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An event-triggered coincidence algorithm for fast-neutron multiplicity assay corrected for cross-talk and photon breakthrough

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ABSTRACT

A model quantifying detector cross-talk and the misidentification of events in fast neutron coincidence distributions is described. This is demonstrated for two experimental arrangements comprising rings of 8 and 15 organic liquid scintillation detectors. Correction terms developed as part of this model are tested with ^{252}Cf and a relationship is developed between the ^{235}U enrichment of U_3O_8 and the order of correlated, fast neutron multiplets induced by an americium–lithium source. The model is also supported by Geant4 simulations. The results suggest that a typical assay, for experimental arrangements that are similar to the examples investigated in this research, will exhibit cross-talk for less than 1% of all detected fast neutrons but, if not accounted for, this can bias the numerical analysis by a margin of 10% and 35% in second- and third-order coincidences (i.e. couplet and triplet counts), respectively. Further, for the case of ^{252}Cf, it is shown that a relatively low proportion of 4% breakthrough by γ rays (that is, photons misidentified as neutrons by the pulse-shape discrimination process) can lead to an erroneous increase of 20% in total neutron counts in the assay of a mixed-field, in this case of ^{252}Cf. These findings will help direct the developments needed to enable organic scintillation detectors with pulse shape discriminators to be applied reliably to nuclear safeguards and non-proliferation verification tasks.

1. Introduction

In nuclear safeguards, fissile material assay is used routinely as one of a number of procedures to ensure that nuclear materials are properly accounted for and not misused. An established technique to this end is neutron multiplicity counting [1,2]. Historically, this method has been deployed using detector systems based on ^{3}He gas for the detection of time-correlated, thermalised neutrons emitted from spontaneous fission (SF) and induced fission (IF). Whilst essentially immune to γ radiation and having high detection efficiencies, ^{3}He-filled proportional counters have the drawback that their sensitivity is optimised for neutrons with energies in the thermal domain. Therefore, the detection apparatus is often arranged to incorporate a stage that is dedicated to the thermalisation of the fast neutrons prior to detection. In addition to reducing the energy of the neutrons, this also increases the source-to-detector time-of-flight due to the time taken for the neutrons to pass through the intermediate stage in which elastic scattering is encouraged to slow the neutrons down to thermal energies.

The implication of this is two-fold in so far as multiplicity and temporal analyses are concerned: (i) the coincidence window needed is substantially wider (that is, of the order of 40–50 μs) [3] compared to the typical time taken for the fission-correlated fast neutron field to die away (typically 20–25 ns), thus influencing acquisition time and statistical uncertainty; and (ii) information on the incident neutron energy is effectively lost in this process, eliminating the prospect of this being exploited for complementary, analytical purposes. Since the rise and fall (the latter being the prompt neutron die-away) of the neutron population in a fission chain can be due to either SF, (α, n) reactions or scattering, and the timing of these different distributions cannot be discerned comprehensively where intermediate thermalisation is necessary, some aspects of the change in the neutron population cannot be determined fully with ^{3}He detectors.

Amongst the earliest reports of fast-neutron multiplicity counting based on the use of organic scintillators in an unmoderated environment is that of Wachter et al. [4]. This highlighted the key potential benefits of fast neutron methods, such as multiplicity sensitivity beyond coincident

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events and significantly-reduced levels of accidentals, over thermal assays. However, it also highlighted the need to correct for: cross-talk, i.e., a chance coincidence where a single neutron can scatter from one scintillation detector to another depositing energy in both, therefore provoking a correlated combination of several events between detector elements (referred to in that work as *multiple-order scattering*); pileup; and the mis-assignment of photons as neutrons (hereafter referred to as *photon breakthrough*). Wachter et al. highlighted the particular significance of these corrections for the case of materials exhibiting high (α, n) yields (relative to fission neutrons) in reducing significant discrepancies in mass assessments that might arise otherwise.

Subsequently, preliminary Monte Carlo studies of system designs taking advantage of liquid scintillator-based, fast-neutron assay systems were reported [5]. These designs adopted thermal neutron coincidence counting auto-correlation techniques that were modified to address the differences in the physics between the two detector systems. Since then, several related counter developments and concepts [6] have been reported using active neutron interrogation [7], and have included further modelling and simulation studies [8–10].

Despite having lower detection efficiencies, organic liquid scintillation detectors can have an advantage in environments associated with items that emit radiation at relatively high rates where chance coincidences can dominate, because they have significantly shorter coincidence gate width requirements, as shown in previous studies [11]. The absence of a thermalisation stage enables coincidence gate-widths of the order of 25 ns to be used, for both neutron and γ-triggered coincidence distributions. This has been accomplished, as described in Section 3 of this work, using the Hybrid Instruments Ltd. MFAx analyser [12], coupled with an off-the-shelf Field-Programmable Gate Array (FPGA), to undertake multiplicity and temporal analysis in real-time, i.e. without post-processing. This approach uses a novel algorithm that works with the size of event clusters in contrast to the traditional approach which is based on analysis of the reduced factorial moments. The shorter coincidence gate-width results in a significant reduction in accidental counts to give reduced levels of uncertainty and increased sensitivity to higher orders of net multiplicity.

However, challenges remain due to two principal disadvantages of organic scintillation materials. Firstly, the relatively high sensitivity of organic scintillators to photons in contrast with that of ^{3}He detectors, coupled with shortfalls in the event discrimination mechanism, can lead to 3–5% of photons (depending on the pulse-shape discrimination (PSD) algorithm being used) being misclassified as neutrons as per the photon breakthrough phenomenon defined earlier. This can have a disproportionate impact on neutron count rates as the ratio between number of neutron and photons emitted from either spontaneous or induced fission is typically of the order of 1:10, for the case of ^{252}Cf, for example. Secondly, cross-talk events arising as a result of a single neutron or photon scattering from one detector to another, thus triggering multiple detectors, can masquerade as correlated multiplets. If a correction for these effects is not made then the assay can be undermined as per, for example, the observations of Wachter et al. [4] referred to earlier. Engineering challenges such as temperature stabilisation, automated setup and so forth for a complex array also exist, but can be overcome by design. Whilst it is possible to configure the PSD algorithm to have very high detector cut-offs in order to operate the detector array in a region where these phenomena are not a hindrance, such an approach is not ideal as it comes at the expense of reduced neutron counts, i.e. reduced neutron efficiency.

Several attempts have been made to address these issues, both experimentally [13,14] and analytically [15–17]. Perhaps most simply, coincident events in adjacent detectors might be discarded (usually by the acquisition firmware or in post-processing) on the basis that cross-talk is most likely to occur between neighbouring detectors; indeed, this is implemented in some commercially-available systems by default. However, this is less than ideal as it might lead to an over-correction given the scenario that bona fide correlated events detected in neighbouring scintillators are also removed. This is especially relevant given the typical, polarised angular correlation between fission neutrons, particularly when tested with isotopes with high values of $\bar{\nu}$, such as ^{252}Cf, where a real correlation in neighbouring detectors might be plausible. Furthermore, for safeguards applications, 2D arrangements of detectors are usually simpler to configure and use than 3D arrangements but detector–detector distances of the former cannot be optimised to minimise cross-talk as easily as in the latter, thus motivating the need for the correction developed here.

The characteristics of neutron cross-talk have been examined before [15] using a ^{252}Cf source, however the results were akin to the cosine distribution consistent with the angular distribution of the source rather than the anticipated isotropic distribution anticipated for cross-talk. The analytical methods suggested by Li et al. [16] and Shin et al. [17] address this problem in a complicated manner using a reduced factorial distribution from a shift register based algorithm.

In this paper, we introduce a correction model based on a relatively simple, event-cluster algorithm using a balance equation to address both detector cross-talk and event mischaracterisation. The coefficients for this model can be derived either experimentally or through simulations. In Section 2, a description of the event-triggered coincidence algorithm and the techniques used for the simulation of the coefficients and validation of the model are presented. Section 3 outlines the experimental techniques with which the approach has been tested, the correction models developed as part of this research are described in Section 4 and the validation of these models is discussed in Section 5. Section 6 summarises the conclusions from the research.

2. Algorithms

2.1. Event-triggered coincidence algorithm

Coincidence counting based on thermal neutrons tends to have relatively large emission-to-detection times in the range 1–100 μs, due to the time necessary for thermalisation. This usually limits a one-shot coincidence algorithm [2] to low count rates. Hence, most assessments based on the detection of thermal neutrons use a coincidence algorithm based on a shift-register [2] to avoid dead time corrections. In the shift-register method, triggers are issued for every incoming event and each starts a new counting window, as illustrated in Fig. 1a. This yields a reduced factorial moment distribution of incoming neutron events; this approach is accepted universally for fissile materials assay in nuclear safeguards. The every-event triggered coincidence distribution is commonly referred to as a multiplicity histogram, and the orders of "multiplicity" are referred to as singles, doubles, triples, quadruples, etc.; these being the 1st, 2nd, 3rd, 4th, etc. net reduced factorial moments on the pulse train.

However, mixed-field analysers used with liquid scintillators have significantly-reduced electronic dead-time, being capable of processing up to 3 million events per second [12]. Moreover, because thermalisation is unnecessary, both the emission-to-detection time and the signal duration are small, i.e., both of the order of several tens of nanoseconds. Hence, the use of an *event-triggered coincidence algorithm* is viable in a fast neutron assay. In this method, when a neutron is first detected such that no prior events have occurred constituting a trigger, the system will issue a trigger. This opens a user-defined prompt gate for the prompt coincidence counter and disables the trigger mechanism. During this window, the algorithm scans for incoming photon and/or neutron events and these are counted. Following the end of the prompt gate, the system is idled for 150 ns and then a delayed gate is opened to assess the accidental coincidence distribution. At the end of each of the two windows, a signal is issued which increments the corresponding foreground and background coincidence distributions and re-activates the trigger mechanism. Hence, in the event sequence illustrated by way of example in Fig. 1b, only the 1st, 5th and 6th triggers are issued, as this is when the trigger architecture is sensitive to incoming events. This prevents the same neutron event to be counted multiple times and, as