Identification of shielded neutron sources with the liquid scintillator BC-501A using a digital pulse shape discrimination method

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Abstract

Fast and unambiguous identification of shielded neutron sources is of paramount importance in nuclear nonproliferation and international safeguards applications. Usually, the identification of neutron spectra relies on unfolding procedures that are unstable and time consuming. In this paper, we present a new application of an existing technique, which can be used for neutron source identification by direct analysis of the pulse height spectrum, i.e., without resorting to energy spectrum unfolding. This technique is based on the acquisition of neutron and \( \gamma \)-ray pulses by using a liquid scintillation detector and a fast waveform digitizer. An optimized digital pulse shape discrimination method based on standard charge integration is used to discriminate neutrons from \( \gamma \)-rays. The neutron pulses are then analyzed to generate a pulse height distribution. The accuracy of the neutron source identification technique was tested on three neutron sources: Cf-252, Am–Be, and Am–Li. Several source–shielding configurations were tested to assess the influence of potential shielding of the source and the sensitivity of the technique. For this investigation, lead and polyethylene shielding blocks were used. The measured pulse height distributions were compared with the distributions simulated with the MCNP-PoliMi code, and very good agreement was obtained. The results show that for both shielded and unshielded configurations the identification of Cf-252, Am–Be, and Am–Li shielded sources is easily achievable by direct analysis of the measured pulse height distributions, without the need of subsequent neutron energy spectrum unfolding.

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1. Introduction

Accurate knowledge of neutron energy spectra is required in many areas, such as nuclear nonproliferation, international safeguards, nuclear material control and accountability, and national security. In particular, for safeguards applications, a fast and robust technique for the identification of typical neutron sources (Cf-252, Am–Be, Pu-240, etc.) is essential. The identification of the neutron sources with organic liquid scintillators using fast digitizers is a very promising technique and is based on the observable differences in the neutron pulse height spectra of the various sources. In fact, liquid scintillators are the main components of various measurement systems.

A common feature of radiation detectors is their sensitivity to more than one type of radiation. This is also true of liquid scintillation detectors, which for several decades have been mainly used as neutron detectors, but which have lately been increasingly used for neutron/\( \gamma \)-ray detection. The main reasons for the popularity of the liquid scintillation detectors are their excellent pulse shape discrimination (PSD) properties and fast timing performance [1–5]. Neutron and \( \gamma \)-ray PSD is a well-known method, which is frequently used for organic liquid scintillation detectors in mixed neutron/\( \gamma \)-ray radiation fields [6,7]. It is also well known that the total light output generated in the scintillation detector can be fairly accurately represented by the sum of the two exponential

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decays referred to as the fast and slow components of the scintillation process (for more accurate representation, a sum of three components is preferable; see, for example, Ref. [8]). The fraction of the amount of light in the slow component is a function of the type of particle inducing the scintillation process. In general, the slow component is larger for heavier particles, i.e., the tails of the average neutron pulses are consistently larger than the tails of the \( \gamma \)-ray pulses [9,10]. This difference in the intensity of slow neutron and \( \gamma \)-ray components serves as a basis for PSD methods used to identify and characterize pulses. Several analog methods for PSD have been developed and studied in the past; examples are charge-comparison and rise-time methods [6,11,12].

Two main trends in the improvement of the PSD properties of liquid scintillators for applications in high count-rate fields have been pursued. The first improvement has been achieved by optimizing the size and shape of detectors [13,14]. The second important improvement allowed for increasing the measurement count rate and maximizing the amount of information obtained from a detector. This step has been achieved by rapid development of waveform digitizers, especially in the recent past, although the first digital neutron–\( \gamma \)-ray PSD measurements were carried out in the early 1980s (see, for example, Ref. [15]).

In this paper, we present a new application of a technique that acquires neutron pulse height spectra using a liquid scintillation detector, a fast digitizer, and an optimized PSD method. This technique is used for fast and accurate identification of shielded neutron sources based on the direct analysis of the resulting pulse height spectra. To our best knowledge, this is the first time this technique has been used for direct identification of shielded neutron sources. This technique is fast, robust, and allows for highly accurate identification of different neutron sources.

The pulses were measured with the liquid scintillator BC-501A and a Tektronix waveform digitizer TDS-5104. The pulses were collected directly from the anode of the detector’s photomultiplier (PMT). A digital PSD method based on ratios of pulse integrals over different time periods was optimized and applied to discriminate neutrons from \( \gamma \)-rays. This method is a digital version of the well-known analog charge integration method.

Three neutron sources were used for investigation: Cf-252, Am–Be, and Am–Li. These sources have distinct neutron energy spectra, both in shape and average neutron energy. The results presented in this paper show that the source identification is possible by the direct analysis of the measured pulse height distributions.

In order to verify the quality of the neutron distributions acquired from measured data, these pulse height distributions were compared with distributions simulated with the MCNP-PoliMi code [16], which is a substantial enhancement of the standard MCNP code [17]. Among other calculations, this code allows for accurate simulations of pulse height distributions for various detectors, taking into account the physical processes governing the pulse creation. Finally, to assess the influence of the potential shielding surrounding the source, and to assess the sensitivity of the PSD method, several source–shielding configurations were tested. In a real-scenario situation, the probability of the presence of shielding around the source is relatively high, either due to transportation reasons, or to intentional source or nuclear material proliferation. We addressed this aspect by using shielding blocks made of lead (Pb) and polyethylene (PE).

The remainder of the paper is organized as follows. Section 2 describes the experimental setup as well as the PSD method used, Section 3 gives a detailed description of Monte Carlo simulations, Section 4 contains final results and discussion, and Section 5 gives conclusions.

2. Digital neutron/\( \gamma \)-ray PSD

2.1. Data collection

A fast waveform digitizer, 1-GHz digital oscilloscope Tektronix TDS-5104, was used to acquire and store tens of thousands of pulses from the anode of the liquid scintillator BC-501A for subsequent PSD analysis. This high-performance digitizer offers 8-bit resolution with a maximum sampling rate of 5 GS/s. The oscilloscope fast-frame acquisition mode allowed for capturing the pulses with the highest resolution. The digitizer was connected to the anode of the PMT via 50\( \Omega \) impedance. The PMT has the same diameter as the scintillator (see the scintillator description below). All data was saved to a PC and postprocessed by using scripts written in Matlab\textsuperscript{\textregistered} language. The measurement time for each source–shielding configuration was approximately 5 min, and the postprocessing time was approximately 10 min.

The neutron sources Cf-252 and Am–Li were placed at a distance of 50 cm from the detector. The construction of the Am–Be source was such that it did not fit the measurement desk when placing at a distance of 50 cm; therefore, it was placed 30 cm from the detector (see Fig. 1). The shielding blocks used in the study were placed next to the detector front face.

The detector BC-501A is a cylindrical liquid organic scintillator model 4.62MAB-1F3BC-501A/5, which is 7.7 cm thick with a diameter of 15.2 cm. The detector container is made of aluminum. The front face of the detector has a thickness of 2 mm. The sidewall is composed of two layers: the external layer has a thickness of 2 mm and the internal layer is approximately 0.5 mm thick. The PMT tube is mounted on the back circular surface of the detector.

2.2. PSD method

As already mentioned in Section 1, the difference in the intensity of slow light components of the pulses generated by neutron and \( \gamma \)-rays is commonly used in PSD methods. One of the methods based on this phenomenon is the charge
integration method, which is very popular due to its simplicity. This method compares the total charge of the pulse with the charge obtained by the partial pulse integration. The range of the partial integral can cover either the slow component or the fast component of the pulse.

We applied the charge integration method offline to the measured pulses. A ratio of two areas was obtained by integration of the pulse in various time intervals. The first area is the total area of the pulse \( A_1 \), and the second area is the tail of the pulse \( A_2 \), as shown schematically in Fig. 2. The integrals are given in a unit of \((V \times ns)\). The tail integral \( A_2 \) is calculated from a certain point above the pulse maximum (see \( T_{2\text{start}} \) in Fig. 2) to the end of the pulse \( (T_{1,2\text{stop}}) \). The optimal start point of this integral was found to be such that \( T_{2\text{start}} - T_{\text{max}} = 11 \text{ ns} \) (see Section 2.3 for details). The total integral \( A_1 \) is calculated by integrating between the values \( T_{1\text{start}} \) and \( T_{1,2\text{stop}} \). The spread of the positions of the pulse maxima \( (T_{\text{max}}) \) is below a value of 10 ns. This has only a negligible effect on the total pulse integral value.

The area ratio \( R \) is defined as

\[
R = \frac{A_2}{A_1}.
\]  

Because the intensity of the light in the slow component is higher for neutrons than for \( \gamma \)-rays, neutron pulses have generally a larger ratio \( R \). Fig. 3 shows an example of measured pulses of all energies from the Am–Be source placed at a distance of 30 cm from the detector. The measurement threshold was set to a light output of 0.094 MeVee (MeV electron equivalent). This corresponds to an electron energy of 0.094 MeV. In Fig. 3, the separation of the neutron and \( \gamma \)-ray pulses can be clearly observed. In fact, only the pulses lying in the valley between the peaks are difficult to classify. A discrimination point \( R_c \) to accurately classify the detected particles must be chosen in this valley to minimize the amount of misclassified pulses.

![Fig. 1. Experimental setup with the scintillation detector BC-501A, Am–Be source, and 1-in. Pb shield.](image1)

![Fig. 2. Integration of pulses over different time ranges for the PSD method. The integral time scale is also shown.](image2)

![Fig. 3. Visualization of pulses measured with the Am–Be source placed at a distance of 30 cm from the liquid scintillator BC-501A. Only a small number of pulses are misclassified with correctly chosen discrimination point \((R_c \approx 0.26)\).](image3)
misclassified pulses. The classification point for the data in Fig. 3 lies at a value of $R_c = 0.26$. Above the classification point, all pulses are classified as neutrons, while below this point the pulses are classified as $\gamma$-rays. Clearly, some of the neutrons and $\gamma$-ray pulses are always misclassified when using this PSD method (or any other method). Therefore, optimization of this method is required to minimize this effect.

2.3. Optimization of the PSD method

The number of misclassified neutrons and $\gamma$-rays can be minimized by optimizing the following parameters: (1) the integration range of the total integral, (2) the integration range of the tail integral, (3) the location of the classification point, and (4) the pulse height threshold. Optimizations (2) and (3) were applied in the past to improve the performance of the PSD method [18]. The start point of the tail integral was set to 11 ns from the peak of the pulse, and the end point to 170 ns from the peak of the pulse. Optimization (1) was eliminated by setting the total pulse range in the measurement to a fixed value of 200 ns, while optimization (4) was abandoned due to the loss of information in the neutron low-energy range when it is applied.

The PSD method was optimized by using correctly attributed neutron and $\gamma$-ray pulses, which were obtained from two known radioactive sources. The neutrons emitted by a Cf-252 source were detected using the time-of-flight (TOF) method, to specifically identify the neutrons in a $\gamma$-ray background. The Cf-252 source was placed in the middle of an ionization chamber at a distance of 1 m from the detector. The ionization chamber was used to provide a start signal for the digitizer and to determine the time zero for the measured neutron pulses. The neutron pulses obtained with this method have been named “TOF-attributed neutrons.” Examples of misclassification of neutrons are accidental coincidences, which result in the attribution of $\gamma$-ray pulses as neutron pulses. The $\gamma$-ray pulses were measured using a Cs-137 source placed on the face of the detector. Fig. 4 shows the neutron and $\gamma$-ray pulses used for the optimization. It can be seen that pulse misclassification occurs mainly for pulses with low total-integral values (low energies) [18].

3. Monte Carlo simulations of the detector pulse height distributions

3.1. MCNP-PoliMi code

The MCNP-PoliMi code was developed to provide accurate information on neutron and $\gamma$-ray time quantities.
This code is a very useful tool for the simulations of correlated events. Specifically, at each collision, all relevant information about neutron and $\gamma$-ray collisions is recorded to a file, which is subsequently analyzed by using a special postprocessing code. For this application, the most relevant information written to the collision file is:

(i) reaction type,
(ii) target nucleus,
(iii) position of collision,
(iv) deposited energy,
(v) collision time, and
(vi) incident particle energy.

3.2. Detector pulse height distributions from the MCNP-PoliMi code system

The MCNP-PoliMi code, in conjunction with the postprocessor, can simulate neutron and $\gamma$-ray pulse height distributions for liquid and plastic scintillation detectors. Naturally, the type of detector used in the simulation significantly influences the amount of light produced in the detector as a function of incident energy; this is taken care of in the postprocessing analysis of the simulation results. The MCNP-PoliMi pulse height distributions were compared to the distributions acquired from the measured data using the optimized PSD method.

In the simulations, all three neutron sources tested with the PSD method were considered as point and isotropic

<table>
<thead>
<tr>
<th>Average source energies</th>
<th>Cf-252</th>
<th>Am–Be</th>
<th>Am–Li</th>
</tr>
</thead>
<tbody>
<tr>
<td>$E_n$ (MeV)</td>
<td>2.13</td>
<td>4.22</td>
<td>0.36</td>
</tr>
</tbody>
</table>

Fig. 6. Histograms from the pulses generated by the Cf-252, Am–Be, and Am–Li sources. The Am–Li source produces only a small number of neutrons relative to $\gamma$-rays. In each figure, the peaks on the left belong to $\gamma$-ray pulses, and the peaks on the right represent neutron pulses.
sources. Therefore, some observable differences between the measured and simulated data can be attributed to the source simplifications in the MCNP-PoliMi runs. The source distributions implemented in MCNP-PoliMi were used in the simulations. The MCNP-PoliMi neutron source energy spectra are shown in Fig. 5. The neutron spectrum for Cf-252 is identical to the spectrum originally used in the MCNP-DSP code [19]. The implementation of Am–Be and Am–Li is described in Ref. [20]. The average neutron energies of these sources are shown in Table 1. It should be noted that the measured pulse heights were transformed to light outputs by means of the standard Cs-137 calibration measurement.

In order to calculate the detector pulse height distribution, the recorded collision file is analyzed. There are several adjustable parameters for the analysis:

(i) pulse generation time,
(ii) dead time,
(iii) acceptance threshold,
(iv) light output range of interest, and
(v) light output binning.

After these parameters have been adjusted by the MCNP-PoliMi user, and the collision file has been analyzed, the pulse height distributions are created from the accepted light pulses.

4. Results

4.1. Measured pulses from the Cf-252, Am–Be, and Am–Li neutron sources

Fig. 6 shows the histograms created from the pulses generated by the Cf-252, Am–Be, and Am–Li neutron sources. In the measurements shown in Fig. 6, no shielding was used. For all three sources, very good separation of neutrons from $\gamma$-rays is observed. Further, it is apparent...
that the Am–Be source provides the highest number of neutrons relative to the γ-rays. On the other hand, the Am–Li source generates only a small number of neutrons if no Pb shielding is used. As a result, it is very difficult to obtain accurate “neutron” information, and Pb shielding should be used to eliminate the strong γ-ray background. It should be noted that due to the low-energy neutrons from the Am–Li source a higher pulse height resolution was used in the measurement. This resolution is a factor of two higher than the resolution used for the two other sources. This change of resolution is reflected in Fig. 6 in the different position of the neutron and γ-ray peaks for the Am–Li source.

Fig. 7 shows the pulse tail integrals \( A_2 \) as a function of the total integrals \( A_1 \) of the pulses from the Cf-252, Am–Be, and Am–Li neutron sources. A very good separation of neutrons from γ-rays can be observed for all neutron sources. It also should be noted that the integral ranges differ as a consequence of the substantially different neutron spectra for these sources (see Fig. 5).

4.2. Different detector-shielding configurations

In this investigation, about 27,000 neutron and γ-ray pulses were acquired for each source–shielding configuration. Fig. 8 shows the comparison of various detector-shielding configurations for the Cf-252 source. In this comparison, the influence of the Pb and PE shieldings is presented on the performance of the PSD method. The results are shown as a ratio of tail-to-total integral as a function of the pulse height. As expected, the presence of the Pb shield between the source and the detector causes the elimination of a certain number of γ-rays, which results in a larger number of neutrons, compared to the case without the shielding. No dramatic increase in the relative number of the neutron pulses is observed by transition

![Graphs showing pulse height vs. ratio of tail-to-total integral for different shielding configurations](image-url)
from 1 in. of Pb to 3 in. of Pb. On the other hand, by using 3 in. of PE shield, the number of neutrons decreases significantly as a result of neutron scattering and subsequent neutron absorption.

4.3. Measurements versus simulations

Fig. 9 shows the comparison of simulated and measured pulse height distributions for the Cf-252 source with different source–shielding configurations. The measured data were collected with the same binning as the simulated data and normalized to the area to allow direct comparison. In Fig. 9, for all configurations, a very good agreement has been achieved. In the cases of the configurations without the shielding and with 1 in. of PE, respectively, the measurement data exhibit lower statistical accuracy than the configurations with the Pb shielding. This is due to the lower number of neutrons in the dataset.

The measurement errors shown in Fig. 9 were calculated as the square roots of the measured data values.

Fig. 10 shows the measured and simulated pulse height distributions for the Am–Be and the Am–Li sources. For both sources, the configurations with Pb shields are shown. The configuration with 1 in. of PE is shown for the Am–Be source only because this configuration, when used for the Am–Li source, leads to a complete loss of neutron information.

While in the case of the Am–Be a very good agreement can be observed in Fig. 10, for the Am–Li source, a relatively large difference is observed between the measurement and the simulation. This difference is caused by a relatively low number of neutrons emitted from this source when compared to the γ-rays. Consequently, it is very difficult to acquire accurate information necessary to achieve a reasonably good agreement between the measured and simulated data. In addition, because of the low
average energy of the Am–Li neutrons the influence of the measurement background on the data is enhanced, which can also alter the measured data.

Fig. 11 shows the comparison of the measured pulse height distributions for all three sources and several source–shielding configurations used in this work. The results are shown using identical ranges of the normalization areas for the Cf-252 and Am–Be data. For the Am–Li data, this range was different due to substantially different neutron energy spectrum of the source and corresponding different light output spectrum.

It is apparent that the general shape of the distributions is not significantly changed with the presence of the tested shields. However, the presence of PE increases uncertainty of the measured data, especially at high light outputs. In Fig. 11, a very clear distinction between the Cf-252 and the Am–Be source is observed below the light output value of 0.4 MeVee. Below this value, statistical error plays only a minor role. Above this value, the general slope of the distributions can be still used for the source identification, even though statistical uncertainties are increased. The 3-in. PE blocks used for the Cf-252 and the Am–Be cause larger fluctuation of the measured distribution points when compared to other source–shielding configurations, especially above 1 MeVee. This fluctuation, however, still allows the identification of the sources. The Am–Li source can be very easily distinguished from two other sources due to its low pulse height distribution profile.

Based on the results presented in this paper, all tested sources can be clearly distinguished from each other, in an accurate and timely manner, which makes it easy to identify typical neutron source distributions, such as the fission spectrum of Cf-252, the double-peak Am–Be spectrum, and the low-energy Am–Li spectrum.
5. Conclusions

An existing technique has been applied for the first time to accurately identify neutron sources, based on the direct analysis of measured neutron pulse height spectra using a liquid scintillator BC-501A and a fast Tektronix waveform digitizer TDS-5104. An optimized digital PSD method was applied to a large number of pulses originating from various sources to obtain the pulse height distributions. These distributions were compared to distributions simulated with the MCNP-PoliMi code and excellent agreement was obtained for the Cf-252 and the Am–Li source. The measurement and simulation results for the Am–Li source are slightly less good due to the relatively low number of neutrons emitted from this source when compared to the γ-rays and consequent uncertainty in the measured neutron pulse height spectrum. In order to test the influence of the source shielding on the identification technique, lead and polyethylene blocks were used. It is apparent that the presence of the tested shields does not significantly alter the distributions, and the results can be clearly distinguished even with 3 in. of polyethylene (Cf-252, Am–Be) between the source and the detector. The presence of lead helps to eliminate the γ-ray background and, thus, improves the performance of the source identification technique.

The results show that the identification of Cf-252, Am–Be, and Am–Li sources in both shielded and unshielded configurations is easily and robustly achievable by the direct analysis of the measured pulse height distributions, without the need of subsequent neutron energy spectrum unfolding. While unfolding methods are frequently used in mixed neutron/γ-ray fields to identify neutrons, the technique presented in this paper has several important advantages: (i) the measurement time for the neutron pulse height distribution is shorter (unfolding requires very high accuracy of measured pulse height distribution data and therefore much longer measurement times); (ii) there is no need for accurate measurements of the detector response matrix using monoenergetic neutrons, as is needed for unfolding procedures; and (iii) additional time is saved by eliminating the neutron spectrum unfolding procedure altogether.

The results prove the potential of liquid scintillators to become the standard components of reliable neutron-detection systems in nonproliferation and nuclear safeguards applications. In the future, we plan to test other neutron sources (for example, Pu–Be, Pu-240) to verify the robustness and versatility of this technique.

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References
