LA-UR-17-27526 (Accepted Manuscript)

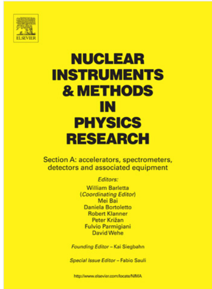
Experimental evaluation of the extended Dytlewski-style dead time correction formalism for neutron multiplicity counting

Lockhart, Madeline Louise
Henzlova, Daniela Constance
Croft, Stephen
Cutler, Theresa Elizabeth
Favalli, Andrea
McGahee, Christopher Oliver
Parker, Robert Francis

To be published in: Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment

DOI to publisher's version: 10.1016/j.nima.2017.09.025

Accepted Manuscript



Experimental evaluation of the extended Dytlewski-style dead time correction formalism for neutron multiplicity counting

M. Lockhart, D. Henzlova, S. Croft, T. Cutler, A. Favalli, Ch. McGahee, R. Parker

PII: S0168-9002(17)30995-6
DOI: http://dx.doi.org/10.1016/j.nima.2017.09.025
Reference: NIMA 60104

To appear in: *Nuclear Inst. and Methods in Physics Research, A*

Received date : 25 August 2017
Accepted date : 13 September 2017

Please cite this article as: M. Lockhart, D. Henzlova, S. Croft, T. Cutler, A. Favalli, C. McGahee, R. Parker, Experimental evaluation of the extended Dytlewski-style dead time correction formalism for neutron multiplicity counting, *Nuclear Inst. and Methods in Physics Research, A* (2017), http://dx.doi.org/10.1016/j.nima.2017.09.025

Experimental Evaluation of the Extended Dytlewski-Style Dead Time Correction Formalism for Neutron Multiplicity Counting

M. Lockhart[1,#], D. Henzlova[1], S. Croft[2], T. Cutler[1], A. Favalli[1], Ch. McGahee[1,*], R. Parker[1]

[1]Nuclear Engineering and Nonproliferation Division, Los Alamos National Laboratory, Los Alamos, NM 87545, USA

[2]Global Nuclear Security Technology Division, PO Box 2008, Bldg 5700, MS-6166, Oak Ridge, TN 37831-6166, USA

Abstract

Over the past few decades, neutron multiplicity counting has played an integral role in Special Nuclear Material (SNM) characterization pertaining to nuclear safeguards. Current neutron multiplicity analysis techniques use singles, doubles, and triples count rates because a methodology to extract and dead time correct higher order count rates (i.e. quads and pents) was not fully developed. This limitation is overcome by the recent extension of a popular dead time correction method developed by Dytlewski. This extended dead time correction algorithm, named Dytlewski-Croft-Favalli (DCF), is detailed in reference [1], which gives an extensive explanation of the theory and implications of this new development. Dead time corrected results can then be used to assay SNM by inverting a set of extended point model equations which as well have only recently been formulated. The current paper discusses and presents the experimental evaluation of practical feasibility of the DCF dead time correction algorithm to demonstrate its performance and applicability in nuclear safeguards applications. In order to test the validity and effectiveness of the dead time correction for quads and pents, ^{252}Cf and SNM

Corresponding author: henzlova@lanl.gov
#Texas Tech University, 2500 Broadway, Lubbock, Texas 79409
*Pennsylvania State University, 201 Old Main, University Park, Pennsylvania 16802

sources were measured in high efficiency neutron multiplicity counters at the Los Alamos National Laboratory (LANL) and the count rates were extracted up to the fifth order and corrected for dead time. In order to assess the DCF dead time correction, the corrected data is compared to traditional dead time correction treatment within INCC. The DCF dead time correction is found to provide adequate dead time treatment for broad range of count rates available in practical applications.

Keywords: neutron multiplicity counting, dead time correction, quads, pents

1. Introduction

The identification and characterization of Special Nuclear Materials (SNM) is essential in safeguards measurements performed by the Department of Energy (DOE) and other agencies. SNM such as plutonium and uranium have unique signatures based on the emission of correlated neutrons from spontaneous or induced fission and many Non-Destructive Assay (NDA) techniques can be used to detect and extract the correlated rates to detect, identify, and quantify SNM. Current methods of correlated neutron counting are based on measurement of singles (S), correlated pairs (doubles, D) and, in multiplicity counting, correlated triplets (triples, T) [2]. Using the extracted correlated count rates, SNM can be characterized using point model equations, which relate the neutron coincidences to the properties of an item and allow for the calculation of ^{240}Pu effective mass, leakage multiplication, and random neutron contribution from (α,n) interactions in the item [2,3].

Passive Neutron Multiplicity Counting (PNMC) based on correlated rates up to triples is a highly developed and widely used assay method to quantify SNM forming a pillar of technical nuclear safeguards. However, with improved data acquisition techniques and analysis methods, the practical feasibility and implications of extending the neutron multiplicity counting beyond triples can now be assessed. Addition of higher order correlated rates (quads and pents) can expand experimental information that can in turn be used in the improved characterization of SNM. With high efficiency neutron multiplicity counters there is potential to measure higher order correlated rates, quads and pents, if current methods of analysis are extended to include their explicit calculation for utilization in SNM characterization. To be able to use these additional observables in practical applications, dedicated dead time correction algorithms must be developed. Both of these aspects were addressed in recently developed advanced dead time correction algorithm based on the popular dead time correction method developed by Dytlewski [4]. The Dytlewski-Croft-Favalli (DCF) dead time correction algorithm [1] was developed at Los Alamos National Laboratory (LANL) and extends the PNMC towards higher order correlated rates with self-consistent treatment of dead time. To actually use the dead time corrected quads and pents to assay SNM properties, extended point model equations have also recently been developed [5]. Evaluation of these equations and quads and pents importance in SNM characterization represents a separate subject of research and will be presented in a future publication.

The following sections present experimental evaluation of the DCF algorithm with the focus on 1/ examination of the feasibility of quads and pents measurement and 2/ evaluation of performance of the DCF dead time correction. The second evaluation includes measurements of ^{252}Cf sources and plutonium items. While the ^{252}Cf data is used to assess the DCF dead time correction for full range of correlated rates (singles through pents), the plutonium measurements focus on DCF performance in the characterization of plutonium using standard point model equation (i.e. relying on singles through triples only).

2. DCF Dead Time Correction and Analysis Parameters

The Dytlewski formalism for dead time correction was originally developed to correct correlated neutrons up to triples [4]. The DCF dead time correction algorithm provides an extension of the original Dytlewski equations toward higher order correlated rates (quads and pents) and was described in full detail in [1]. In addition to extending the PNMC beyond triples counting, the DCF also includes self-consistent dead time treatment of singles through triples. The original Dytlewski scheme corrected singles in an ad hoc way using empirical expressions [6]. The DCF algorithm therefore provides room for improved performance over traditional approaches currently in use for dead time correction of singles through triples count rates; in particular it offers potential to correct for anomalous behaviors such as negative correlated count rates. Negative correlated count rates (triples and less often doubles) are experimentally observed in challenging measurement scenarios involving materials with very high neutron emission rates and cannot be appropriately corrected using existing empirical dead time correction methods.

The DCF remains simple to apply. It takes the form of matrix multiplication and is described by a single effective dead time parameter. Furthermore, the DCF algorithm was developed for all modes of neutron pulse train analysis and therefore extends the traditional PNMC beyond the standard shift register-based analysis. In the process of acquiring data from a multiplicity counter, one of two methods is used. The most common mode of data acquisition is through the standard Multiplicity Shift Register (MSR). Another method is list mode data acquisition. The latter allows the complete neutron detection pulse train to be recorded and stored, making this approach the most flexible for data analysis [7]. With a complete record of all detection times, the data can be analyzed using a variety of approaches and is not limited by a reliance on MSR hardware. While MSR counting is limited to one method of analysis, data acquired in list mode can be analyzed with any of the following gating schemes: MIXED, RTI (Randomly-Triggered Inspection) and STI (Signal-Triggered Inspection).

The most widely used gating scheme in MSR counting is MIXED. This traditional analysis method combines information from signal-triggered gates and randomly-triggered gates to extract correlated rates. In the signal-triggered gates, the gate opens with each incoming neutron pulse after a short pre-delay, which serves as a pause to allow the detection efficiency to return to normal after the disruption caused by the incoming event. In the randomly-triggered gates, the gates open independently from the time in which a pulse or signal arrives. The RTI approach is solely based on the latter method and represents a variant on an approach developed in the field of reactor noise analysis [8]. On the other hand, STI only utilizes information contained in the signal-triggered gates. Extension of these approaches into PNMC was presented in [9,10].

For randomly-triggered gates, the sampling frequency is also taken into consideration. The main sampling techniques include consecutive sampling, fast accidental sampling (FAS), and sampling at neutron detection frequency (i.e. corresponding to the frequency of signal-triggered gate openings). In the consecutive sampling technique, gates are triggered one after another with no overlap. In the case of FAS, gates are overlapping and triggered at a fixed frequency.

With DCF all of these approaches can be used to extract and dead time correct correlated rates up to pents. This allows for comparison among these approaches for any potential improvements in the overall performance. Note that the STI method does not lend itself to a practical implementation [1] and therefore will not be included in this evaluation. The current experimental evaluation will focus on FAS sampling technique due to its improved statistical uncertainty over the other sampling methods [11].

2.1 Gate Width and Gate Utilization Factors

It was described in [12] that an optimum gate width can be determined for different modes of neutron pulse train analysis described above. However, the implementation of a variable optimum gate width in analysis is likely unrealistic in practical applications. In all current practical safeguards applications a single gate width is typically used. Therefore, for current analysis purposes, the gate width was set to a fixed value for each detector used for measurements and is specified in the detector parameters in Table 1.

Because the gate width is finite, all correlations may not be detected within the gate time. Taking into account the die-away time (τ), pre delay (T_p), and the gate width (T_g) parameters of a given detector, the gate utilization factor (GUF), i.e. the fraction of correlated neutrons detected within the gate time period, can be calculated using Eq. 1 and 2. The MIXED GUF is defined for doubles through pents as f_n, n=1-5 (also denoted as n=d,t,q,p in some resources). Similarly, RTI GUF is denoted by w_n, n=1-5. Note that f_1=w_1=1, meaning that the singles count rate is unaffected by the pre-delay and gate width. The determination of the GUF is vital because they are required in the RTI expressions for correlated rates that have been developed for the DCF correction [1] as well as in the point model expressions [2]. Note that DCF MIXED expressions for singles through pents are independent of GUF values; for the MIXED analysis GUF knowledge is only required in the point model equations for extraction of the SNM characteristics.

$$f_n = \left[e^{-T_p/\tau}\left(1 - e^{-T_g/\tau}\right)\right]^{n-1} = f_2^{n-1} \qquad \text{(Eq. 1)}$$

$$w_n = \sum_{k=0}^{n-1}(-1)^k \binom{n-1}{k}\left(\frac{1-e^{-kT_g/\tau}}{kT_g/\tau}\right) \qquad \text{(Eq. 2)}$$

Eq. 1 and 2 assume single die-away time approximation that is not always valid for neutron multiplicity detectors and in practice [13], GUF values are therefore typically determined experimentally [2]. However, it provides a good initial estimation and in the current analysis theoretical GUF values will be used to maintain consistency among the different pulse train analysis methods.

2.2 Dead Time Parameter and its Optimization

In order to use the DCF correction, an effective dead time parameter is required for each detector. The value of this parameter may not necessarily be the same across the different pulse train analysis methods. For the purpose of this analysis, the initial estimate for the dead time parameter is based on the INCC multiplicity dead time parameter for each detector used in the measurements. The values of these parameters are provided in Table 1.

To find the most suitable value of the dead time parameter for each analysis method and detection system used in this paper, further optimization of the dead time parameter was performed using a set of well characterized ^{252}Cf sources. A set of ^{252}Cf sources with varying neutron emission rates, but of similar age and impurities are a suitable choice because the ^{252}Cf sources do not have multiplication or random neutron contribution (i.e. (M=1) and (α,n)=0). Without multiplication and (α,n) production, the point model equations are simplified significantly [2]. Under this presumption, the count rate ratios (i.e. D/S, T/S, Q/S, P/S) are theoretically proportional to powers of the detector efficiency and should therefore be constant with increasing source strength if the optimum dead time treatment is implemented. Such analysis is therefore strongly indicative of the quality of dead time correction treatment and also allows for optimization of the dead time parameter.

3. Experimental Evaluation

The key focus of this experimental evaluation is two-fold – 1/ to evaluate feasibility of higher order count rate measurements and 2/ to evaluate performance of the DCF dead time correction algorithm for practical applications. To evaluate both of these aspects, measurements were performed at LANL with three neutron multiplicity counters and a wide range of ^{252}Cf and PuO_2 items. It is important that the counters have high neutron detection efficiency to extract higher order moments with less uncertainty and to allow investigation of feasibility of quads and pents measurements for practical applications.

The three detectors used were Epithermal Neutron Multiplicity Counter (ENMC) [14], ENMC with Inventory Sampling Counter (INVS) insert [15], and Advanced Recovery and Integrated Extraction System (ARIES) neutron counter [16]. ENMC and ENMC/INVS were used to measure ^{252}Cf sources while ARIES was used to measure PuO_2 sources at an operating nuclear facility. The individual detector parameters are summarized in Table 1. The dead time coefficients (A, B, δ) were used in the standard INCC dead time correction [6] that serves as a reference for DCF performance.

{space for Table 1}

ENMC is a ^3He-based detector containing 121 ^3He tubes arranged in 4 rings. The He^3 tubes are 1” (2.54 cm) in diameter and have an internal pressure of 10 atm. The central cavity is approximately 17.0” (43.2 cm) tall and 7.6” (19.4 cm) in diameter and the active length of the counter is 28.0” (71.1 cm). ENMC groups the 121 tubes into 27 channels equipped with AMPTEK amplifiers and a derandomizing buffer [17].

The performance of the ENMC is augmented by the addition of an INVS insert. The INVS was designed to be inserted into the sample chamber of ENMC for the assay of smaller items. It contains an additional 21 He^3 tubes arranged in two rings around the central cavity. The small sample chamber is 2.0” (5.1 cm) in diameter and 6.0” (15.2 cm) high and the active length of the insert is 20.0” (50.8 cm). With the insertion of the INVS, the efficiency of ENMC is increased by more than 20% relative, however it is limited to smaller diameter samples.

The ENMC and ENMC/INVS were used to measure series of ^{252}Cf sources and each source was measured for 900 s. An overview of sources available for measurements is provided in Table 2. The data from the ENMC and EMC/INVS counters was acquired in list mode using a Pulse Train Recorder (PTR32) list mode module that allows recording of up to 32 channels simultaneously [7]. The key advantage of list mode acquisition is flexibility in data analysis of the recorded complete pulse train and was chosen to enable both, MIXED and RTI analysis methods to fully test the DCF algorithm capabilities.

{space for Table 2}

To demonstrate practical feasibility of the DCF dead time correction algorithm, it is also desirable to evaluate its performance on materials of operational interest. For this purpose a range of PuO_2 items with different masses was measured in the ARIES multiplicity counter that resides in an operating facility at LANL. An overview of the measured PuO_2 items is summarized in Table 3. The measurements corresponded to 1 hour and data were acquired using standard MSR electronics and INCC software. Only MIXED DCF analysis was performed on this dataset to provide a direct comparison with the empirical dead time correction within INCC. The detector parameters (pre-delay, gate and efficiency) used in the analysis were taken over from INCC in order to allow for a direct comparison and are listed in Table 1. The GUF values used in the point model equations to calculate ^{240}Pu effective mass were also taken from INCC and correspond to f_d=0.6640, f_t=0.4482.

{space for Table 3}

3.1 Feasibility of Quads and Pents Measurements

In order to assess feasibility of quads and pents measurements, a range of ^{252}Cf sources with increasing neutron emission rates was measured and corresponding quads and pents uncertainties were evaluated for a fixed measurement time corresponding to 900 s. Longer measurement time would decrease the uncertainty, but 900 s was selected as a representative of a typical NDA assay. The measurements were acquired as a series of 90 cycles of 10 s each to allow for sample based error estimation. The results for MIXED and RTI analysis for ENMC and ENMC/INVS are shown in Tables 4 and 5, respectively, to provide a direct comparison between the two analysis approaches. Both analyses were performed using FAS. 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 the DCF dead time correction for these higher order correlated rates will be discussed in the following section.

{space for Table 4 and 5}

The dead time uncorrected rates presented in Tables 4 and 5 also provide an important experimental demonstration of the effects of dead time on the measured higher order correlated rates. As can be seen in the ENMC dataset, quads and pents exhibit saturation trends or even turn to negative values with increasing count rate; an effect that was theoretically predicted in [18]. The results presented in Tables 4 and 5 illustrate the complexity and challenges associated with reliable measurements of quads and pents. As can be seen in Table 4 and 5, a very good uncertainty (of the order of 5% or less) can be achieved in 900 s for quads for low to medium count rates (up to ~ 200 kHz) for ENMC and for high count rates (up to ~ 1 MHz) for higher efficiency ENMC/INVS. Uncertainties of pents for the same measurement time are noticeably higher for all the measured ^{252}Cf sources. In addition, pents uncertainties exhibit complex trends and can be prohibitively large for cases where measured pents turn to negative values. Note that saturation tendencies (or count rates turning negative) are generally observed for lower singles count rates as the multiplicity order increases. This is also confirmed by the ENMC data, where pents turn negative, an effect not seen in quads (nor doubles and triples) for the same singles count rate range. It can also be seen that the uncertainties for quads deteriorate with increasing measured count rates. Comparison of RTI and MIXED approaches reveals tendency towards slightly better uncertainties in the case of the RTI analysis, an observation in agreement with previous findings documented in [12].

Overall, the current results provide an initial assessment of the feasibility of experimental extraction of quads and pents. The results also demonstrate that for sufficiently high efficiency counters (>60% in this study), uncertainties of 5% or less can be achieved in quads for measurement times of the order of 900 s. Note that these uncertainties would drop to <1% level for a 2 hr measurement time, which may still be practically feasible in some applications. Pents present a more challenging scenario due to their 5th order dependence on the neutron detection efficiency. Although pents suffer from noticeably worse uncertainty over the 900 s measurement period, the full feasibility assessment can only be made once the actual requirements on uncertainty and its propagation into the final physics quantities (e.g. through the extended point model equations) are taken into account; an area that will be subject of further research.

3.2 Performance of DCF Dead Time Correction

The following results demonstrate the performance of the DCF dead time correction on series of ^{252}Cf sources and practical feasibility of this algorithm on a set of PuO_2 materials of operational interest. The results presented here contain a comparison between dead time uncorrected data, the DCF corrected data for both MIXED and RTI gating schemes with FAS, and INCC empirical dead time correction. The INCC dead time correction has only been developed for MIXED analysis algorithm up to triples and therefore the corresponding dead time corrected data is included for MIXED analysis only [6].

The DCF dead time correction was performed using an optimum dead time parameter selected for each analysis algorithm and detector combination. As discussed previously, the count rate ratios (*D*/*S*, *T*/*S*, *Q/S*, and *P/S*) for the measured set of ^{252}Cf sources should be constant and independent of the source strength. To find an optimum dead time parameter, its value was varied until an optimum was found based on this method. The initial dead time estimate for ENMC and ENMC/INVS corresponded to 37 ns and 100 ns, respectively, and was varied between 20-40 ns and 60-110 ns, respectively, to optimize the final dead time correction. The optimum dead time parameters for each detector are summarized in Table 6. The DCF dead time corrected results using the optimum dead time parameters are plotted with the uncorrected data and traditional INCC corrected results in Figures 1 and 2 for ENMC and ENMC/INVS, respectively. The ENMC/INVS dataset includes sources with very high count rates which extend the evaluation of the performance of the DCF algorithm towards more extreme measurement scenarios.

For the ENMC dataset the DCF corrected results exhibit rather constant trends, independent of source strength, for all correlated orders and both analysis methods (MIXED as well as RTI). The effect of dead time treatment relative to dead time uncorrected results is clearly visible. In addition, the DCF corrected results also demonstrate capability of this algorithm to correct for negative count rates observed in dead time uncorrected pents (Figure 1, bottom). The ENMC DCF results analyzed using MIXED approach (Figure 1, left) show very good agreement with

INCC for D/S as well as T/S ratios confirming validity of the DCF algorithm for this count rate range.

The ENMC/INVS dataset exhibits more complex trends. Evaluation of this set of high rate ^{252}Cf sources in a very high efficiency neutron counter provides a unique opportunity to access the effectiveness of DCF for more extreme experimental scenarios. In case of ENMC/INVS the optimum dead time parameter was chosen mainly based on the D/S ratios. The D/S trend is independent of source strength for both MIXED and RTI approaches, however ratios begin to increase as a function of count rate for the higher order correlations. It can be clearly seen that for count rate ratios beyond doubles the DCF exhibits upward trend for both, MIXED and RTI analysis. This suggests that a lower dead time parameter would be more appropriate for correcting triples through pents than required by optimum doubles correction. This is an interesting observation suggesting a potential dependence of dead time correction parameter on multiplicity order and will be further explored with more extensive datasets. A comparison with INCC highlights a similar trend also in this traditional dead time correction, where T/S ratio exhibits an upward trend similar to DCF. The D/S and T/S DCF results analyzed using MIXED approach (Figure 2, left) show very good agreement with INCC.

Overall, the ENMC results, that represent more typical count rate range in practical applications, indicate capability of DCF algorithm to effectively correct for dead time effects for all correlated rates. In addition, capability to correct for negative count rates observed in dead time uncorrected pents was demonstrated. The ENMC/INVS results provide a rather extreme case of count rates less often encountered in practical applications, which, however, represents an important test scenario for the full capability of the DCF. The ENMC/INVS DCF results for D/S and T/S exhibit similar trends as INCC, generally validating the DCF performance. However, observed trends for correlated rates beyond doubles indicate potential dependence of dead time correction parameter on multiplicity order.

{space for Figure 1 and 2}

To further test the implementation of DCF correction algorithm the practical application of the DCF dead time corrected correlated rates for SNM characterization was evaluated using datasets from plutonium bearing materials. The purpose of this evaluation is to determine the capability to extract and correct correlated count rates up to triples which are then used to calculate the effective mass of ^{240}Pu.

For this evaluation, the ARIES neutron counter was used to measure a range of Pu items with masses up to ~4.5 kg at an operational nuclear facility. Because an equivalent set of ^{252}Cf sources was not available in the facility for this counter, the multiplicity dead time parameter (50.7 ns) utilized in standard INCC dead time correction was used in the DCF formalism and varied to investigate its influence on the $^{240}Pu_{eff}$ mass. As discussed earlier, only MIXED analysis was performed on this dataset to provide a direct comparison with the standard INCC results.

Figure 3 shows a comparison of the mass ratio of measured versus declared (known) ^{240}Pu effective ($^{240}Pu_{eff}$) mass with the measured mass extracted using DCF dead time correction for several values of dead time parameter as well as using INCC dead time correction. As can be seen from Figure 3, the DCF results for dead time parameter of ~40.7 ns demonstrate good agreement with INCC. The dataset covers broad range of Pu masses up to ~4.5 kg and the results shown therefore provide an initial validation of the DCF algorithm performance and demonstrate its capability to provide results in close agreement with INCC, a nuclear safeguards standard. In other words, the DCF approach has passed the initial test, namely that it can be used in a way that preserves established performance norms if that is the goal.

{space for Figure 3}

4. Discussion

A summary of the optimum dead time parameter values for DCF for both analysis approaches (MIXED, RTI), and counters is shown in Table 6. Table 6 also includes the initial dead time parameter estimate based on the INCC multiplicity dead time parameter.

{space for Table 6}

Table 6 highlights that optimum dead time parameter is not necessarily the same for both analysis approaches (MIXED and RTI). In general, the MIXED analysis requires a higher value of the dead time parameter than RTI. Note that the optimum dead time parameter values for the MIXED approach are in very close agreement with the standard INCC multiplicity dead time parameter. This further validates selection of the latter value for the DCF analysis of PuO_2 materials measured in the ARIES counter.

It should be pointed out that the DCF expressions for the RTI rates depend on the corresponding GUF values [1]. As described in section 2.1., theoretical GUF values were used in the current evaluation. It can be anticipated that experimental GUF values could favor slightly different values of the RTI DCF dead time parameters, however it can be expected that the trends observed in the present evaluation (Figures 1-3) will remain largely unchanged. The choice of theoretical GUF values for this work was primarily motivated by a desire to maintain consistency among the different pulse train analysis methods and provides a robust initial assumption to assess the DCF performance for RTI analysis. A detailed study of experimental GUF values and their effect on the DCF RTI results will be subject of future work.

The optimum values of the dead time parameters presented in Table 6 were selected primarily based on the D/S ratio. However, the results in Figure 2 indicate a tendency towards lower optimum dead time parameter values for higher order correlated count rates, especially for quads and pents. This trend seems to be more pronounced for conditions more significantly affected by dead time, such as high count rates measured in high efficiency counters ENMC/INVS.

Although the optimum dead time parameter varies for fairly extreme count rates, the results shown in Figures 1-3 demonstrate that the DCF dead time corrected results provide adequate dead time treatment for typical measurement scenarios. In addition, the DCF extends the PNMC towards new capabilities in alternative analysis approaches (RTI) and extraction and dead time correction of higher order correlated rates.

5. Conclusions

This paper presented an experimental evaluation of performance of novel dead time correction algorithm based on traditional Dytlewski formalism. The DCF algorithm was developed to allow for extraction and dead time correction of higher order correlated rates (quads and pents) from measured pulse train and also to provide improved performance over existing empirical dead time correction methods that cannot, for example, correct for anomalies such as negative count rates. The key motivation in extending traditional PNMC towards higher order correlated rates lies in the potential to improve characterization of assayed SNM via additional experimental observables. The capability to measure and use higher order moments represents a new area of research within nuclear safeguards and its full potential is still being explored. The current paper focused on key aspects of the DCF dead time correction algorithm in order to establish its performance for potential use in nuclear safeguards applications for all multiplicity orders from singles up to pents.

The paper explored two key areas – 1/ evaluation of feasibility to experimentally extract quads and pents with sufficient uncertainty and 2/ performance evaluation of DCF dead time correction algorithm for all count rates from singles up to pents.

To appropriately address the two key questions, high efficiency neutron multiplicity counters were used for the measurements. The evaluation of measured quads and pents was performed on dead time uncorrected data to remove any uncertainties associated with the correction and reveal magnitude of the dead time effects. The measured quads and pents and their corresponding uncertainties for 900 s measurement time demonstrated statistically meaningful results for quads with uncertainties of ~5% or less over a broad range of count rates (0.3 – 1,000 kHz). The observed uncertainties for pents were noticeably worse (greater than tens of percent). Both, quads and pents exhibited saturation and also negative values with increasing count rate, that clearly require sophisticated dead time correction treatment to fully recover. The high pents uncertainties and often negative values represent an important observation that may indicate limited applicability of this experimental observable to certain measurement scenarios (high efficiency, however more moderate (< 100 kHz) neutron emission rates).

The evaluation of the DCF dead time correction included comparison of DCF dead time corrected count rate ratios (D/S, T/S, P/S, and Q/S) with uncorrected and (where applicable) INCC dead time corrected results. The correction was studied on series of well-characterized ^{252}Cf sources as well as range of PuO_2 items of operational mass range and included both, standard MSR analysis (MIXED) and alternative analysis based on randomly triggered gates only (RTI). The results revealed a very good performance of DCF over standard count rate range and for the broad range of PuO_2 items with masses up to 4.5 kg. In particular, good agreement with INCC type dead time correction was observed for the measured ^{252}Cf sources and PuO_2 materials. This serves as a demonstration of capability of DCF to correct for dead time effects in majority of measurement scenarios including items of practical interest. In addition, DCF demonstrated capability to correct negative count rates observed for some quads and pents.

High count rate measurement scenarios (500 – 1,000 kHz) were also evaluated with range of ^{252}Cf sources measured in 80% efficiency neutron counter. These results indicated potential dependence of dead time correction parameter on multiplicity order with lower dead time parameter values favored by higher order correlated rates (triples – pents) compared to doubles rates. A full implication and magnitude of this effect will have to be further evaluated.

The results presented provide a critical first step in the overall assessment of whether it is feasible to use quads and pents in practical safeguards measurements. They will be further explored in conjunction with advanced analysis models currently under development to fully assess the quads and pents measurement feasibility and their uncertainty limits for practical use. Further work will also focus on evaluation of influence of GUF values on the DCF RTI results.

6. Acknowledgements

This work was sponsored by the U.S. Department of Energy (DOE), National Nuclear Security Administration (NNSA), Office of Nonproliferation Research and Development (NA-22). We thank Dr. Peter Santi and Dr. Bill Geist for encouraging and supporting this work.

7. References

[1] S. Croft, A. Favalli, "Extension of the Dytlewski-Style Dead Time Correction Formalism for Neutron Multiplicity Counting to Any Order", Nuclear Instruments and Methods in Physics Research A 869, p. 141-152 (2017).

[2] N. Ensslin, W. C. Harker, M. S. Krick, D. G. Langner, M. M. Pickrell, J. E. Stewart, "Application Guide to Neutron Multiplicity Counting", Los Alamos National Laboratory Manual, LA-13422-M (1998).

[3] D. Reilly, N. Ensslin, H. Smith, Jr., and S. Kreiner, Passive Nondestructive Assay of Nuclear Materials, Los Alamos National Laboratory Technical Report LA-UR-90-732 (1991).

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

[5] A. Favalli, S. Croft, P. Santi, "Point Model Equations for Neutron Correlation Counting: Extension of Böhnel's Equations to Any Order", Nuclear Instruments and Methods in Physics Research A 795, p. 370-375 (2015).

[6] M. Krick, B. Harker, W. Geist, J. Longo, "INCC Software Users Manual", Los Alamos National Laboratory Technical Report, LA-UR-10-6227 (2009).

[7] D. Henzlova, H. O. Menlove, M. T. Swinhoe, J. B. Marlow, I. P. Martinez, C. D. Rael, "Neutron Data Collection and Analysis Techniques Comparison for Safeguards", IAEA-CN-184/178, IAEA nuclear safeguards symposium, Vienna, Austria (2010).

[8] R.P. Feynman, F. de Hoffmann, and R. Serber, J. Nuclear Energy, vol. 3 (1956) 64-69.

[9] S. Croft, A. Favalli, D. K. Hauck, D. Henzlova, P.A. Santi, "Feynman Variance-to-mean in the Context of Passive Neutron Coincidence Counting", Nuclear Instruments and Methods in Physics Research A 686 p. 136-144 (2012).

[10] S. Croft, D. Henzlova, and D. K. Hauck, "Extraction of Correlated Count Rates Using Various Gate Generation Techniques: Part I Theory", Nuclear Instruments and Methods in Physics Research A 691, p. 152-158 (2012).

[11] S. Croft, P. Blanc and N. Menaa, "Precision of the Accidentals Rate in Neutron Coincidence Counting", proceedings of Waste Management Symposium March 7-11, 2010 on Improving the Future by Dealing with the Past, Phoenix, Arizona, USA (2010).

[12] D. Henzlova, S. Croft, H. O. Menlove, M. T. Swinhoe, "Extraction of Correlated Count Rates Using Various Gate Generation Techniques: Part II Experiment", Nuclear Instruments and Methods in Physics Research A 691 (2012) p. 159-167.

[13] S. Croft, R. D. McElroy and S. C. Kane, "Coincidence gate utilization factors for neutron correlation counters with up to three components in the die-away profile", Proceedings of the ICEM '07: The 11th International Conference on Environmental Remediation and Radioactive Waste Management, Bruges (Brugge), Belgium, 2-6 September 2007, copyright ©2007 by ASME, ISBN 0-7918-3818-8, paper 7173 (2007).

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

[15] J. E. Stewart, H.O. Menlove, D.R. Mayo, W.H. Geist, L.A. Carrillo, G.D. Herrera, "The Epithermal Neutron Multiplicity Counter Design and Performance Manual: More Rapid Plutonium and Uranium Inventory Verifications by Factors of 5–20", Los Alamos National Laboratory Manual, LA-13743 (2000).

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

[17] J. E. Swansen, "Deadtime Reduction in Thermal Neutron Coincidence Counter", Nuclear Instruments and Methods in Physics Research B 9, p. 80-88 (1985).

[18] D. K. Hauck, S. Croft, L. G. Evans, A. Favalli, P. A. Santi, and J. Dowell, "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 p. 57-69 (2013).

List of Figures:

Figure 1: DCF corrected count rate ratios for a set of ^{252}Cf sources measured in ENMC as a function of singles rate. INCC comparison is only available for MIXED results. Error bars for D/S and T/S are smaller than the size of the symbols.

Figure 2: DCF corrected count rate ratios for a set of ^{252}Cf sources measured in ENMC/INVS as a function of singles rate. INCC comparison is only available for MIXED results. Note that dead time uncorrected Q/S and P/S ratios are shown on the right axis. Error bars for D/S and T/S are smaller than the size of the symbols.

Figure 3: Measured versus declared $^{240}Pu_{eff}$ mass extracted using DCF dead time correction algorithm compared with INCC.

List of Tables:

Detector	Pre-delay [s]	Gate Width [s]	Die-away [s]	Efficiency [%]	Dead Time Coefficient A (1E-6) [s]	Dead Time Coefficient B (1E-12) [s]	Multiplicity Dead Time δ [ns]
ENMC	1.5	24	21.8	65.0	0.0954	0.0289	36.8
ENMC/INVS	1.5	24	18.8	80.0	0.341	0.017	100.0
ARIES	3.0	64	51.0	51.8	0.182	0.000	50.7

Table 1: Operating parameters of multiplicity counters used in the evaluation.

Table 2: Overview of neutron emission rates of measured ^{252}Cf sources.

Source ID	Neutron emission rate [n/s]	Multiplicity counter	date
Cf4	6.7E+02	ENMC	01/01/2010
Cf11	6.8E+04	ENMC	01/01/2010
Cf12	1.3E+05	ENMC	01/01/2010
A7-866	2.6E+05	ENMC	01/01/2010
A7-867	5.1E+05	ENMC	01/01/2010
FTC-CF-1184	4.8E+05	ENMC/INVS	07/11/2016
FTC-CF-5065	1.1E+06	ENMC/INVS	07/11/2016
FTC-CF-2593	1.2E+06	ENMC/INVS	07/11/2016
FTC-CF-3097	2.0E+06	ENMC/INVS	07/11/2016

Table 3: Overview of the known characteristics of the PuO_2 materials measured in the ARIES system.

Source ID	Declared Pu mass [g]	Declared ^{240}Pu effective mass [g]
Pu1	748.7	45.4
Pu2	1492.6	90.8
Pu3	2996.4	181.6
Pu4	4430.7	260.1

Table 4: Singles count rate and correlated count rates for quads and pents for MIXED (top) and RTI (bottom) analysis methods with corresponding uncertainties for ^{252}Cf sources measured in ENMC.

Source	S_{MIXED}	σ [%]	Q_{MIXED}	σ [%]	P_{MIXED}	σ [%]
Cf4	426 ± 1	0.29	20.7 ± 0.4	1.8	2.7 ± 0.2	6.6
Cf11	43,690 ± 30	0.07	2,020 ± 55	3	222 ± 72	32
Cf12	83,739 ± 18	0.02	3,467 ± 110	3	13 ± 182	1,422
A7866	167,104 ± 24	0.01	5,227 ± 302	6	-1,637 ± 560	34
A7867	322,203 ± 34	0.01	4,332 ± 787	18	-6,420 ± 1,983	31
Source	SRTI	σ [%]	QRTI	σ [%]	PRTI	σ [%]
Cf4	426 ± 1	0.29	24.1 ± 0.8	3.2	3.8 ± 0.8	21.0
Cf11	43,691 ± 30	0.07	2,134 ± 44	2	171 ± 54	32
Cf12	83,739 ± 18	0.02	3,624 ± 93	3	-32 ± 137	427

A7866	167,102 ± 24	0.01	5,195 ± 247	5	-1,469 ± 442	30
A7867	322,203 ± 33	0.01	2,545 ± 649	26	-6,780 ± 1,503	22

Table 5: Singles count rate and correlated count rates for quads and pents for MIXED (top) and RTI (bottom) analysis methods with corresponding uncertainties for ^{252}Cf sources measured in ENMC/INVS.

Source	S_{MIXED}	σ [%]	Q_{MIXED}	σ [%]	P_{MIXED}	σ [%]
FTC-CF-1184	410,699 ± 42	0.010	-21,287 ± 1,280	6	-16,273 ± 3,433	21
FTC-CF-5065	910,678 ± 58	0.006	-80,018 ± 4,680	6	21,956 ± 13,732	63
FTC-CF-2593	985,405 ± 57	0.006	-85,939 ± 5,007	6	17,917 ± 15,042	84
FTC-CF-3097	1,566,867 ± 63	0.004	-75,923 ± 8,442	11	78,696 ± 32,485	41
Source	S_{RTI}	σ [%]	Q_{RTI}	σ [%]	P_{RTI}	σ [%]
FTC-CF-1184	410,699 ± 42	0.010	-27,283 ± 899	3	-13,280 ± 2,065	16
FTC-CF-5065	910,679 ± 58	0.006	-75,972 ± 3,150	4	29,520 ± 8,042	27
FTC-CF-2593	985,406 ± 56	0.006	-74,242 ± 3,545	5	38,864 ± 9,167	24
FTC-CF-3097	1,566,867 ± 63	0.004	-13,231 ± 6,137	-46	84,224 ± 19,951	24

Table 6: Summary of dead time parameters for each counter used in the evaluation of ^{252}Cf data.

Detector	INCC Dead time estimate [ns]	DCF MIXED [ns]	DCF RTI [ns]
ENMC	37	40	30
ENMC/INVS	100	105	70

Figure1

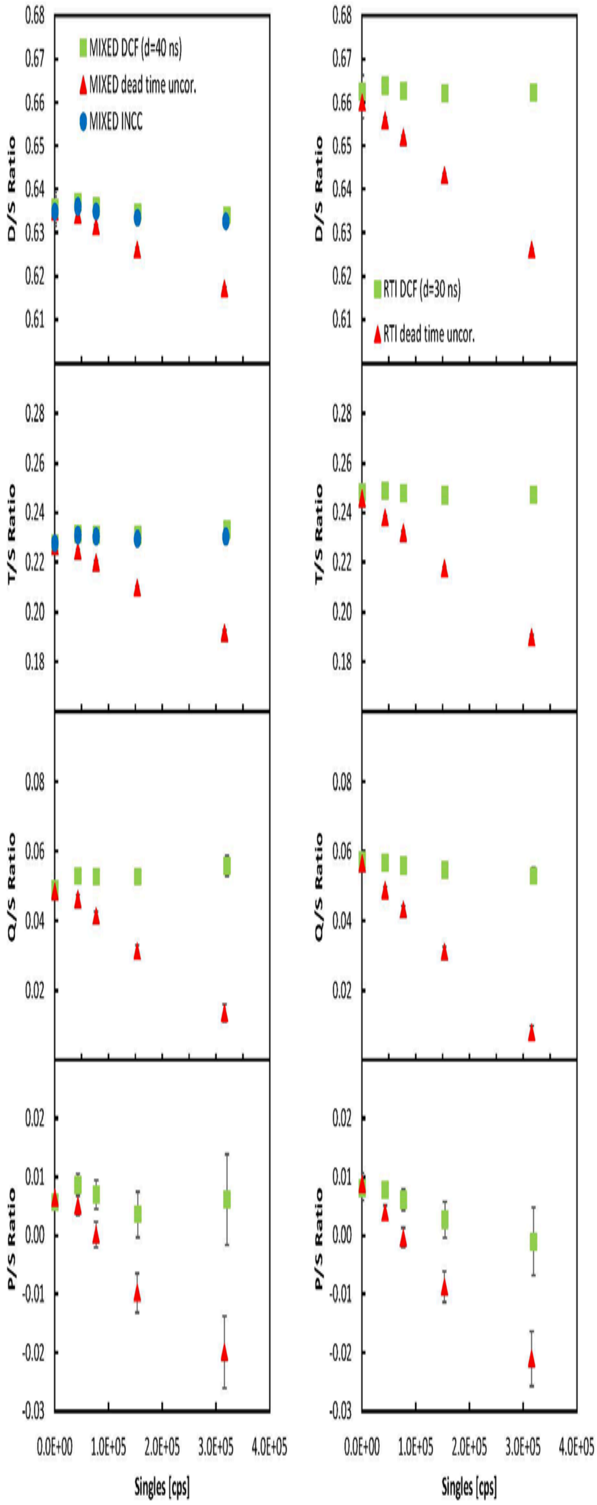
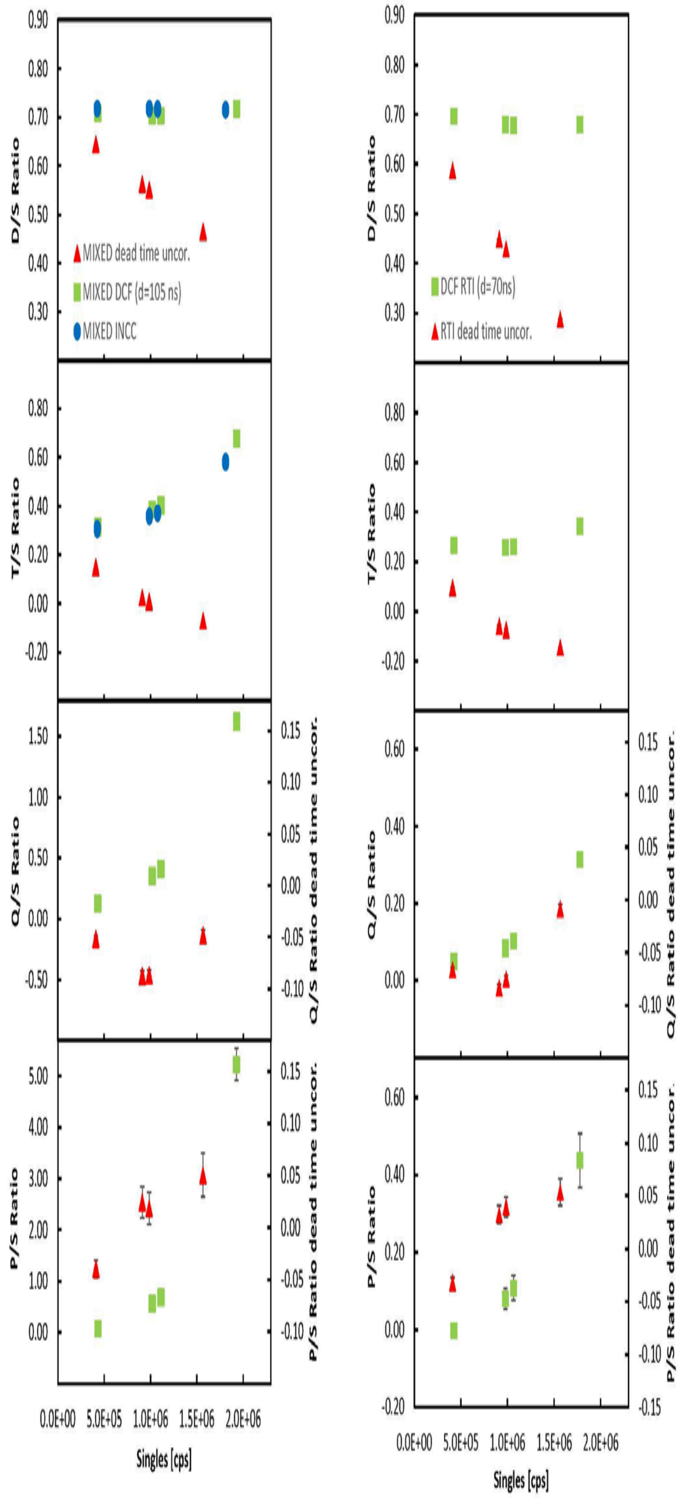
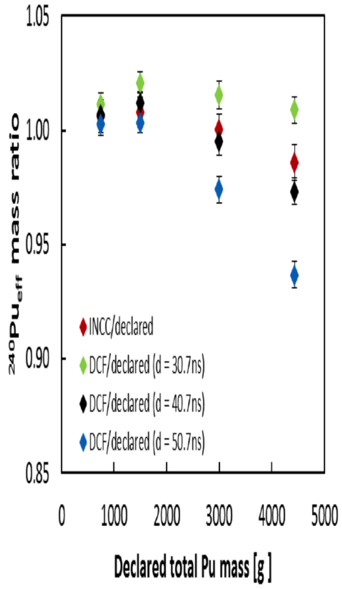

MIXED DCF (d=40 ns)
MIXED dead time uncor.
MIXED INCC
RTI DCF (d=30 ns)
RTI dead time uncor.
D/S Ratio
T/S Ratio
Q/S Ratio
P/S Ratio
Singles [cps]
0.0E+00
1.0E+05
2.0E+05
3.0E+05
4.0E+05

Figure2



MIXED dead time uncor.
MIXED DCF (d=105 ns)
MIXED INCC
DCF RTI (d=70ns)
RTI dead time uncor.
D/S Ratio
T/S Ratio
Q/S Ratio
P/S Ratio
Q/S Ratio dead time uncor.
P/S Ratio dead time uncor.
Singles [cps]
0.0E+00
5.0E+05
1.0E+06
1.5E+06
2.0E+06

Figure3


1.05
1.00
0.95
0.90
0.85
$^{240}Pu_{eff}$ mass ratio
INCC/declared
DCF/declared (d = 30.7ns)
DCF/declared (d = 40.7ns)
DCF/declared (d = 50.7ns)
0
1000
2000
3000
4000
5000
Declared total Pu mass [g]