

Time-of-Flight Spectroscopy of ^{252}Cf Spontaneous Fission Neutrons: Influences of Detector Voltage, Pulse-Shape Discrimination and Shielding

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Abstract—Experimental measurements to explore the effect of detector voltage, pulse-shape discrimination (PSD) threshold and detector shielding on time-of-flight measurements of the ^{252}Cf neutron spectrum made with organic scintillation detectors are described. It is found that detector voltage has a major effect, whilst changing the PSD threshold and shielding the detectors to optimize sensitivity to the desired γ -neutron correlation results in a small effect.

I. INTRODUCTION

MANY of the recorded attempts to measure energy spectra of spontaneous fission (SF) neutrons experimentally have employed the time-of-flight (ToF) technique, often for ^{252}Cf but also for ^{244}Cm and ^{240}Pu , for example [1-7]. The technique is straightforward in principle, being based on an estimate of the time taken for a neutron emerging from fission to travel a known distance. However, it can be complicated in practice by the effects of consistency of detector set-up, changes in a given measurement environment in terms of shielding and scatter, detector crosstalk and misclassification of neutrons and photons. Nonetheless, it remains one of the few deterministic routes to obtaining an experimental measurement of the SF neutron spectrum, with which to complement simulations and stochastic measurements based on unfolding of overlapping sets of event data.

TOF spectroscopy can be performed using associated particle methods, looking for correlated gamma- neutron detections assumed to have originated in the same fission event. As the gamma rays will travel at the speed of light, at these scales of distance they can be treated as detected at the instant they are produced, and therefore provide a start time for the travel of the neutron, and a means of measuring its time of flight, and therefore energy.

In this paper, we describe the results of research performed to determine the extent of the effects of systematic changes on a detector set-up designed to measure TOF SF spectra for ^{252}Cf neutrons. The effects of changing detector voltage, shielding around the detector and relative severity of pulse-shape discrimination settings have been explored.

II. METHOD

Two EJ-309 organic scintillator detectors (of type VS-1105-2, Scionix, Netherlands) have been used in this research. Events from these detectors have been processed by a specially adapted mixed-field analyzer (Hybrid Instruments, UK). This analyzer uses an FPGA to perform real time pulse digitization, pulse shape particle discrimination (PSD), and high precision timing, between the two detector channels, allowing the system to perform real time TOF spectroscopy.

The detectors were positioned to optimize the duration between these events whilst ensuring that detection efficiency is not compromised too greatly, as greater separation between detectors and source will increase the neutron flight time and therefore improve energy resolution but impair efficiency through reduced 4π coverage. For this work a detector distance of 70cm for the main detector and 20cm for the trigger detector was used. The closer detector to the source is used as the trigger detector, and gamma events detected in this detector opens a coincidence window a user specified time width (in this case 100ns). If during this window a neutron is detected in the second detector, the two events are presumed to be coincident, and the time between these detections is logged and along with other coincident events used to calculate the neutron energy spectrum.

Two ^{252}Cf sources were used for these measurements. To investigate the effect of the detector set up a 70 MBq sealed source accommodated in a 1 m³ water tank in the Department of Engineering at Lancaster University, UK was used. This source is stored within the water tank and moved to the edge for use. However, as both the large mass of water and associated building clutter in close proximity to the source and detectors provides a potentially large source of scatter neutrons, an additional measurement was made using a bare ^{252}Cf source at the National Physical Laboratory (NPL, London, UK). This bare source measurement was performed in a low scatter environment, using the same setup in terms of equipment and detector layout, providing a comparable measurement relatively

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unperturbed by scatter. A photograph of the Lancaster set-up is provided in Fig. 1.

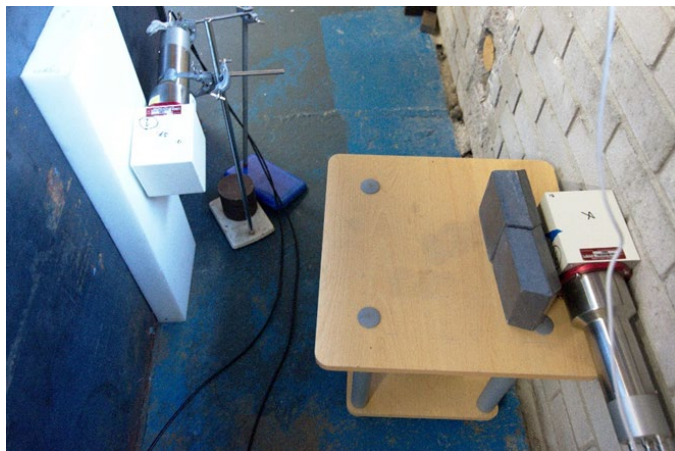


Fig. 1. Photograph of the experimental set-up, ^{252}Cf source (in tank) to the left behind a 5-cm HDPE slab and the trigger detector, and the neutron counter shielded with 5 cm lead to the right.

To explore the influences of detector voltage on the ToF measurements, three measurement campaigns were performed at high, medium and low voltage settings (in relative terms recognizing that to sustain consistent PSD there are limits beyond which the technique is undermined, so these were avoided). To explore the influence of PSD two threshold settings were investigated: i) a setting offering a compromise between avoiding misclassification and maintaining event throughput and ii) that which is more conservative to err on the side of γ rays for the trigger detector and on the side of neutrons for the neutron detector. To assess the influence of shielding: i) 5 cm of high-density polyethylene (HDPE) was installed local to the trigger detector, in order to reduce the potential for neutrons to masquerade as trigger γ rays when misclassified, and ii) 5 cm of lead to perform the inverse for the ToF detector.

III. RESULTS

Results for the measurements are presented below. Fig. 2 shows the results of the bare source measurement taken at NPL. Alongside the measured data the known ^{252}Cf Maxwellian spectrum, taken from literature [8] is plotted. To account for the intrinsic detection efficiency of the scintillant, this spectrum has been folded with the detector efficiency, and is referred to as MaxEFF. Indeed, for all of the presented spectra, it is worthy to note that assuming an approximate cut-off in neutron energy of 700 keV, only the trends above this energy are to be considered; less than this level is indicative of the response of the scintillant rather than of ^{252}Cf itself.

The higher low energy cutoff of the bare source NPL measurement shown in Fig. 2 is the function of the threshold settings used, which in this case were higher than for subsequent measurements.

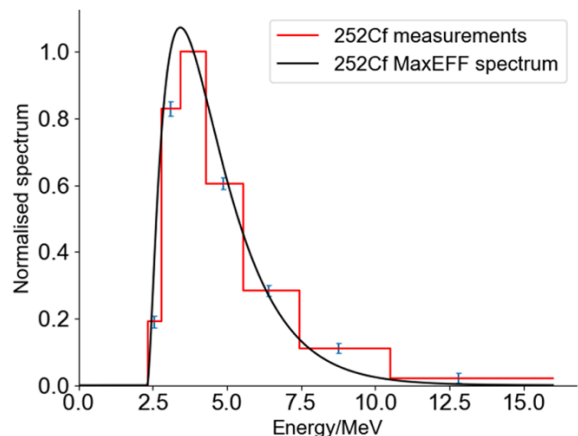


Fig. 2. Spectra taken from a bare ^{252}Cf source in the low scatter facility at NPL. The relatively high cutoff at 2.5MeV is a function of the detector voltage and threshold settings. The 252Maxeff spectrum plotted alongside the data is the Maxwellian spectrum for ^{252}Cf taken from literature [8], folded with the known detector response.

Results for each parameter measurement taken at the Lancaster University source are presented in Figs. 3-5. Firstly, Fig.3 shows the effect of changing detector voltage on the measured spectrum. As can be seen, changing the detector voltage has a marked effect on the low energy cutoff of the spectrum. This can be understood in terms of the reduced pulse height with reduced voltage. With a lower applied voltage, a neutron depositing a certain amount of energy in the scintillant will produce a smaller output pulse, more likely to fall below the threshold value and be discarded. Therefore at lower neutron energies the detection efficiency will be disproportionately affected. As the neutron energy is measured as a function of time rather than energy deposition, the high energy end of the spectrum is largely unaffected.

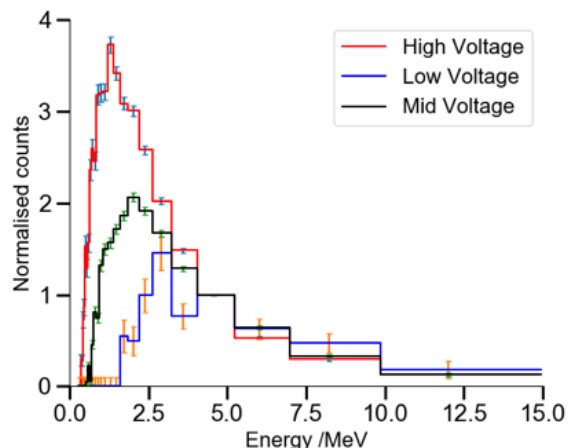


Fig. 3. ^{252}Cf neutron spectrum (tank) with changing detector voltage.

Fig. 4 shows the effect that changing the PSD settings has on the measured spectrum. As can be seen, for events above the peak at 2.5MeV, the response is unaffected within statistical uncertainty. Below this peak however, strict PSD is seen to reduce the detector response very slightly. This effect is consistent with the PSD process reducing the detection

efficiency at low energies, as there is greater misidentification at the small pulse heights which are more overwhelmingly more likely to be produced by low energy neutrons.

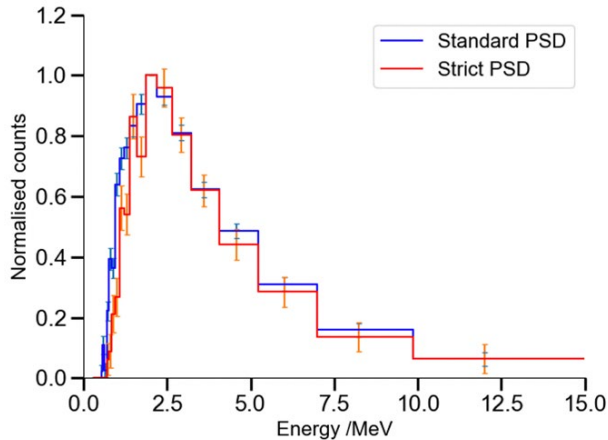


Fig. 4. ^{252}Cf neutron spectrum (tank) with changing PSD threshold. Showing the effect of a strict PSD approach against one less conservatively chosen.

Fig.5 shows the effect of providing shielding to the detectors. This produces no discernable effect below the 2.5MeV peak, but, whilst close to the statistical limit, suggests that the shielding lowers the response for $E_n > 1.5$ MeV. This effect is hypothesized to be produced by the reduction via shielding of the cases where neutron-neutron events are erroneously classified as γ -neutron events. In these cases, the slower travel time (compared with a gamma ray) of the initial, triggering neutron will delay the start of the timing of the second neutron, which will then be presumed to have travelled more quickly and therefore possess a higher energy than in reality. This has the effect of erroneously introducing high energy events into the spectrum.

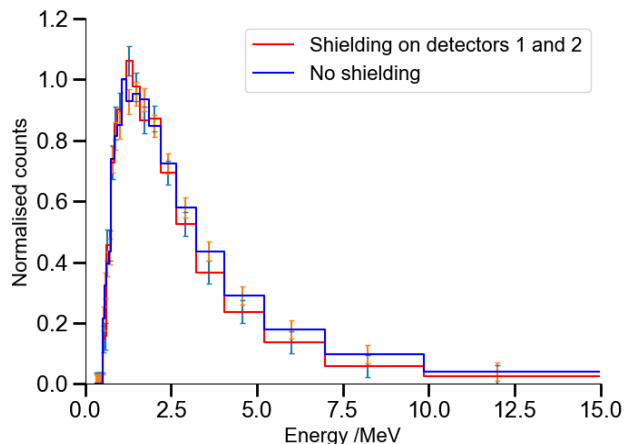


Fig. 5. ^{252}Cf neutron spectrum (tank) as a function of shielding.

Whilst the shielding will also reduce the number of neutron- γ events misclassified as γ -neutron events, these coincidental detections are not correlated and therefore have no probabilistic preference towards a certain energy. As a result, they will be removed along with other accidental coincidences during data processing.

IV. DISCUSSION

The bare source measurements show good agreement with the known spectrum, as per a T -parameter [8] of 1.42, indicating the consistency of the approach with the prior art [7]. This demonstrates that this method of obtaining neutron time of flight measurements can be used to perform spectrometry of fission sources in the field and in real time. This is a marked development over traditional ToF measurements which require either accelerator facilities, or special source preparation within ionization chambers [7] [9]. Whilst in this research fission sources have been used, there is no reason that it cannot be used on other neutron sources where the neutron emission is accompanied by another radiation product, provided that it is temporally correlated and produced within short (<100ns) timescales. An example is [10] where a similar approach has been used to measure the AmBe neutron spectrum. Perhaps one consideration that must be made when making these measurements however is that some knowledge of the neutron source in that only the component with correlated particle emission contributes to the spectrum. This requirement for prior knowledge however is a common requirement for performing neutron spectrometry using other frequently used techniques, such as spectrum unfolding approaches [11]. The ToF technique however has the advantage of providing a deterministic measurement of the spectrum, rather than one that is subject to stochastic interpretation.

The further measurements show that it is relatively sensitive to changes in experimental conditions and setup, although this is the case for other neutron spectrometry methods as well. In this case however it can be seen from these measurements that experimental changes and inconsistencies can have disproportionate effects on different parts of the spectrum.

For example, comparison of the spectra measured under contrasting shielding schemes shows an appreciable effect in changing the relative contributions between high and low energy neutrons. Whilst subtle, these measurements suggest using polyethylene shielding on the trigger detector reduces the contribution towards the spectrum above the peak neutron energy. As previously stated, this can be understood as an effect of reducing the numbers of misidentified neutron detections. What is still not understood from this measurement alone is the magnitude of the scatter component that this PE shielding will potentially introduce, and further measurements or modelling work would prove valuable to provide further insight into this component of the measured spectrum.

Shielding the second detector from excess gamma rays would appear to offer no advantage from a spectrum fidelity point of view but may offer advantages in high count rate environments where reducing data volume for processing is desirable.

Measurements exploring the detector voltage show the effect of this parameter on the measured spectrum is fairly marked, most clearly, a reduction of the detector voltage results in an increase of the low energy cutoff observed in the spectrum. Indeed, if the low energy end of the spectrum (down to approximately 700keV, the intrinsic cutoff in the scintillant response) is of interest, the voltage settings chosen must be high

enough to reflect this. In addition to the cutoff threshold, a reduction of photomultiplier voltage shows a general reduction in detection efficiency. This behavior is not particularly surprising in the light of detector behavior whilst used in a more usual pulse height mode. As increasing the voltage applied across the photomultiplier tube increases the magnitude of the pulse produced during a particle detection, a lower incident particle energy is required to illicit a detector response that falls above the threshold setting on the detector. Similar to standard pulse height spectrum measurements this effect will be observed more strongly at lower energies, and therefore the detector efficiency will be affected disproportionately with reducing energy. A characteristic that contrasts with pulse height spectrum measurements however, is that regardless of photomultiplier voltage (and therefore pulse height), the neutron energy is measured consistently across different voltages, as the neutron energy is measured as a function of time of flight rather than the quantification of energy deposited in the scintillant. Detector voltage therefore must be carefully set and maintained between measurements to provide consistency of detection efficiency across the spectrum, but an energy axis calibration (such as using peaks in the gamma spectrum of check sources as is usual when making pulse height spectrum measurements) is not required.

Particle discrimination is an important aspect of all neutron measurements where the detector is sensitive to gamma rays as neutrons are not produced in isolation. In this technique however the intrinsic difference in arrival time can be largely used to accurately sort between neutron and gamma events and so particle discrimination is primarily used as a means of reducing data throughput and thus the burden on processing capacity. Conversely, when performing neutron spectrometry using pulse height spectrum measurements, the particle discrimination settings must be carefully balanced to reduce gamma contamination of the spectrum whilst also avoiding discarding true neutron counts. As can be seen in these results, the loss of low energy neutrons to the PSD process can be seen in the spectrum and a sensible strategy is therefore to avoid overzealous PSD settings, allowing miss-characterized gamma rays to be sifted out by virtue of their early arrival at the detector in comparison with true neutron events.

V. CONCLUSIONS

This work shows the ability of a portable real time system utilizing two scintillator detectors to perform associated particle ToF spectroscopy. This offers an opportunity to make practical deterministic measurements of the neutron energy spectra of fission sources. Furthermore, provided the experimental parameters are chosen appropriately, a good low energy response can be obtained, and the technique is resistant to gamma contamination of the spectrum.

Whilst this method offers a powerful means of measuring neutron spectra, the subsequent measurements have illustrated some of the subtleties associated with obtaining consistent results. Firstly, the scatter contribution must be understood in a given environment, although this is largely the case for any

neutron energy measurements. Likewise, detector set up must be kept consistent in terms of photomultiplier voltages, thresholds and particle discrimination settings.

It has also been demonstrated that the effects of these inconsistencies will not tend to be seen across the spectrum evenly. In the case of over-conservative PSD settings and lowered photomultiplier voltage, changes to the spectrum are observed either predominantly or exclusively at the low energy portion of the spectrum. For different shielding approaches however, the effect is at the high energy component, or at least the relative balance of low-to-high energy response.

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REFERENCES

- [1] D. A. Hicks, J. Ise Jr. and R. V. Pyle, 'Spontaneous-fission neutrons of californium-252 and curium-244', *Phys. Rev.* 98 (5) pp. 1521-1523 (1955).
- [2] P. M. Belov et al., *Yadernaya fizika* 9 (4) pp. 727 (in Russian) (1969).
- [3] F. Herold, Savannah River Laboratory Report, DR-949 (1969), Proc. Of a Consultants Meeting, Vienna, pp. 18 (1972).
- [4] Yu.S. Zamyatin et al., 'Measurement of the energy spectra and the average number of prompt neutrons produced in the fission of actinide elements', *Nuclear Data for Reactors (Proc. of the 2nd Int. Conf. on Nucl. Data for Reactors, Helsinki, 1970), IAEA, Vienna, vol.2, p.183 (1970), in Russian.*
- [5] Aleksandrova, Z.A., Bol'shov, V.I., Kuznetsov, V.F., Smirenkin, G.N., Tarasko, M.Z., 'Spectra of the prompt neutrons arising from the spontaneous fission of ^{252}Cf , ^{244}Cm , and ^{240}Pu ', *Sov. At. Energ.* 36 (4) pp. 355-358 (1974).
- [6] O. I. Batenkov, G. S. Boykov, L. V. Drapchinsky, M.Ju. Majorov and V.A. Trenkin, 'The measurement of prompt neutron spectrum in spontaneous fission of ^{244}Cm ', pp. 158-165. In: 'Selected papers of ISTC workshop on nuclear data of minor actinides', May 27-31 JAERI. Tokai, Japan(1996), https://inis.iaea.org/collection/NCLCollectionStore/_Public/28/057/28057899.pdf?r=1&r=1.
- [7] W. Mannhart, 'Properties of Neutron Sources', Proc. of an Advisory Group Meeting on Neutron Sources, Leningrad, June 1986, IAEA-TECDOC-410, IAEA, Vienna, pp. 158 (1987).
- [8] J. Terrell, 'Fission neutron spectra and nuclear temperatures', *Phys. Rev.* 113 (2) pp. 527-541 (1959).
- [9] M. Devlin et al., "The Prompt Fission Neutron Spectrum of $^{235}\text{U}(n,f)$ below 2.5 MeV for Incident Neutrons from 0.7 to 20 MeV," *Nucl. Data Sheets*, vol. 148, pp. 322-337, 2018.
- [10] J. Pal, S. Saha, C. C. Dey, B. K. Sinha and M. B. Chatterjee, "Neutron energy spectrum of an Am-Be source by gamma gated NTOF technique," *Radiat. Phys. and Chem.*, vol. 51, no. 4-6, pp. 475-477, 1998.
- [11] M. Reginatto, "Overview of spectral unfolding techniques and uncertainty estimation," *Radiat. Meas.*, vol. 45, no. 10, pp. 1323-1329, 2010.