, D. Henzlova (2014) Derivation of Higher Order Moments for Neutron Multiplicity Analysis Methodologies, *Los Alamos National Laboratory Technical Report* LA-UR-15-20496.