The Impact of Gate Width Setting and Gate Utilization Factors on Plutonium Assay in Passive Correlated Neutron Counting

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ABSTRACT

In the field of nuclear safeguards, passive neutron multiplicity counting (PNMC) is a method typically employed in non-destructive assay (NDA) of special nuclear material (SNM) for nonproliferation, verification and accountability purposes. PNMC is generally performed using a well-type thermal neutron counter and relies on the detection of correlated pairs or higher order multiplets of neutrons emitted by an assayed item. To assay SNM, a set of parameters for a given well-counter is required to link the measured multiplicity rates to the assayed item properties. Detection efficiency, die-away time, gate utilization factors (tightly connected to die-away time) as well as optimum gate width setting are among the key parameters. These parameters along with the underlying model assumptions directly affect the accuracy of the SNM assay. In this paper we examine the role of gate utilization factors and the single exponential die-away time assumption and their impact on the measurements for a range of plutonium materials. In addition, we examine the importance of item-optimized coincidence gate width setting as opposed to using a universal gate width value. Finally, the traditional PNMC based on multiplicity shift register electronics is extended to Feynman-type analysis and application of this approach to Pu mass assay is demonstrated.

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1. INTRODUCTION

In Passive Neutron Multiplicity Counting (PNMC) applications, high detection efficiency is typically required in order to detect correlated pairs or higher order multiplets of neutrons emitted by an assayed item [1,2]. The correlated neutron signal is a direct signature of nuclear fissions occurring in the item and can be used to quantify the amount of fissioning material. To achieve high detection efficiency, thermal neutron counters typically utilize a well-type geometry with the central cavity surrounded by one or more rings of thermal neutron detectors (typically ^3He filled proportional counters) embedded in high density polyethylene (HDPE). The cavity is enclosed by top and bottom end-plugs made of suitable material (typically HDPE or an assembly of aluminum with HDPE, or graphite) to re-scatter the item-emitted neutrons back into the counter's active volume. Various designs of well-counters are available with performance characteristics optimized for measurements of different items of interest. A detailed overview of existing coincidence and multiplicity well-counters can be found in [1,2]. To characterize the assayed special nuclear material (SNM), the measured multiplicity rates (singles, S , doubles, D , and triples, T) are linked to the item properties (mass, multiplication and relative random neutron contribution from (α,n) reactions, denoted as α) through several assumptions encompassed within point-model [2]. Several well-counter specific parameters are required in order to extract the item characteristics from the measured multiplicity rates. Detection efficiency is an essential parameter and is typically extracted via measurements of well-characterized (i.e. certified by

47 national certification body such as National Institute of Standards and Technology, NIST or
48 National Physical Laboratory, NPL etc.) reference ^{252}Cf sources with uncertainties of typically
49 $\sim 1.5\%$. To account for differences in ^{252}Cf and Pu neutron emission spectra subsequent re-
50 calibration is typically performed using a well-characterized Pu standard or through application
51 of a Monte Carlo calculated correction factor. The neutron die-away time represents another key
52 parameter characteristic of a given counter design. The neutron die-away time corresponds to the
53 mean lifetime of neutron population in the counter and represents the average time of slowing
54 down and diffusion of neutrons within the counter material before detection or loss via parasitic
55 neutron capture (outside the active volume) or via escape from the detection system. In practice
56 the slowing down times are short compared to the diffusion timescales and do not significantly
57 affect the measured signals, since the detected events correspond to neutrons after the initial
58 slowing down process. Die-away time is a fundamental characteristic of counter design and is
59 closely related to the amount of moderating material (typically HDPE) and distribution and
60 intrinsic efficiency of individual thermal neutron detectors within the system. Die-away time
61 determines the optimum operating parameters of a counter as it defines the length of time
62 window (coincidence gate) needed to detect a sufficient fraction of correlated fission events.
63 Short die-away time is desirable in practical applications in order to reduce chance coincidence
64 events that are detected along with the genuine correlated events and thus deteriorate the
65 measurement precision. Although die-away time is a key parameter that defines other essential
66 counter operating characteristics, in practical application it is needed only to determine the gate
67 utilization factors (GUF) that essentially represent correction factors for the use of finite
68 coincidence gate in the counting of correlated events. The decay of the neutron population within
69 the counter is typically approximated by a single exponential with the die-away time as the $1/e$

decay parameter. Due to the finite coincidence gate width used to perform correlated counting and roughly exponential decay of the neutron population not all correlated neutrons are detected and counted within the selected finite gate width [2,3]. GUF values correct for the ‘missing events’ and thus effectively extrapolate the detected finite-gate multiplicity rates (S , D , T) to the near-ideal, ‘infinite-gate’ quantities required in point model expressions. It is therefore the operating coincidence gate width and corresponding GUF values that are essential for counter performance and quality of Pu assay.

The nuclear safeguards community, including the International Atomic Energy Agency (IAEA), traditionally employs multiplicity shift register (MSR) electronics to perform time correlation analysis (TCA) on the neutron detection times (pulse train). In MSR, set of two coincidence gates (of the same width) is used to extract multiplicity rates (D , T): triggered-gate and random-gate. The triggered-gate is opened by each detected neutron and all correlated multipliers within this gate, following a short predelay, T_p , are counted to create a triggered-gate multiplicity distribution. The predelay is a triggered-gate specific parameter that is needed to remove a portion of the pulse train affected by instrumental artifacts (deadtime, baseline shift, etc.) following each trigger pulse and is typically selected to allow sufficient time for these effects to dissipate before the coincidence gate is inspected. The triggered-gate multiplicity distribution contains both genuine as well as randomly correlated multipliers. Contrary to the triggered-gate, the random-gate is opened randomly with respect to the neutron detection times. No predelay is required for this gating scheme. Two approaches can be utilized to generate the random-gate multiplicity distribution: use of sufficiently long delay following the trigger pulse to assure that any correlations with the trigger have dissipated or use of sequence of gates opened at a fixed rate. The latter approach is also known as fast accidentals (FAS) and has been shown to improve

the precision of MSR multiplicity rates by up to a factor of $\sqrt{2}$ [4]. The random-gate multiplicity distribution then contains multiplets that are randomly correlated with respect to the trigger pulse and is used to remove this random contribution from the triggered-gate multiplicity distribution. In the MSR, the correlated events from triggered-gate and random-gate are thus combined to extract the genuine correlated multiplicity rates (D , T). This type of analysis (also known as MIXED) employs a finite coincidence gate and the measured D and T rates represent so-called finite-gate multiplicity rates. Triggered-gate GUF values are then required to extrapolate these measured quantities to the corresponding ‘infinite-gate’ multiplicity rates.

Alternatively, in the field of sub-criticality experiments, Feynman variance-to-mean analysis is more commonly employed. This approach utilizes solely information contained within the random-gate. Similarly as in the case of MSR approach, two methods can be utilized to generate the random-gate multiplicity distribution: use of sufficiently long delay following the trigger pulse to assure that any correlations with the trigger have dissipated or use of sequence of gates opened at a fixed rate. The latter approach is typically employed to perform Feynman variance-to-mean analysis, however, the information contained in the random-gate multiplicity distribution generated using the long delay may also be used to extract correlated events as shown in [5]. A link between the Feynman variance-to-mean analysis and PNMC was recently established and discussed in detail in [5-7] and relies on the realization that multiplicity distribution constructed from series of random-gates can itself be utilized to extract the genuine correlated multiplicity rates. This approach was termed Randomly Triggered Interrogation (RTI). Equivalence between the MSR and RTI approaches for a range of ^{252}Cf sources was experimentally demonstrated [5], suggesting that both approaches can be used interchangeably to extract multiplicity rates (i.e. S , D , T) within PNMC and their potential respective benefits can be

further evaluated. Similarly as in the MSR analysis, finite gate width is used to extract the measured correlated multiplicity rates using RTI approach. Corresponding random-gate GUF values are thus required to extrapolate the measured quantities to the near-ideal ‘infinite-gate’ multiplicity rates.

Theoretical expressions for triggered-gate as well as random-gate GUF values are available [7,8] and generally rely on a single die-away time assumption, derived for an idealized detection system, where neutron population lifetime can be described by a single exponential. The corresponding expressions are written as follows:

$$f_n = [e^{-T_p/\tau}(1 - e^{-T_g/\tau})]^{n-1} = f_2^{n-1} \quad (1)$$

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

In these expressions $n \geq 2$, T_p represents predelay, T_g is the selected gate width and τ is the system die-away time.

Realistic systems, however, often utilize several rings of neutron detectors embedded in varying amounts of HDPE and although single die-away time assumption is typically valid to a good degree, multiple die-away time components always exist due to the finite and heterogeneous nature of the system. In safeguards applications it is thus common practice to determine the GUF values for each coincidence/multiplicity counter experimentally, instead of using Eq. (1,2), and to treat them as fixed counter-specific constants. In the standard MSR analysis, expressions for triggered-gate GUF values based on the measured multiplicity rates can be derived assuming a non-multiplying system with no random (α, n) neutron contribution (i.e. a ^{252}Cf neutron source) and correspond to [2]:

$$f_d = \frac{2v_{S1}D_{MSR}}{\varepsilon v_{S2}S_{MSR}} \quad (3)$$

$$f_t = \frac{3f_d v_{S2}T_{MSR}}{\varepsilon v_{S3}D_{MSR}} \quad (4)$$

where S_{MSR} , D_{MSR} , T_{MSR} correspond to deadtime-corrected measured MSR singles, doubles and triples rates, respectively; and v_{si} ($i=1-3$) represent the first–third factorial moments of the spontaneous fission neutron distribution. Thus it can be seen that f_d and f_t can be extracted directly from the deadtime-corrected measured ^{252}Cf multiplicity rates. It should be pointed out that since the GUF values essentially reflect the die-away time of the system, which is independent of the type of item measured, the use of ^{252}Cf source provides adequate representation of the die-away behavior that can be utilized also for Pu-bearing items. Eq. (3,4) were derived directly from point model equations for multiplicity rates and thus do not rely on the single die-away time assumption. Similarly as in MSR analysis, also in the case of RTI analysis the random-gate GUF values can be extracted experimentally using similar expressions [7]:

$$w_2 = \frac{2v_{S1}D_{RTI}}{\varepsilon v_{S2}S_{RTI}} \quad (5)$$

$$w_3 = \frac{3w_2 v_{S2}T_{RTI}}{\varepsilon v_{S3}D_{RTI}} \quad (6)$$

where S_{RTI} , D_{RTI} , T_{RTI} correspond to deadtime-corrected measured RTI singles, doubles and triples rates, respectively. Therefore, similar set of equations can be utilized to extract the random-gate doubles and triples GUF values (w_2 and w_3) experimentally as will be demonstrated in this work for a range of realistic plutonium items.

157 Additionally, in the traditional coincidence/multiplicity counting applications, it is a common
158 practice to use a universal coincidence gate width setting for the entire range of materials
159 assayed in a given counter. The need to use a universal coincidence gate width setting was
160 originally motivated by the multiplicity shift register electronics utilized to collect and pre-
161 analyze the incoming pulse train to form multiplicity distributions. Having only a single
162 coincidence gate width option was typical for MSR operation. When list mode data acquisition is
163 used, the neutron detection time is recorded for the entire pulse train and available for future
164 analysis. Consequently, the recorded pulse train can be re-analyzed using an arbitrary number of
165 coincidence gate width settings allowing detailed analysis and assessment of impact of these
166 settings on the assay results. The variation of optimum coincidence gate width was studied for a
167 range of ^{252}Cf sources in [9]. In this paper we will expand this work to a range of plutonium
168 items representative of Pu items of operational interest and evaluate its impacts on accuracy of
169 Pu assay. Using the optimum gate width setting established for each item, the experimental
170 (single die-away time assumption-free) extraction of GUF values will be employed for MSR as
171 well as RTI approaches. The ^{240}Pu effective mass of a range of assayed items extracted using
172 experimentally as well as theoretically obtained GUF values will be compared to the known
173 declaration, which will provide a useful metric to evaluate use of experimental versus theoretical
174 GUF values along with the importance of item-optimized gate width choice.

176 2. Pu ASSAY AND RELATED OPERATING PARAMETERS

177 To extract physical characteristics of an assayed item, PNMC utilizes the point model
178 assumptions and corresponding equations that relate the measured (finite-gate), deadtime-

179 corrected multiplicity rates (S_i , D_i , T_i , where i =MSR, RTI, respectively) to the item parameters.

180 These equations may be written as follows [2]:

$$181 \quad S_i = F_S \varepsilon v_{S1} (1 + \alpha) M_L \quad (7)$$

$$182 \quad D_i = \frac{F_S \varepsilon^2 g_2 M_L^2}{2} \left[v_{S2} + \left(\frac{M_L - 1}{\bar{v}_{I1} - 1} \right) v_{S1} (1 + \alpha) v_{I2} \right] \quad (8)$$

$$183 \quad T_i = \frac{F_S \varepsilon^3 g_3 M_L^3}{6} \left[v_{S3} + \left(\frac{M_L - 1}{\bar{v}_{I1} - 1} \right) [3v_{S2} v_{I2} + v_{S1} (1 + \alpha) v_{I3}] + 3 \left(\frac{M_L - 1}{\bar{v}_{I1} - 1} \right)^2 v_{S1} (1 + \alpha) v_{I2}^2 \right] \quad (9)$$

184 where the variables used are:

185 F_S = spontaneous fission rate

186 ε = neutron detection efficiency

187 M_L = neutron leakage multiplication

188 $\alpha = (\alpha, n)$ to spontaneous fission neutron ratio

189 g_2 = doubles GUF (where $g_2 = f_d$, w_2 for MSR and RTI analysis, respectively)

190 g_3 = triples GUF (where $g_3 = f_t$, w_3 for MSR and RTI analysis, respectively)

191 v_{si} ($i=1-3$) = first – third factorial moments of the spontaneous fission neutron distribution

192 v_{ii} ($i=1-3$) = first – third factorial moments of the induced fission neutron distribution

193

194 Based on these equations, the item parameters (F_S , M_L , α) can be extracted. The spontaneous
 195 fission rate can be related to the so-called effective ^{240}Pu mass ($^{240}\text{Pu}_{\text{eff}}$), which is defined as
 196 mass of ^{240}Pu that would produce the same double coincidence response as that obtained from all
 197 the even isotopes contained in the assayed item. This is a standard definition adopted in the
 198 nuclear safeguards community for assays of Pu-bearing items [1]:

$$199 \quad ^{240}\text{Pu}_{\text{eff}} = 2.52^{238}\text{Pu} + ^{240}\text{Pu} + 1.68^{242}\text{Pu} \quad (10)$$

From Eq. (7-9) it can be seen that several counter specific parameters are required for extrapolation of measured deadtime-corrected multiplicity rates to near-ideal (infinite-gate and efficiency corrected) quantities, namely detection efficiency, ϵ , along with GUF values (f_d, f_t and w_2, w_3 for MSR and RTI analysis, respectively). As stated earlier, the efficiency is among the key counter specific parameters and is typically extracted via a series of careful measurements of well-characterized reference ^{252}Cf sources and traceable Pu standards of well-known mass, isotopic composition, chemical form and purity. Therefore, in this study the efficiency will be treated as a well-characterized constant with the appropriate uncertainty and the main focus will be on the influence of selection of GUF values and optimum coincidence gate width.

Selecting the operating coincidence gate width, T_g , which is tightly connected to the detector die-away time, is essential for neutron coincidence/multiplicity counting applications, since it is directly related to the measurement precision. Wider gate widths allow for higher contributions of chance correlations that result in significant deterioration of measurement precision especially at high rates. Thus coincidence gate width values optimized for correlated measurement precision are typically used in PNMC. A theoretical relationship between the optimum coincidence gate width and die-away time can be derived via minimizing the corresponding doubles rate precision and is generally used as an initial guidance for universal operating gate width setting. In standard MSR electronics this expression corresponds to $T_g \sim 1.27\tau$ as shown in [1,10]. Extension of these considerations towards RTI approach was performed in [9], where it was shown that for purely random gates, this relationship is modified to $T_g \sim 2.14\tau$. Wider optimum gate width setting is thus favored by the RTI analysis. Alternatively, the optimum gate width can be determined experimentally using series of coincidence gate width settings and extracting multiplicity precision for each gate width value. Graphical representation of relative

standard deviation (RSD) versus coincidence gate width then exhibits a shallow minimum corresponding to the optimum gate width setting. This approach was applied in [9] to a series of ^{252}Cf sources to provide a comparison with the above theoretical estimates. It was shown that the optimum coincidence gate width varies strongly with the ^{252}Cf neutron emission rate with longer gates being favored for weaker sources, although it is common practice to use a universal coincidence gate width value for a given detector type. In this paper, this approach will be extended towards plutonium items with range of masses and α -values to demonstrate the effect of item-optimized coincidence gate width setting and evaluate its importance for practical applications.

3. OPTIMUM GATE WIDTH FOR Pu ITEMS

For the purpose of this evaluation, measurements were performed in Los Alamos National Laboratory (LANL) Epithermal Neutron Multiplicity Counter (ENMC) using a range of available Pu items. The ENMC represents a flagship neutron multiplicity counter with optimized efficiency and die-away time characteristics for high accuracy multiplicity measurements for nuclear safeguards studies. ENMC is composed of 121 ^3He -gas filled proportional counter tubes of 1" (2.54 cm) diameter surrounding the central cavity in four rings. A photograph of the instrument along with schematic internal layout is shown in Figure 1. The individual tubes are filled at 10 atm pressure. The ENMC detection efficiency corresponds to 64.2% for plutonium energy spectrum with die-away time of 21.8 μs . Additional details of ENMC design and operation can be found in [10].

245 {space for Figure 1}

246

247 An overview of the measured Pu items is provided in Table 1. Measured items covered masses
248 from <1 g to ~100 g with a range of α -values (i.e. random neutron contribution from (α ,n)
249 reactions). During measurements, each item was positioned in the center of the counter cavity.
250 Measurement times corresponded to 1-2 hours to assure < 1% precision in the triples rates for the
251 range of settings utilized in this work.

252

253 {space for Table 1}

254

255 Measurements were performed using list mode data acquisition that allows recording and storing
256 of individual neutron detection times in a binary pulse train. The pulse train was subsequently re-
257 analyzed using a series of gate width values between 16 – 256 μ s in a factor of 2 increments.
258 Doubles and triples relative standard deviation (RSD) was extracted for each gate width setting
259 using MSR as well as RTI pulse train analysis approaches. To generate a series of random
260 coincidence gates in MSR as well as RTI analysis a fixed clock frequency of 1MHz was used.
261 The overview of RSD in % versus gate width settings is provided in Figures 2 and 3.

262

263 {space for Figure 2 and 3}

264

Figures 2 and 3 illustrate a typical behavior of RSD with increasing gate width. A shallow minimum for both the MSR and RTI approaches is clearly visible, with precision steadily deteriorating for further increases of gate width. It can be seen that for all Pu items the minimum is shallower for RTI compared to MSR approach. Comparison of the doubles and triples RSD plots demonstrates that an equivalent optimum gate width setting is favored by both multiplicity orders, thus confirming similar findings for ^{252}Cf in [5]. The fact that the minimum exhibits a rather shallow trend along with observation that D and T RSDs favor similar optimum gate width values are key features supporting universal optimum gate width selection practiced in current safeguards. A summary of the optimum gate width values extracted from Figures 2 and 3 for a subset of items representing the full range of measured count rates is provided in Table 2.

{space for Table 2}

In Table 2 it can be seen that the item-optimized gate width value decreases with increasing measured singles rate that includes the random neutron contribution from (α,n) reactions rather than solely with increasing ^{240}Pu effective mass. Therefore, it can be concluded that the observed optimum gate width dependence is mainly related to the neutron detection rate and confirms earlier findings using a series of ^{252}Cf sources [5]. The standard universal optimum gate width setting for ENMC and MSR analysis corresponds to 24 μs and as Table 2 suggests, for all but the smallest Pu item, is well in the range of extracted values. Measurements of small items imply low count rates, where the random coincidences are relatively unimportant compared to the need to collect correlated counts with sufficient statistics, which explains why a wider gate is

generally favored in this domain. It should be pointed out that PNMC is generally not applied in the low-mass item range due to the high precision in the triples rates required to solve Eq. (7-9) with sufficient accuracy, which would involve excessive measurement times. As suggested by the approximate theoretical expression in [9], RTI approach favors wider optimum gate width setting.

Another important observation can be inferred from Figures 2 and 3 related to the relative precision of the MSR and RTI approaches. It can be seen that for all large mass items (> 3 g in this study) the RTI approach provides better RSD than MSR approach. For small Pu items (< 1 g in this study) the extracted maximum precision is comparable for both analysis methods. These results confirm similar findings for range of ^{252}Cf sources presented in [5,10] and suggest the potential for improved measurement conditions (shorter measurement times) when utilizing RTI approach with FA for a certain subset of items. Further evaluation extending current dataset towards higher mass Pu items will be needed to fully assess the potential benefit of the RTI technique.

4. IMPACT OF GUF ON Pu ASSAY

Using the item-optimized gate width values summarized in Table 2, the finite-gate multiplicity rates (S, D, T) can be extracted for each measured item using MSR as well as RTI analyses. The corresponding triggered-gate and random-gate GUF values (f_d, f_t and w_2, w_3 , respectively) must be applied to extrapolate to the ‘infinite-gate’ multiplicity rates to extract item specific parameters based on Eq. (7-9). Three approaches will be considered here to evaluate the optimum counter operating settings using measured versus declared ^{240}Pu effective mass as a

comparison metric. First, the item-optimized gate width settings summarized in Table 2 will be used and the corresponding triggered-gate and random-gate GUF values will be extracted using Eq. (1,2) based on a single die-away time assumption with a standard ENMC die-away time of 21.8 μ s (item-optimized T_g , theoretical GUF). Second, the item-optimized gate width settings will be used in conjunction with the experimentally extracted GUF values based on Eq. (3-6) (item-optimized T_g , experimental GUF). A well-characterized ^{252}Cf source will be used for this purpose. Third, the two bespoke approaches based on the item-optimized coincidence gate width setting will be compared with results obtained using standard ENMC operating parameters (universal coincidence gate width) to evaluate the importance of the item-optimized gate width setting.

Assuming the item-optimized coincidence gate width values from Table 2, the deadtime-corrected finite-gate S , D , T rates can be extracted using MSR and RTI approaches. The deadtime correction used in this analysis corresponds to an empirical form extracted using range of ^{252}Cf sources with detection rates that overlap with the range of currently investigated Pu items [5]. The deadtime correction adopted here can be expressed as follows:

$$S_i = S_{mi} \cdot e^{\delta_{Si} S_{mi}} \quad (11)$$

$$D_i = D_{mi} \cdot e^{\delta_{Di} S_{mi}} \quad (12)$$

$$T_i = T_{mi} \cdot e^{\delta_{Ti} S_{mi}} \quad (13)$$

Where $i=\text{MSR}$ and RTI , respectively and δ_{ji} , ($j=S, D, T$) corresponds to singles, doubles and triples deadtime parameters, respectively for the MSR and RTI approaches. The deadtime parameter values for both analysis approaches are summarized in Table 3. The advantage of this deadtime correction implementation lies in its consistency for both pulse train analysis

approaches that provides an empirical formulation valid for the range of items investigated in this paper. More elaborate deadtime correction algorithms developed from first principles are currently under evaluation and will be reported in a separate publication. The approach adopted here is adequate for the present purposes, since deadtime corrections are relatively small due to a combination of counter design and the modest counting rates involved.

{space for Table 3}

Using the deadtime-corrected S , D , T rates measured in the MSR and RTI approaches for each coincidence gate width setting, the experimental triggered-gate (f_d , f_i) and random-gate (w_2 , w_3) GUF values can be extracted using Eq. (3-6). Similarly, the theoretical triggered-gate and random-gate GUF values can be calculated based on Eq. (1,2) for each gate width, T_g , assuming the die-away time of 21.8 μ s. Tables 4 and 5 provide a summary of the experimental and theoretical GUF values for the MSR and RTI approaches for the range of coincidence gate width settings explored in this work. The corresponding uncertainties were extracted from expressions (3-6) and (1,2) for the experimental and theoretical GUF values, respectively. For the experimental values, the uncertainties of the measured S_i , D_i , T_i rates (i = MSR, RTI, respectively) were utilized that typically correspond to <1%. The relative standard deviations on ν_{S1} , ν_{S2} and ν_{S3} are estimated to be approximately 0.27%, 0.062% and 0.23%, respectively based on the data listed in [11,12]. Finally, the uncertainty of the detection efficiency (ϵ) is affected by the accuracy of the absolute yield of the certified ^{252}Cf source that, depending on the certifying laboratory, typically corresponds to ~1.5%. For the theoretical GUF values an uncertainty of die-

away time of 1% was used [13] that includes systematic effects extracted based on practical experience with operating range of high-performance counters.

{space for Tables 4 and 5}

Assuming the GUF values from Tables 4 and 5, the deadtime-corrected S , D , T rates measured for a range of Pu items can be extrapolated to the near-ideal ‘infinite-gate’ multiplicity rates. Using point model equations (Eq. (7-9)), the corresponding ^{240}Pu effective mass (as well as multiplication and α -values) can be calculated for each measured Pu item using its item-optimized gate width setting and assuming experimental as well as theoretical GUF values. In addition, a standard ENMC coincidence gate width setting of $T_g=24\text{ }\mu\text{s}$ was used for all the measured Pu items for comparison with the item-optimized gate width settings. The assayed ^{240}Pu effective mass for the three T_g /GUF combinations is summarized in Tables 6 and 7 for MSR and RTI analysis, respectively. The corresponding assay-over-declaration percent difference is shown in Figures 4 and 5 for the two analysis approaches.

{space for Table 6}

{space for Figure 4}

Table 6 and Figure 4 provide a useful comparison of expected precision and accuracy for traditional MSR assay using different counter operating settings. From Table 6 it can be seen that all three investigated T_g /GUF combinations provide very good quality assay results. Comparing in detail the assay results from the two approaches using item-optimized T_g combined with experimental and theoretical GUF values it can be seen that the use of theoretical GUF values results in overall poorer performance with average assay-over-declaration ratio of $0.981 \pm 0.010\%$ and $0.985 \pm 0.009\%$ for pure and impure Pu items, respectively; compared to $0.994 \pm 0.009\%$ and $0.998 \pm 0.008\%$ when using experimental GUF. This comparison thus serves as an illustration of the impact of use of experimental GUF values extracted directly from the measured multiplicity rates as opposed to relying on a single die-away time assumption. The use of experimental GUF values essentially provides a self-consistent way to correct for the deficiencies of the single exponential die-away time assumption to realistic features of the given counter. Results for a different counter might differ slightly depending on the counter design that defines the extent to which the single die-away approximation is valid.

Comparing the use of item-optimized gate width setting with the traditional approach of assuming a universal gate width value, it can be concluded that both approaches yield about equally good performance with average assay-over-declaration ratio of $0.994 \pm 0.009\%$ and $0.992 \pm 0.010\%$ for item-optimized and universal T_g , respectively for pure PuO_2 items. In the case of impure Pu items, the average assay-over-declaration ratio corresponds to $0.998 \pm 0.008\%$ and $0.998 \pm 0.009\%$ for the item-optimized and universal T_g , respectively. This comparison therefore provides a justification and feasibility of use of a universal coincidence gate width over a span of Pu items with range of masses and impurities similar to items of safeguards interest.

{space for Table 7}

{space for Figure 5}

Table 7 and Figure 5 illustrate the application of Pu mass assay to RTI-based analysis of the acquired pulse train. The data serve as a demonstration of feasibility of this technique in the context of traditional PNMC and Pu assay. Contrary to MSR analysis, it can be seen that all three T_g /GUF combinations result in similar assay results and there is no apparent direct benefit to using experimental instead of theoretical GUF values for this item-set and counter combination. The assay results for universal coincidence gate width setting agree very well with the results obtained using item-optimized gate width settings. The overall assay results for the item-optimized gate width with experimental GUF values correspond to assay-over-declaration ratios of $0.987 \pm 0.017\%$ and $1.002 \pm 0.007\%$ for pure and impure items, respectively. Similarly for the item-optimized gate width with theoretical GUF, the assay results in assay-over-declaration ratios of $0.988 \pm 0.012\%$ and $0.998 \pm 0.006\%$ for the pure and impure items, respectively. Finally, for the universal coincidence gate width setting, the assay results in assay-over-declaration ratio of $0.987 \pm 0.017\%$ and $1.004 \pm 0.007\%$ pure and impure items, respectively.

Both, MSR as well as RTI analysis exhibit slight negative bias (on average -1.06% and -1.25% for MSR and RTI analysis, respectively), which may in part be attributed to imperfections in the empirical deadtime correction selected within this work as well as to uncertainties in the utilized GUF values. Further work on more realistic deadtime correction is currently underway by our group, which should provide a better description of pulse losses in a correlated pulse stream. The

deadtime correction applied in this work nevertheless represents an approach consistent with common practice utilized in current nuclear safeguards [1,2]. In addition, it should be pointed out that the point model equations (Eq. (7-9)) were applied in an absolute sense to extract the assayed item parameters ($^{240}\text{Pu}_{\text{eff}}$ mass, multiplication and α) and thus require an estimate of the absolute detection efficiency. As discussed earlier, this quantity is determined through measurements of traceable (i.e. attested by national certification body) ^{252}Cf sources with absolute yields that exhibit uncertainties typically in the range of $\sim 1.5\%$ [14]. Thus the detection efficiency will suffer similar uncertainty that will affect the extracted GUF values, potentially contributing to the observed negative bias. It is possible to adjust the model parameters within reasonable limits to achieve better overall agreement with declarations, an approach typically pursued in common practice to allow for point-model violations and include potential item dependencies. Such an approach has not been adopted here to allow for a direct comparison of the two analysis approaches and highlight possible differences among the different parameters used.

Another important aspect of this assay comparison is the precision of the ^{240}Pu effective mass. The results demonstrated good equivalence of using universal T_g with the results extracted for item-optimized coincidence gate width settings. A closer look at the values presented in Tables 6 and 7 (columns 2 and 4) reveals that despite the very good assay results, the use of universal gate width exhibits poorer precision than the corresponding results extracted assuming item-optimized T_g . This is an expected trend as suggested by plots in Figures 2 and 3 that highlight the precision deterioration with off-optimum coincidence gate width settings. For the items evaluated in this paper, the effect is especially noticeable for low-mass items (Pu1-Pu3) as these favor wider gate width settings than is the standard universal ENMC setting. The precision

deterioration for these items is however, fairly small and can be neglected in most practical applications targeting broader ranges of item masses. For very small items the effect can be anticipated to have higher impact and wider gate width is typically used in specific applications, such as waste assay [15].

Comparing precision values between Table 6 and 7 it can be seen that assay results obtained with RTI analysis exhibit better precision for the entire range of items. This is a consequence of the improved precision of D and T rates observed in Figures 2 and 3. Also in this case the improvement is rather moderate and likely not bringing significant assay time improvements for most practical applications, where measurement time is not critical. Further evaluation will be needed to assess potential benefits of RTI analysis technique for applications beyond the items assayed in this work (such as very low-mass or high-mass Pu items).

5. CONCLUSIONS

In nuclear safeguards, bulk SNM assay is often performed using multiplicity shift register electronics and a set of predetermined detector-specific parameters is required to fully characterize the assayed item and extract its physical properties (mass, multiplication and contribution of random (α, n) neutrons). For the latter, point-model assumptions are used that relate the measured deadtime-corrected multiplicity rates (S, D, T) to these physical properties. Detection efficiency, pre-delay, operating gate width setting and corresponding GUF values are the key parameters required to extrapolate from the measured, finite-gate multiplicity rates to corresponding ‘infinite-gate’ (gate-independent) and efficiency corrected rates required in the point model equations. In current safeguards practice, a universal coincidence gate width is

typically determined via measurement of a single known ^{252}Cf (or Pu when available) source and the corresponding measured rates are utilized to extract the GUF values.

It was shown previously, using a range of ^{252}Cf sources that the optimum gate width setting varies with neutron detection rate [5]. In addition, the applicability of PNMC was recently extended to include an alternative pulse train analysis technique (the RTI approach), derived from an approach typically employed to assess the multiplication of sub-critical systems. This allowed an expansion of the range of PNMC techniques and evaluation of potential benefits of this alternative approach. In this work the focus was on the evaluation of the effects of the choice of gate width setting and corresponding GUF values on the Pu assay precision and accuracy as well as on demonstrating the feasibility of Pu assay using RTI pulse train analysis technique recently demonstrated to provide equivalent multiplicity rates for a range of ^{252}Cf sources [5]. The current work utilized the versatility of list mode data acquisition, which allows re-analysis of recorded pulse train to conveniently access a range of gate widths as well as to employ alternative analysis algorithms. The main objective was to evaluate the necessity for an item-optimized gate width setting as opposed to a universal T_g value extracted for a single representative item and assess the assay accuracy using corresponding experimentally extracted GUF values. Theoretical GUF values were utilized as well for comparison and to demonstrate the effects of a single die-away time assumption on the assay results. These two approaches were compared with assay based on a standard ENMC operating gate width. Finally, the analysis was extended to the RTI approach in order to illustrate the optimum gate width selection and experimental GUF extraction for this analysis technique and to demonstrate its feasibility.

The results of this work provide an overview for the selected system (ENMC) performance for a range of detector-specific parameters on a set of well-characterized Pu items with mass of $< 1\text{g}$

to ~100 g and covering a range of impurities. The variation of optimum gate width setting was evaluated and confirms previous findings that optimum gate width setting is closely related to the neutron detection rate irrespective of other item characteristics – i.e. small Pu items with high impurity will favor narrower gate width settings similar to high-rate items, due to the high contribution of neutrons from (α ,n) reactions. This observation is valid for both analysis approaches (MSR and RTI). Optimum gate width evaluation also demonstrates that the precision of the RTI technique is generally less sensitive to variations of gate width setting than the MSR technique, due to its shallower RSD dependence on gate width for D as well as T rates. In addition, Pu assay results obtained in this work demonstrate improved measurement precision for RTI compared to MSR approaches for the entire range of investigated Pu items measured in ENMC. This observation is a direct consequence of the precision of underlying multiplicity rates and confirms earlier findings with a range of ^{252}Cf sources. The precision improvement is fairly moderate and is unlikely to be of practical importance for the type of items evaluated in this work; however, potential benefits should be evaluated for other types of items such as waste, scrap or high-mass multiplying bulk Pu items as well as for other counter designs.

Absolute Pu assay was performed using both multiplicity analysis techniques (MSR and RTI). Focusing on the traditional MSR analysis technique, the results suggest very good assay results for the entire range of items for both item-optimized coincidence gate width settings as well as the universal gate width value, typical for the ENMC counter. The assay-over-declaration ratio for pure (impure) Pu items corresponds to $0.994\pm0.009\%$ ($0.998\pm0.008\%$) and $0.992\pm0.010\%$ ($0.998\pm0.009\%$) for the two gate width approaches, respectively. A slight difference between the two gate choices can be observed in the measurement precision, with the item-optimized gate width approach generally resulting in lower uncertainty than using the universal gate width. The

effect is typically the most significant for small items due to the optimum coincidence gate width of these items being further off the universal value. For the items studied, the effect is however too small to be of practical importance in safeguards applications. These results provide useful evidence for the universality of the optimum gate width setting and support the choice of a single optimum gate width value that provides considerable simplification of the data acquisition/analysis for a range of assayed items. Comparing with previous evaluations of a range of ^{252}Cf sources [5], it can be expected that the optimum gate width setting does not significantly change with further increases in neutron detection rate. On the contrary, very low mass items might require using a wider coincidence gate width setting due to precision differences and improved measurement times, as is common practice in waste assay. Nevertheless, quantitative PNMC analysis is not usually applied at very low count rate levels as it requires high precision in the triples rates and thus would require excessive assay times. The comparison with theoretical GUF values for the MSR analysis demonstrates the benefit of using experimental (single die-away assumption free) GUF values in order to obtain more accurate Pu assay results. The assay-over-declaration ratio assuming theoretical GUF values corresponds to $0.981 \pm 0.010\%$ and $0.985 \pm 0.009\%$ for pure and impure Pu items, respectively.

Results of RTI analysis also exhibit very good agreement between the analysis based on optimum gate width settings and analysis based on universal gate width value. No significant advantage in using experimental over theoretical GUF values is observed in this case for the counter and item set combination studied in this work. The assay-over-declaration ratio in this case corresponds to $0.987 \pm 0.017\%$ ($1.002 \pm 0.007\%$) and $0.987 \pm 0.017\%$ ($1.004 \pm 0.007\%$) for pure (impure) Pu items the two gate width approaches, respectively. These results should provide a general feasibility demonstration of the RTI approach to Pu assay along with direct

demonstrations of experimental determination of optimum gate width and corresponding GUF values that can be extracted using similar expressions as in the case of the MSR approach. Current work also paves the way to properly select analysis parameters for the RTI technique in order to evaluate and demonstrate the feasibility of this new algorithm.

6. ACKNOWLEDGMENTS

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609 Figure 4: Assay/declaration for a range of Pu items measured in ENMC and analyzed using MSR
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613 approach with three different T_g/GUF combinations.

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Table 1: Summary of PuO₂ items measured in the ENMC.

Source ID	Declared ²⁴⁰ Pu _{eff} [g]	Material type	Impurity	α -value (measured)
Pu1	0.56	PuO ₂	Oxide	1.7
Pu2	0.56	PuO ₂	Al	6.8
Pu3	0.70	PuO ₂	-	0.2
Pu4	3.61	PuO ₂	Diatomite	4.2
Pu5	29.26	PuO ₂	-	0.5
Pu6	54.31	PuO ₂	-	0.5
Pu7	92.08	PuO ₂	-	0.5
Pu8	104.46	PuO ₂	-	0.5

Table 2: Overview of optimum gate width settings extracted for MSR and RTI pulse train analyses for a subset of Pu items spanning full range of measured count rates.

Item ID	Declared $^{240}\text{Pu}_{\text{eff}}$ [g]	Measured S [cps]	α -value (measured)	MSR optimum GW [us]	RTI optimum GW [us]
Pu3	0.72	539	0.2	64	128
Pu2	0.56	2817	6.8	32	64
Pu4	3.61	12389	4.2	32	32
Pu5	29.26	30320	0.5	16	32
Pu8	104.46	112638	0.5	16	32

Table 3: Deadtime coefficients for S , D , T rates extracted for the MSR and RTI analysis techniques.

Deadtime coefficient [μs]	MSR	RTI
δ_S	0.036 ± 0.003	0.036 ± 0.003
δ_D	0.144 ± 0.013	0.212 ± 0.011
δ_T	0.618 ± 0.080	0.849 ± 0.079

Table 4: Summary of experimental triggered-gate (f_d , f_t) and random-gate (w_2 , w_3) GUF values extracted from Eq. (3-6) for a range of gate width values utilized in this work.

Gate width [μ s]	Experimental GUF							
	f_d	σ [%]	f_t	σ [%]	w_2	σ [%]	w_3	σ [%]
16	0.496	1.5	0.253	2.2	-	-	-	-
24	0.622	1.1	0.395	1.5	0.409	1.1	0.209	1.5
32	0.710	1.5	0.511	2.2	0.487	1.5	0.290	2.2
64	0.867	1.5	0.754	2.2	0.678	1.5	0.525	2.2
128	-	-	-	-	0.824	1.5	0.735	2.2

Table 5: Summary of theoretical triggered-gate (f_d , f_t) and random-gate (w_2 , w_3) GUF values extracted from Eq. (1,2) for a range of gate width values utilized in this work.

Gate width [μ s]	Theoretical GUF							
	f_d	σ [%]	f_t	σ [%]	w_2	σ [%]	w_3	σ [%]
16	0.485	0.6	0.236	1.2	-	-	-	-
24	0.623	0.5	0.388	1.0	0.394	0.7	0.191	1.3
32	0.718	0.4	0.516	0.7	0.476	0.6	0.274	1.2
64	0.884	0.1	0.781	0.2	0.677	0.4	0.525	0.7
128	-	-	-	-	0.830	0.2	0.745	0.3

Table 6: Overview of ^{240}Pu effective mass values measured in ENMC using MSR analysis and three different T_g/GUF combinations along with the corresponding declared values in the last column. The quoted percent errors represent statistical uncertainties.

Source ID	$^{240}\text{Pu}_{\text{eff}}$ [g] (item-optimized T_g , exp. GUF)	$^{240}\text{Pu}_{\text{eff}}$ [g] (item-optimized T_g , theor. GUF)	$^{240}\text{Pu}_{\text{eff}}$ [g] (universal T_g)	Declared $^{240}\text{Pu}_{\text{eff}}$ [g]
Pu1	0.56 (0.21%)	0.55 (0.21%)	0.56 (0.23%)	0.56
Pu2	0.56 (0.44%)	0.55 (0.45%)	0.56 (0.48%)	0.56
Pu3	0.71 (0.13%)	0.70 (0.13%)	0.71 (0.15%)	0.70
Pu4	3.63 (0.27%)	3.59 (0.28%)	3.63 (0.28%)	3.60
Pu5	28.82 (0.12%)	28.53 (0.13%)	28.77 (0.12%)	29.26
Pu6	54.05 (0.13%)	53.41 (0.13%)	53.84 (0.13%)	54.31
Pu7	91.19 (0.15%)	89.70 (0.15%)	90.73 (0.15%)	92.08
Pu8	103.55 (0.16%)	101.82 (0.16%)	103.24 (0.16%)	104.46

699 Table 7: Overview of ^{240}Pu effective mass values measured in ENMC using RTI analysis and
700 three different T_g/GUF combinations along with the corresponding declared values in the last
701 column.

Source ID	$^{240}\text{Pu}_{\text{eff}}$ [g] (item-optimized T_g , exp. GUF)	$^{240}\text{Pu}_{\text{eff}}$ [g] (item-optimized T_g , theor. GUF)	$^{240}\text{Pu}_{\text{eff}}$ [g] (universal T_g)	Declared $^{240}\text{Pu}_{\text{eff}}$ [g]
Pu1	0.56 (0.19%)	0.56 (0.19%)	0.56 (0.22%)	0.56
Pu2	0.56 (0.40%)	0.56 (0.40%)	0.56 (0.44%)	0.56
Pu3	0.71 (0.13%)	0.71 (0.13%)	0.71 (0.16%)	0.70
Pu4	3.64 (0.22%)	3.61 (0.22%)	3.64 (0.24%)	3.60
Pu5	28.81 (0.10%)	28.89 (0.10%)	28.77 (0.11%)	29.26
Pu6	53.73 (0.11%)	53.81 (0.11%)	53.69 (0.11%)	54.31
Pu7	89.85 (0.12%)	89.97 (0.13%)	89.85 (0.13%)	92.08
Pu8	101.94 (0.14%)	102.04 (0.14%)	101.94 (0.14%)	104.46

702

Figure1

Figure2

Figure3

Figure4

Figure5