The angular dependence of pulse shape discrimination and detection sensitivity in cylindrical and cubic EJ-309 organic liquid scintillators

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ABSTRACT: Liquid scintillators are used widely for neutron detection and for the assay of nuclear materials. However, due to the constituents of the detector and the nitrogen void within the detector cell, usually incorporated to accommodate any expansion that might occur to avoid leakage, fluctuations in detector response have been observed associated with the orientation of the detector when in use. In this work the angular dependence of the pulse-shape discrimination performance in an EJ309 liquid scintillator has been investigated with $^{252}$Cf in terms of the separation of $\gamma$-ray and neutron events, described quantitatively by the figure-of-merit. A subtle dependence in terms of pulse-shape discrimination is observed. In contrast, a more significant dependence of detection sensitivity with the angle of orientation is evident.

KEYWORDS: Neutron detectors (cold, thermal, fast neutrons); Scintillators, scintillation and light emission processes (solid, gas and liquid scintillators)
1 Introduction

Organic liquid scintillation detectors have long been used for fast neutron detection ever since the late F.D. Brooks discovered that the difference in scintillation decay times between heavily-ionizing particles and photons could be exploited to discriminate fast neutrons and γ rays [1]. However, for a long period of time their use was confined to experimental laboratories and physics research installations on account of their being highly flammable and potentially hazardous to use in industrial environments. Recently, low-hazard variants have become available and very recently solid forms of organic materials offering similar pulse-shape discrimination (PSD) properties to those of liquids were discovered [2]. These developments, along with the advent of real-time digital electronic PSD processing [3–5], have stimulated a great deal of research interest in the use of organic scintillation materials for industrial applications associated with, for example, nuclear safeguards and security [6, 7].

With these capabilities it is feasible to realize fast-neutron multiplicity assay with very low accidentals rates and a relatively small number of detectors [8, 9]. Often, to remove the $1/r^2$ spatial dependence from measurements by positioning of detectors equidistant from a source or sample under scrutiny, it is desirable to arrange detectors comprising an array throughout the range of $4\pi$. In this case some cells in such a system can be required to operate consistently with one another despite being at contrasting angles of orientation; similar demands are made of portable systems where the orientation of detectors is rarely consistent from one measurement to another. Whilst solid scintillators are not expected to be affected by orientation on account of the material stiffness of the scintillant, with liquids there is the settling of the fluid that in some circumstances has the potential to influence the amount of liquid in contact with the photomultiplier tube (PMT) and thus affect the light transport to the photocathode. In figure 1 a detector is depicted schematically in nine different orientations with respect to a source located at $0^\circ$.

In each of these orientations the expansion volume inside the cell will be in a different location within the detector. When the detector is at $-90^\circ$, it is anticipated that the bubble will settle above the liquid scintillant, but below the optical window of the PMT, offering maximum influence on the light transport of the detector. As the angle is increased towards the horizontal position at $0^\circ$, the effect of the bubble is expected to decrease until it has no influence on the light transport to the PMT through the scintillant. This is the origin of the anticipated effect on the response of an individual detector.
The pioneering work in this regard of late is that of Naeem et al. [10] who explored the dependence of pulse height spectra (PHS) with the angle of orientation of organic liquid scintillation detectors. In this work, two contrasting detector volumes were explored both with simulations and with exposure to a $\gamma$-ray source. The position of the Compton edge in the PHS for the larger detector was observed to undergo a significant change as a result of positioning, whilst the smaller detector with a design feature to keep the bubble off the PMT window did not exhibit such a dependency.

In this paper we describe the influence of orientation on two contrasting detector shapes with the objective of determining whether the PSD performance of either detector is affected.

2 Experimental details & results

The experiments in this research were carried out using a 75 MBq $^{252}$Cf source situated in a large (approximately 1 m $\times$ 1 m $\times$ 1 m) water bath comprised of a plate steel box (of thickness 33 mm) in the Engineering Department at Lancaster University, U.K. The source is moved to the front face of the inner hull during exposure, with the apparatus set up outside the vessel as shown in figure 2a and 2b, such that the source is a distance of approximately 150 mm away from the front face of the detector.

The first set of experiments used a VS-1105-21 detector (Scionix, Netherlands) that has a sensitive volume of dimensions 100 mm $\times$ 100 mm $\times$ 120 mm containing the low-hazard EJ-309 scintillant (Eljen Technology, Sweetwater, TX). This was connected to a single channel of a 4-channel mixed-field PSD analyzer (MFA, Hybrid Instruments Ltd., U.K.) out of a stack of four MFAs, as shown in figure 2c. These units perform PSD processing in real-time with an event throughput of $3 \times 10^6$ events per second and 6 ns time jitter [11]. The PSD operation is based on the pulse-gradient analysis method [3] in which two samples are taken, one of the pulse at the peak and the other in the falling edge of the pulse. These are compared following processing with the moving-average filter and interpreted as the first and second integral respectively.
 Firstly, experimental runs at $0^\circ$ (horizontal), $+45^\circ$, $-45^\circ$ and $-90^\circ$ were performed to calibrate the HV supply for the detector with $^{137}$Cs. These were carried out to match the gains on the detector to ensure they were consistent with each other whilst varying between the angular orientations. The duration of these runs were set to be sufficient to collect several thousand events. The PHS obtained via this approach were all observed to be consistent in shape and in terms of the position of the Compton edge, meaning that the detector would not need further calibration between the varying angular orientations when under investigation. After these gain-matching measurements were taken, the source was changed to the $^{252}$Cf source described previously. A scatter plot of first versus second integral was then collected in order to set the PSD thresholds, figure 3; to obtain the data necessary for this the detector was exposed to the source for one hundred thousand and one hundred pulses (due to the analyser recording an extra 0.1% of counts required), with the detector at $0^\circ$. The thresholds for determination of gamma and neutron events is set by placing three points on the scatter plot between the plumes. The corresponding scatter plot after it has been normalised is shown in figure 4. The processing of the data for this plot is elaborated upon in section 3.

After the calibration run the detector was set at each of the angles with respect to the source as previously depicted in figure 1. Each angle was set using a clamp stand and a magnetic elevation meter, accurate to $\pm 0.5^\circ$. The system was run for 5 minutes to allow it to stabilise and then the $^{252}$Cf source was exposed.

The $\gamma$-ray and neutron counts produced by the MFA according to the previously set thresholds are provided at varying angles in table 1. The ratio of $\gamma$-rays to neutrons is consistent throughout,
Figure 3. Example scatter plot at $0^\circ$ using the cubic detector with thresholds set to colour the gamma events red and the neutron events blue.

Figure 4. Normalised scatter plot at $0^\circ$ using a cubic detector of type VS-1105-21, $\gamma$ rays (lower), neutrons (upper). Data have been normalized.

although it was observed that the acquisition time to reach the desired counts varied at some angular orientations. To quantify this effect each run at each orientation was repeated for a set exposure time of 2 minutes, and the number of counts observed in each case compared; these data are shown in table 2.
Table 1. γ-ray and neutron composition of 100100 events with $^{252}\text{Cf}$ as a function of angle for the cubic cell.

| Angle/° | γ-ray events | Neutron events | γ-ray/neutron ratio |
|---------|--------------|----------------|-------------------|
| 90      | 82302        | 17798          | 4.624 ± 0.038     |
| 67.5    | 82320        | 17780          | 4.630 ± 0.038     |
| 45      | 82267        | 17833          | 4.613 ± 0.038     |
| 22.5    | 82390        | 17710          | 4.652 ± 0.039     |
| 0       | 82187        | 17913          | 4.588 ± 0.038     |
| −22.5   | 82282        | 17818          | 4.618 ± 0.038     |
| −45     | 82226        | 17874          | 4.600 ± 0.038     |
| −67.5   | 82161        | 17939          | 4.580 ± 0.038     |
| −90     | 82316        | 17784          | 4.629 ± 0.038     |

Table 2. γ-ray and neutron composition with $^{252}\text{Cf}$ as a function of angle for 2-minute exposure for the cubic cell.

| Angle/° | γ-ray events | Neutron events | γ-ray/neutron ratio | Total Counts |
|---------|--------------|----------------|--------------------|--------------|
| 90      | 194970       | 41530          | 4.695 ± 0.025      | 236500 ± 486 |
| 67.5    | 262231       | 56969          | 4.603 ± 0.021      | 319200 ± 565 |
| 45      | 329524       | 71576          | 4.604 ± 0.019      | 401100 ± 633 |
| 22.5    | 380668       | 82832          | 4.596 ± 0.018      | 463500 ± 681 |
| 0       | 392768       | 85732          | 4.581 ± 0.017      | 478500 ± 692 |
| −22.5   | 313248       | 69452          | 4.510 ± 0.019      | 382700 ± 619 |
| −45     | 271227       | 59773          | 4.538 ± 0.021      | 331000 ± 575 |
| −67.5   | 213214       | 46186          | 4.616 ± 0.024      | 259400 ± 509 |
| −90     | 172920       | 36980          | 4.676 ± 0.027      | 209900 ± 458 |

The next stage of the experiment carried out identical tests with a second detector, this time with a scintillant cell of a different shape but still containing the organic liquid material EJ-309. The cell shape measured 130 mm in diameter with a volume approx. 1 liter. This would allow comparison between the two shapes with respect to the effect of orientation on discrimination performance. This was carried out in exactly the same way as described above for the first detector but with a cylindrical detector cell as opposed to cubic as in the first instance. The detector HV was calibrated as before with a $^{137}\text{Cs}$ source, only this time it was noted that the PHS showed that there was a significant shift in position of the Compton edge when at the orientation of $−90°$. The detector was then exposed to the $^{252}\text{Cf}$ source to determine the PSD thresholds and a normalised scatter plot of the result is shown in figure 5.

As before the detector was then set to each of the varying angular orientations and the events of each radiation type recorded for a set one hundred thousand and one hundred counts. The results showing the contribution of each radiation type for each orientation with the cylindrical cell detector is shown in table 3. As can be seen, at $−90°$ for this detector the ratio of γ-rays to neutrons is lost. The scatter plot corresponding to the $−90°$ measurement, showed a significant variation in the position of the neutron and γ-ray distributions at this angle; as mentioned a significant shift was
also observed in the PHS supporting why the ratio appears to have disappeared. To resolve this it was required that the PSD threshold needed to be adjusted for this individual specific angle with the cylindrical detector. Following adjustment, the measurements at this angle were repeated with the revised PSD thresholds and the results are given in Table 4.

Table 3. $\gamma$-ray and neutron composition with $^{252}$Cf as a function of angle for 100100 counts for the cylindrical cell.

| Angle/° | $\gamma$-ray events | Neutron events | $\gamma$-ray/neutron ratio |
|---------|----------------------|----------------|---------------------------|
| 90      | 67051                | 33049          | 2.029 ± 0.014             |
| 67.5    | 66947                | 33153          | 2.019 ± 0.014             |
| 45      | 66758                | 33342          | 2.002 ± 0.013             |
| 22.5    | 66487                | 33613          | 1.978 ± 0.013             |
| 0       | 66146                | 33954          | 1.948 ± 0.013             |
| −22.5   | 66378                | 33722          | 1.968 ± 0.013             |
| −45     | 66464                | 33636          | 1.976 ± 0.013             |
| −67.5   | 64813                | 35287          | 1.837 ± 0.012             |
| −90     | 22041                | 78059          | 0.282 ± 0.002             |

Once the threshold had been recalibrated three runs were performed to check consistency of the new scatter plot. For this one particular angle there is still evidence that there are two distinct regions, although the regions exhibit less separation than is seen in all other plots from different angular orientations. With separation between neutron and $\gamma$-ray plumes reduced, an increase in the uncertainty in PSD is likely and reduces confidence with which one radiation type can be
Table 4. $\gamma$-ray and neutron composition with $^{252}$Cf at $-90^\circ$ for 100,000 counts for the cylindrical cell.

| Measurement | $\gamma$-ray events | Neutron events | $\gamma$-ray/neutron ratio |
|-------------|----------------------|---------------|---------------------------|
| 1$^{\text{st}}$ | 88631                | 11469         | 7.73 ± 0.08               |
| 2$^{\text{nd}}$ | 88471                | 11629         | 7.61 ± 0.08               |
| 3$^{\text{rd}}$ | 88751                | 11349         | 7.82 ± 0.08               |

successfully discriminated from the other leading to the misleading increase in the $\gamma$-ray/neutron ratio.

As with the cubic cell, it was observed with the cylindrical cell that the acquisition time for some positions varied with the angular orientation, table 5 provides the counts observed for a set exposure time of 2 minutes with the cylindrical cell at each orientation. With this second set of tests the PSD threshold for $-90^\circ$ was set to the new PSD threshold that produced the data in table 4.

Table 5. $\gamma$-ray and neutron composition with $^{252}$Cf as a function of angle for 2-minute exposure for the cylindrical cell.

| Angle$^\circ$ | $\gamma$-ray events | Neutron events | $\gamma$-ray/neutron ratio | Total Counts     |
|-------------|----------------------|---------------|---------------------------|------------------|
| 90          | 402935               | 131965        | 3.053 ± 0.010             | 534900 ± 731     |
| 67.5        | 492799               | 164101        | 3.003 ± 0.009             | 656900 ± 810     |
| 45          | 586236               | 203564        | 2.880 ± 0.007             | 789800 ± 889     |
| 22.5        | 625133               | 219367        | 2.850 ± 0.007             | 844500 ± 919     |
| 0           | 696869               | 246531        | 2.827 ± 0.007             | 943400 ± 971     |
| $-22.5$     | 591757               | 209143        | 2.829 ± 0.007             | 809000 ± 895     |
| $-45$       | 455139               | 159761        | 2.849 ± 0.008             | 614900 ± 784     |
| $-67.5$     | 352793               | 124507        | 2.834 ± 0.009             | 477300 ± 691     |
| $-90$       | 245707               | 31993         | 7.680 ± 0.046             | 277700 ± 527     |

3 Analysis

To simplify the extraction of figure-of-merit (FoM) values for each data set, the data collected at each angle were normalized and the baseline removed for each case. This produced a linearized plot of the type shown in figures 4 and 5. The FoM for each case was extracted from histograms by taking the number of counts versus channel, calculated using (3.1).

$$\text{FoM} = \frac{s}{\text{FWHM}_n + \text{FWHM}_\gamma}$$  \hspace{1cm} (3.1)

where $s$ is the separation between the two peaks of the histogram, and FWHM$_n$ & FWHM$_\gamma$ are the full-width-at-half-maximum of the neutron and $\gamma$-ray peaks, respectively. An example histogram of the data the FoM results were obtained from is shown in figure 6, with the corresponding data for both the cylindrical and the cubic detector given in table 6.
The values for the FWHM and the separation were extracted with a double-Gaussian fit to the histogram of the form of (3.2),

$$f(x) = Ae^{\frac{(x-\mu_\gamma)^2}{2\sigma_\gamma^2}} + Be^{\frac{(x-\mu_n)^2}{2\sigma_n^2}}$$

(3.2)

where $\mu_\gamma$ and $\mu_n$ are the means for each distribution, and $\sigma_\gamma$ and $\sigma_n$ are the standard deviations for each distribution, with $A$ and $B$ the constants for each radiation type. The consistency of the fit with the data was calculated in each case and the corresponding reduced chi-squared $\chi^2_\nu$ values included in table 6. The double Gaussian plots for all the cubic and cylindrical detector tests can be seen in figures 7 and 8 respectively.

**Figure 6.** A histogram of the number of counts for the cubic detector for the full energy range exposed for 100100 counts at $0^\circ$, with the model fit over the top.

To further explore the indication that the count rate might be dependent on the orientation of the detector, the results from the 2-minute exposure runs have been plotted against the angle in figure 9.

With the cylindrical detector the poorest values seem to occur at the two smallest angles away from the horizontal. This was followed up by an investigation rotating the detector through the same angles but in the horizontal plane as opposed to the vertical as previously explored, these tests are to justify whether the conclusion of different rate affect can be contributed to the detectors front face not being in line with the source. The hypothesis is depicted in figure 10 with a portion of a sphere representing a uniform radiation field and the cube representing the detector cell. The top two diagrams place the centre of the radiation field at a distance of 150 cm from the first receiving point of the detector, whilst the bottom two diagrams place the centre of the radiation field at a distance of 145 cm.
Table 6. Figure-of-Merit as a function of angle for 100100 counts for $^{252}$Cf for cubic and cylindrical detectors.

| Angle/°  | Figure-of-Merit Cubic | $\chi^2$ | Figure-of-Merit Cylindrical | $\chi^2$ |
|----------|------------------------|---------|----------------------------|---------|
| 90       | 0.814 ± 0.044          | 1.00    | 0.789 ± 0.033              | 0.98    |
| 67.5     | 0.814 ± 0.044          | 0.99    | 0.780 ± 0.031              | 0.97    |
| 45       | 0.814 ± 0.044          | 0.99    | 0.763 ± 0.031              | 0.97    |
| 22.5     | 0.795 ± 0.043          | 0.99    | 0.750 ± 0.030              | 0.97    |
| 0        | 0.795 ± 0.043          | 0.99    | 0.789 ± 0.033              | 0.98    |
| -22.5    | 0.773 ± 0.042          | 0.99    | 0.763 ± 0.031              | 0.97    |
| -45      | 0.791 ± 0.044          | 0.99    | 0.780 ± 0.031              | 0.97    |
| -67.5    | 0.795 ± 0.043          | 0.99    | 0.776 ± 0.032              | 0.97    |
| -90      | 0.837 ± 0.045          | 0.99    | N/A                        | N/A     |

Figure 7. Double Gaussian models for each angle tested using the cubic detector exposed to $^{252}$Cf.

To investigate this hypothesis 2-minute tests were carried out with both detectors rotating whilst keeping parallel with the floor, starting perpendicular to the source through to being face on and continuing to being perpendicular with the source on the other side of the detector. The results from the 2 minute exposures using the cubic and cylindrical cell can be seen in tables 7 and 8 respectively. The results are shown graphically in figure 11.

4 Discussion

It is clear from the measurements presented here that when a detector is horizontal the count rates are greater than at any other angle. Also, the effect of the angle on the count rate is less severe when it is tilted in the positive direction i.e. when the nitrogen void is at the top of the cell than
when in the negative direction and the void is at the interface with the photocathode. This indicates that count rate does depend on the orientation of a detector and the shape of the cell influences the severity of the effect; neither of these phenomena are surprising given the sensitivity of these
Figure 10. Pictorial representation of the uniform radiation field bisecting the detector cell.

Table 7. \(\gamma\)-ray and neutron composition with \(^{252}\text{Cf}\) as a function of angle for 2-minute exposure for the cubic cell rotating away from the source.

| Angle/° | \(\gamma\)-ray events | Neutron events | Total   | FoM    |
|---------|------------------------|----------------|---------|--------|
| 90      | 140774                 | 30926          | 171700  | 0.850 ± 0.005 |
| 67.5    | 143429                 | 31671          | 175100  | 0.84 ± 0.002  |
| 45      | 144327                 | 32273          | 176600  | 0.83 ± 0.002  |
| 22.5    | 149226                 | 33074          | 182300  | 0.84 ± 0.005  |
| 0       | 150998                 | 32902          | 183900  | 0.86 ± 0.005  |
| −22.5   | 147343                 | 32757          | 180100  | 0.86 ± 0.005  |
| −45     | 143426                 | 32174          | 175600  | 0.84 ± 0.002  |
| −67.5   | 143473                 | 31727          | 175200  | 0.830 ± 0.002 |
| −90     | 140493                 | 31307          | 171800  | 0.84 ± 0.001  |

Table 8. \(\gamma\)-ray and neutron composition with \(^{252}\text{Cf}\) as a function of angle for 2-minute exposure for the cylindrical cell rotating away from the source.

| Angle/° | \(\gamma\)-ray events | Neutron events | Total   | FoM    |
|---------|------------------------|----------------|---------|--------|
| 90      | 181330                 | 45070          | 226400  | 0.89 ± 0.002 |
| 67.5    | 190070                 | 48230          | 238300  | 0.88 ± 0.005 |
| 45      | 197790                 | 50910          | 248700  | 0.87 ± 0.002 |
| 22.5    | 216968                 | 57832          | 274800  | 0.89 ± 0.005 |
| 0       | 211458                 | 56042          | 267500  | 0.88 ± 0.004 |
| −22.5   | 202684                 | 53816          | 256500  | 0.87 ± 0.002 |
| −45     | 199656                 | 51244          | 250900  | 0.86 ± 0.004 |
| −67.5   | 191502                 | 47598          | 239100  | 0.86 ± 0.001 |
| −90     | 182114                 | 45286          | 227400  | 0.87 ± 0.003 |
Figure 11. Total counts recorded for the 2-minute exposures of the cubic (black squares) and the cylindrical (grey circles) detector as a function of angle on the horizontal plane.

Instruments. However, these observations do indicate that these effects should be accounted for in large arrays of liquid scintillation detectors where either the angles of the detectors comprising the array and the movement of the fluid contrast with one another. This is qualitatively consistent with the previous report [10] and the hypothesis of restricted light transmission occurring as a result of the position of the expansion volume relative to the optical coupling inside a detector (in the absence of a light guide). However, the change in count rate reported here is of a level of sufficient significance to warrant careful calibration of array systems based on liquids. Where an array is made up of detectors of different model types, the contrast in the severity of the effect needs to be born in mind too.

In terms of the discrimination performance, the ratio of $\gamma$-ray count to that for neutrons is plotted in figure 12 as a function of detector angle for both detectors explored in this research (excluding the $-90^\circ$ position for the cylindrical detector). Whilst this exhibits relatively little dependence with angle, it is worthy of note that given the very high throughput of current real-time PSD systems such as that used in this research, the uncertainties achievable even with a relatively low-activity source can be very low, as in this data. Taking this into account, there does appear to be a degree of dependence in discrimination performance, particularly at the vertical positions of $90^\circ$ and $-90^\circ$, but of a degree of significance lower than the count rate dependence reported in this work.

Taken together, the data presented in this research indicate that where possible low-hazard organic liquid scintillation detectors should be used in a horizontal arrangement to avoid the dependency shedding an influence on measurements made, particularly that associated with count rate. However, perhaps of most significance is that a blind assumption that the discrimination performance is independent of orientation, particularly when manipulating detectors into different
Figure 12. The ratio of γ-ray to neutron counts for 2-minute exposures of the cubic detector (black squares) and the cylindrical detector (grey circles) versus angle (uncertainties smaller than the size of the symbols in each case).

positions could be erroneous as macroscopic shifts in pulse shape can occur requiring a recalibration of PSD threshold settings, as demonstrated in this work. It is clear that dramatic shifts in the PHS observed previously [10] can cause the entire discrimination plain to shift thus requiring revision of PSD threshold settings, in some and yet not all positions as observed in this work. Clearly, it remains best practice to check HV and PSD thresholds every time a detector is used and particularly when any positional adjustment or transport has occurred in between measurements. Where regular portability is required, solid scintillation media ought to be independent of these effects notwithstanding a full angular sensitivity assessment of these media has yet to be reported.

It is worthy of note that there are a wide variety of liquid scintillation media, PSD systems and algorithms of choice for research of this type which have not been explored in this research. This variety is likely to exert a further influence on orientation dependence of count rate and discrimination performance.

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