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The design and performance of a large-volume spherical CsI(Tl) scintillation counter for gamma-ray spectroscopy

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Abstract

This paper presents details of the design and performance of a prototype large-volume scintillation detector used for gamma-ray spectroscopy. In this detector, a spherical CsI(Tl) scintillation crystal having a diameter of 5.7 cm was polished and packed in dry MgO powder. The scintillation light from the crystal was viewed using a single $1 \times 1\,\mathrm{cm}^2$ silicon PIN diode. A low-noise preamplifier was also integrated within the detector housing. The measured noise level was equivalent to ~800 electrons (FWHM). Such a configuration provided a very good light collection efficiency, which resulted in an average of 20 electrons being generated per keV of energy deposited in the crystal. One of the key features of the detector design is that it minimises spatial variations in the light collection efficiency throughout the detector. Compared with a standard 3 in. NaI scintillation counter, this feature leads to a much-improved energy resolution, particularly for photon energies above 1 MeV. The results presented in this paper clearly demonstrate that a spherical CsI-photodiode detector could be used as an ideal replacement for the standard 3 in. NaI detectors in many applications, but especially when the incident gamma-ray spectra extend up to ~10 MeV as in neutron-activated gamma-ray analysis. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Spherical; Scintillation detector; Gamma-ray spectroscopy

1. Introduction

The development of good-quality, silicon PIN diodes having sensitive areas of the order of $1\,\mathrm{cm}^2$, together with the availability of compact, low-noise amplifiers in the early 1980s, led to a great interest then in their possible use in scintillation counter applications. The advantages provided by their rugged construction and ability to operate using only low voltages, stimulated particular interest in their application in Space Science and in portable instruments for which compactness was at a premium. The high scintillation efficiency of CsI(Tl), which peaks at 560 nm, made this material the natural choice of those designing detectors for photon-counting applications using silicon PIN diodes. An energy resolution superior to that of a standard NaI scintillation counter could be achieved above ~662 keV in small detectors using shaping times of typically 5 μs. In

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other applications in which small-area diodes have been used to view linear arrays of crystals in ‘current-mode’ for baggage-scanning applications, the less bright but denser material, cadmium tungstate, has been selected.

Much effort was devoted in trying to optimise the design of small detectors around the available PIN diode types, particularly with the view to construct a large-area position-sensitive gamma-ray detector for use in imaging applications in gamma-ray astronomy [1,2]. For example, the ESA INTEGRAL mission due for launch in 2002, includes an array of 4096 such detectors ($10 \times 10 \times 30\,\mathrm{mm}^3$). In contrast, a recent study has made a thorough examination of the best way to apply CsI-photodiode scintillation counters in the detection of energetic gamma rays up to 12 MeV [3]. Clearly, for some applications, there may be a pressing need to use photodiodes. For example, when there is a need to design a large-area, position-sensitive array for imaging applications. However, these devices have not seriously challenged conventional scintillation detectors in most applications because of their relatively small volume and poor low-energy performance. This paper describes the performance of a novel design of CsI-photodiode scintillation counter for use over the range from a few hundred keV up to 10 MeV. This performance has been compared with that of an industry-standard 3 in. NaI scintillation crystal viewed by a photo-multiplier. A detailed analysis for the difference in the performance of these two detectors is also included.

2. The detector design

This prototype detector uses a 100 ml CsI(Tl) sphere (5.7 cm diameter) that was polished and packed in dry MgO powder. It is viewed using a single $10 \times 10\,\mathrm{mm}^2$ Hamamatsu PIN diode (type S3590). A small preamplifier was integrated within the cylindrical aluminium housing as shown in Fig. 1. This detector was fabricated at the Kurchatov Institute, Moscow and tested at Southampton University.

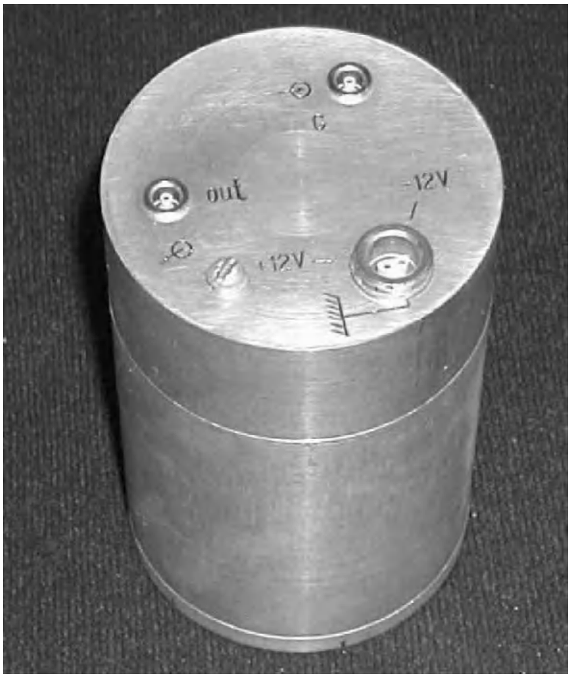

Fig. 1. The prototype $100\,\mathrm{cm}^3$ CsI spherical detector.

It has been shown that the spatial variation of the light collection efficiency combined with the non-uniformity of the photocathode of the PMTs significantly degrades the overall energy resolution of the standard 3 in. NaI/PMT scintillation counters [4]. This is because when the same energy is deposited at different locations within the crystal, different signal amplitudes are detected, particularly when illuminated by high-energy photons. One of the key features of our detector design is that the exit window of the spherical scintillator is only $\sim 1\%$ of its total surface area. As a consequence, most of the detected photons will have undergone a large number of reflections before reaching the PIN diode. This minimises the spatial variations in the light collection efficiency and hence improves the overall energy resolution of the detector. Although the size of the exit aperture is only a small proportion of the total surface area of the sphere, a very good light collection efficiency ($>40\%$) was achieved using such a detector design. Another design consideration is that the use of the PIN diode introduces extra electronic noise. This detector design enables one to reduce the noise contribution by using a relatively small PIN diode whilst at the same time maintaining a good light collection efficiency. In order to demonstrate the superior performance of this new detector, some energy-loss spectra were compared with those recorded using a standard 3 in. NaI scintillation counter (Scionix Type: 76B76/3).

3. Results

3.1. Comparison with a 3 in. NaI detector

The 100 cm^3 CsI detector and a standard 3 in. NaI scintillation counter were exposed to a number of calibration sources including a ^{238}Pu–^{13}C high-energy gamma-ray calibration source, which emits 6129 keV gamma rays from ^{16}O [5]. A comparison between the measured energy-loss spectra using these two detectors is shown in Fig. 2. It is clearly demonstrated that at gamma-ray energies above 1 MeV, the CsI-photodiode detector provided a greatly improved energy resolution compared with the 3 in. NaI detector. At lower energies, the energy resolution of the CsI detector becomes progressively dominated by electronic noise. This results in a crossover point at around 500 keV. Below this energy, the CsI detector has a poorer energy resolution than the standard 3 in. NaI detector. This relatively poor low-energy performance will surely be a limitation of such a CsI detector for some applications. The second generation of these SCINTISPHERE® detectors will be equipped with a choice of large-area avalanche photo-diode or a special photomultiplier in addition to the PIN diode. This should significantly improve the energy resolution, particularly for lower photon energies.

3.2. Signal calibration

The signal generated by this CsI detector was calibrated directly in terms of the number of electrons by illuminating the PIN diode with an Am-241 source. The energy-loss spectrum, shown in Fig. 3, includes both the scintillation signals and the peak resulting from the direct interaction of the 59 keV photons in the silicon diode. By relating this known photon energy to the mean energy required to produce a charge-carrier in the silicon (3.67 eV), the number of electron–hole pairs generated by a unit photon-energy deposited in the scintillation crystal was estimated to be

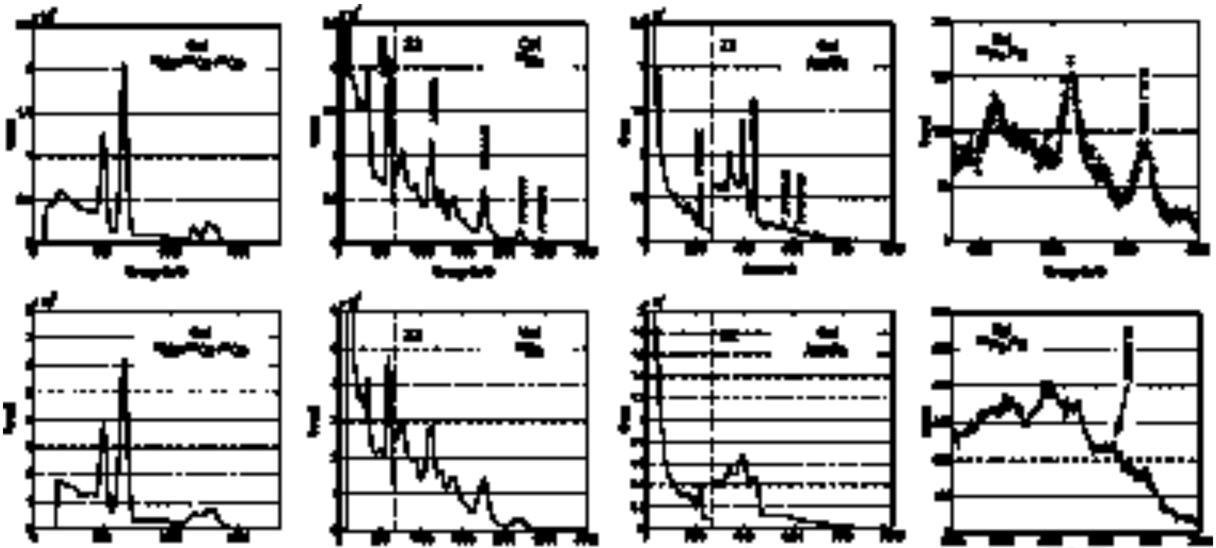

Fig. 2. The comparison between the spectra observed using the CsI detector and the 3 in. NaI detector.

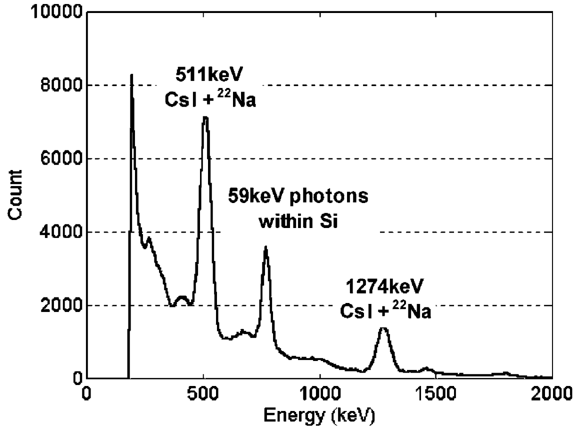

Fig. 3. The spectrum measured using a ^{241}Am point source to irradiate the Si diode. In order to calibrate the number of electron/hole pairs generated, a ^{22}Na source was also used to generate two reference peaks.

$20\,e^{-}h$/keV. This value corresponds to around 13,000 electrons for a 662 keV energy deposit in the crystal. One important indication from this result is that after taking into account the quantum efficiency of the PIN photodiode (~85%), the light collection efficiency achieved was found to be as high as 40% even though the exit aperture was only 1% of the total crystal surface area. This result also indicates that the reflection coefficient of the crystal surface treatment applied to this detector is greater than 99%.

3.3. Equivalent noise charge (ENC)

The spectral broadening caused by the electronic noise in the readout is a combination of the fluctuation in the photo-diode leakage current and the pre-amplifier noise. This can also be measured directly using low-energy gamma-ray sources to irradiate the PIN-diode as in the signal calibration measurements. Since the contribution due to the statistical fluctuations in the number of electron–hole pairs can be determined experimentally, the ENC was calculated using the following equation:

$$\mathrm{ENC}_{(\mathrm{FWHM})} = \sqrt{\mathrm{FWHM}(^{241}\mathrm{Am})^2 - \left(\frac{2.354}{\sqrt{N_{\mathrm{e/h}}}}\right)^2} \tag{1}$$

where the $N_{\mathrm{e/h}}$ is the number of electron/hole pairs generated in silicon by a 59 keV gamma ray from a ^{241}Am source and the FWHM(^{241}Am) is the measured FWHM of the 59 keV photopeak (as shown in Fig. 3). Such a measurement gave an ENC of around 800 electrons (FWHM), which results in an Equivalent Noise Energy (ENE) of ~35 keV (FWHM).

Although the use of larger area, or multiple photo-diodes may provide a better light collection efficiency, it would introduce a higher level of electronic noise. For an optimum energy resolution, a compromise must be made between the light collection efficiency and the noise contribution.

3.4. Spatial variation of the signal amplitude

As early as 1954, detailed surveys were made of the spatial variations in the quantum efficiency of photomultipliers [6]. The conclusion reached then was that significant improvements could be achieved in the performance of gamma-ray spectrometers if a crystal-mounting system could be devised to couple the scintillator just to the most sensitive region of the tube. There has been no indication that later experimenters felt that such a task was either feasible or sufficiently worthwhile. Later work by Fraser-Mitchell and Wright [7] combined several observed patterns of the non-uniformity in the photo-cathode response of a number of photo-multiplier tubes in a Monte Carlo simulation of the spectral resolution of a 3 in. NaI scintillation counter. An illustration of the variation in photo-cathode sensitivity across the photo-cathode of a 3 in. photo-multiplier, kindly supplied by Electron Tubes Ltd., is shown in Fig. 4. We do not have similar information about the variation in quantum efficiency in the particular 3 in. NaI scintillation counter used nor the photodiode attached to the sphere. In the latter case, we have assumed that any such variations are negligible.

In a 3 in. NaI(Tl) detector, the distribution of the scintillation light reaching the photo-cathode is different for interactions which occur at different locations within the crystal. This effect, combined with the photo-cathode variations, spoils the

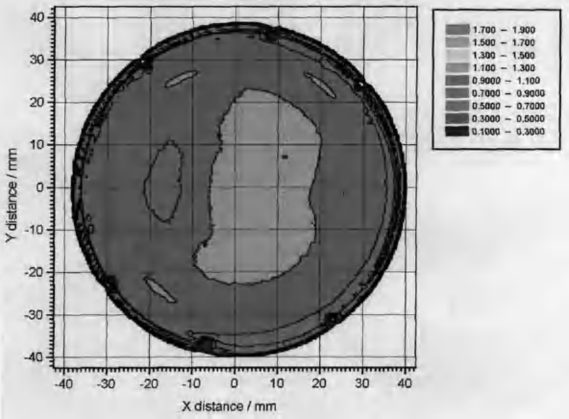

Fig. 4. A typical cathode uniformity contour.

Table 1
Photo-peak positions as a function of the location irradiated[a]

	0°	60°	120°	180°	240°	300°
CsI/PD	365	366	366	366	366	366
NaI/PMT	143	140	139	143	145	144

[a] Measured using a collimated 511 keV gamma-ray beam intersecting both detectors on their sides.

overall energy resolution of the detector. For the spherical CsI detector, since the exit aperture is only a small fraction of the total surface area, the chance of a scintillation photon reaching the exit aperture without reflection is very small. A simple optical Monte Carlo simulation indicated that most of the photons undergo more than 50 reflections before reaching the photodiode. This effect reduces the non-uniformity of the light collection and therefore effectively reduces its contribution to the overall energy resolution.

In order to demonstrate this improvement, a series of measurements have been carried out. A ^{22}Na point source was collimated within a plane perpendicular to the detector-axis and passing through the geometrical centre of the scintillator. The beam was directed so as to intersect the detector at different points around the circumference. This ensured that only a small volume at the edge of the detector was irradiated. Whilst keeping the source and collimator fixed, the detector was rotated about its axis in 60° intervals. From the observed spectra, the position of the 511 keV photo-peak was measured by fitting the full-energy peaks to a Gaussian distribution. For comparison, a similar scan was made using the 3 in. NaI detector. The measured photo-peak positions for both detectors are given, in terms of channel numbers, in Table 1. A second measurement was made using an 81 keV gamma-ray beam, from a collimated ^{133}Ba point source, parallel to the detector axis. Both detectors were irradiated on their front surface along a diameter. Again, the positions of the photo-peaks were derived and listed in Table 2.

Although these results may not reflect all aspects of the spatial variation of the signal amplitude in these two detectors, they provided some important indications about the difference between them. For the 3 in. NaI detector, the measured signal amplitude varied significantly for interactions at different locations in the crystal. When extended sources are used, this effect may contribute as much as an extra 5% of broadening to the overall energy resolution. Furthermore, this contribution may be even more significant for gamma rays having higher energies since their higher penetration capability leads to more interactions occurring close to the photo-cathode. Consequently, photo-cathode non-uniformities will lead to an even greater variation in the measured signal amplitudes. In contrast, the spherical CsI detector showed a much smaller variation. As we will show later in this paper, this improvement significantly reduced the spectral broadening especially at relatively high photon energies.

3.5. Intrinsic energy resolution

It has been shown that the scintillation efficiency (photons/MeV) of most scintillation materials, varies with electron energy, especially between a few keV and several hundred keV [8]. When these scintillation materials are used in gamma-ray spectrometers, this non-proportionality usually leads to a significant spectral broadening, which is intrinsic to the material used (*intrinsic energy resolution*). In this study, we attempted to estimate the intrinsic energy resolution of CsI(Tl) and NaI(Tl) scintillators as a function of gamma-ray

Table 2
Photo-peak positions as a function of the location irradiated[a]

	−3.5 cm	−2.5 cm	−1.5 cm	−0.5 cm	0.5 cm	1.5 cm	2.5 cm	3.5 cm
CsI/PD	—	371	371	371	371	372	371	—
NaI/PMT	144	145	144	143	139	138	137	135

[a] Measured using an 81 keV gamma-ray beam (from a 133 Ba source) irradiating the top surfaces of both detectors.

Table 3
Energy resolutions for the 100 cm^3 CsI detector

Energy	R_{noise}[a] (%)	$R_{\text{statistic}}$[b] (%)	R_{LCE} (%)	$R_{\text{intrinsic}}$ (%)	R_{overall} (%) (estimated)	R_{overall} (%) (measured)
100 keV	42	5.3	1.5	4.0	42.5	
150 keV	28	4.3	1.5	3.6	28.4	
200 keV	21	3.7	1.5	3.2	21.6	
250 keV	16.8	3.3	1.5	4.5	17.8	
300 keV	14	3.0	1.5	5.5	15.4	
356 keV	**12**	**2.8**	**1.5**	**5.3**	**13.5**	**13.4**
400 keV	10.5	2.6	1.5	5.2	12.1	
450 keV	9.3	2.5	1.5	4.9	10.9	
511 keV	**8.2**	**2.3**	**1.5**	**4.4**	**9.7**	**9.5**
662 keV	**6.3**	**2.0**	**1.5**	**3.8**	**7.7**	**7.7**
1.274 keV	**3.3**	**1.4**	**1.5**	**2.2**	**4.5**	**4.7**
2.22 MeV	**1.9**	**1.1**	**1.5**	**1.45**	**3.0**	**2.7–3.2**
4 MeV	1.05	0.83	1.5	1.2	2.3	
6.13 MeV	**0.7**	**0.68**	**1.5**	**0.97**	**2.0**	**2.0**
8 MeV	0.53	0.59	1.5	0.76	1.86	

[a] The value of the noise contribution was measured by irradiating the PD with Am241 source. The measured value for the noise equivalent charge (NEC) was ~800 electrons.

[b] The Gaussian contribution was estimated by assuming 20 electrons/keV.

energy. A number of recent experimental studies have quantified the light-output of a many scintillation materials when excited by electrons and photons of different energies. For example, work by Valentine and Ronney who used a Compton-coincidence technique [9] has shown that the relative light-output per keV of many common scintillation materials varies by as much as 15% over the electron energy range from 10 to 1000 keV. However, a paper by Leutz and D'Ambrosio [10] disputes the magnitude of this non-linearity in NaI detectors.

In order to derive the intrinsic energy resolution for these materials, we chose to use the data from Valentine et al. These data were used in conjunction with a GEANT Monte Carlo simulation to deduce the intrinsic energy resolution as a function of photon energy for detectors based on both CsI(Tl) and NaI(Tl) crystals. The effective light-yield generated by an incident photon was estimated by deriving the various contributions made by each secondary electron generated by the interactions of the photon within the crystal, based on the effective light-yield versus electron energy curve given in Ref. [11]. Note that in order to extend the estimation of the intrinsic energy resolution to photon energies above 10 MeV, we assumed that the effective light-yield per unit energy loss for electrons with energies above 1 MeV is a constant. A similar method has been used by Valentine et al in predicting the intrinsic energy resolution at a particular photon energy. In this work, a relatively good agreement was achieved between the measured and predicted values for the resolution. In our work, we extended the modelling to a number of energies, ranging from 100 keV to 10 MeV. The results of this study are shown in Table 3.

3.6. Energy resolution as a function of energy

The overall energy resolution of the CsI/PD detector and the factors which contribute to this value, can be related as follows:

$$R_{\mathrm{overall}} = \sqrt{R^2_{\mathrm{statistic}} + R^2_{\mathrm{noise}} + R^2_{\mathrm{intrinsic}} + R^2_{\mathrm{LCE}}} \tag{2}$$

where the $R_{\mathrm{statistic}}$ is the contribution from the statistical fluctuation on the number of the charge-carriers generated; R_{noise} is the contribution from the electronic noise; $R_{\mathrm{intrinsic}}$ is intrinsic energy resolution and R_{LCE} is the contribution from the spatial variation of the light collection in the crystal. Since $R_{\mathrm{statistic}}$, R_{noise} and $R_{\mathrm{intrinsic}}$ can be determined either experimentally or through the use of Monte Carlo simulations, we determined the contribution of R_{LCE} by fitting the above equation to a number of measured data points. This indicated that the maximum value for R_{LCE} was around 1.5%. The estimated energy resolution as a function of energy is shown in Table 3 along with several measured values. Clearly, the estimated energy resolutions match the measured ones very well over a wide energy range.

For the 3 in. NaI detector, the overall energy resolution may be given as follow:

$$R_{\mathrm{overall}} = \sqrt{R^2_{\mathrm{statistic}} + R^2_{\mathrm{intrinsic}} + R^2_{\mathrm{transfer}}} \tag{3}$$

where the R_{transfer} is the contribution from the variable probability that a visible photon, generated by a scintillation event in the crystal, will produce a photo-electron that is collected by the first dynode of the PM tube [12]. As was indicated in Section III-D, it is difficult to quantify the term R_{transfer}, since it depends on several unknown factors such as the non-uniformity of the PMT photo-cathode and the actual distribution of the gamma-ray interactions within the crystal. If we assume that an average of 9000 photo-electrons are generated by a 662 keV full-energy event, a similar fitting process revealed that the R_{transfer} term introduces an extra 5% or more, to the overall energy resolution.

A comparison between the CsI and NaI detectors is shown in Fig. 5. In this comparison, we have assumed that the R_{transfer} has a constant value of 5% over the entire energy range. Since the same axis scale is used in both plots, one may easily compare the individual components as well as the overall energy resolution. Clearly, the key to the performance difference between the two detectors is that the unique configuration of the CsI detector that provides a good light collection efficiency with excellent uniformity.

3.7. Improvement in spectrum deconvolution

Our previous study has shown that the spectral quality of a scintillation detector can be improved

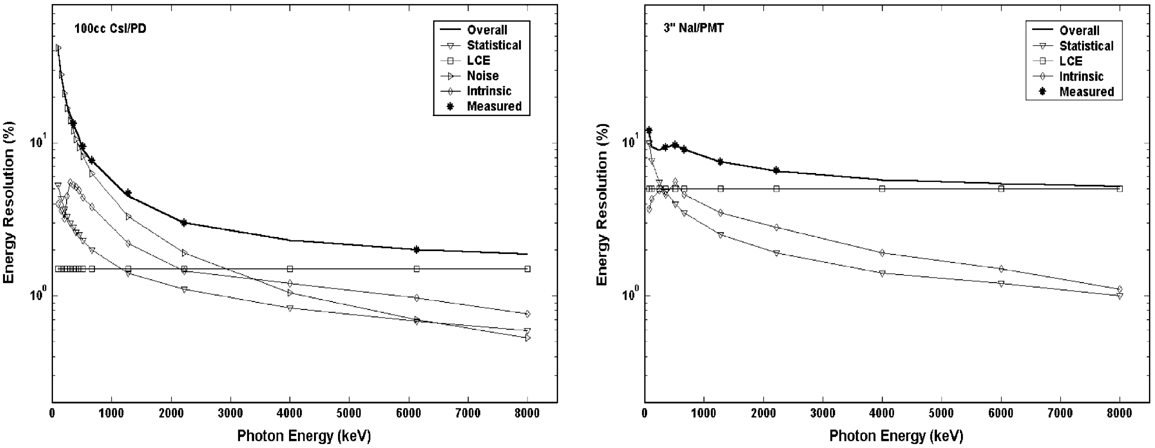

Fig. 5. A comparison of the energy resolution as a function of energy for the CsI detector and a 3 in. NaI detector.

significantly using a post-processing method such as spectrum deconvolution [13]. The accuracy of this process depends crucially on two factors; namely, the degeneracy of the detector response function (DRF) and ones ability to predict the detector's response to the incident gamma rays. The results presented above have demonstrated that the spherical detector design significantly reduces the variance in the light collection whilst providing a good light collection efficiency. Compared with standard 3 in. NaI detector, this results in a much-improved energy resolution and therefore a reduced degeneracy in the DRF. Secondly, by minimising the spatial variation in the light collection process, the DRF of CsI detector can be modelled very precisely. This enables one to predict the incident gamma-ray spectra more accurately using the measured energy-loss spectra. This improvement is illustrated in Fig. 6, in which the deconvolved spectra from these two detectors using a ^{226}Ra point source and a neutron activation gamma ray source, may be compared. Clearly, more spectral details are resolved in the deconvolved CsI spectrum. An energy resolution of around 1.4% at 662 keV has been observed in a deconvolved CsI spectrum, whilst our best attempt only produced an energy resolution of around 3% using the 3 in. NaI detector.

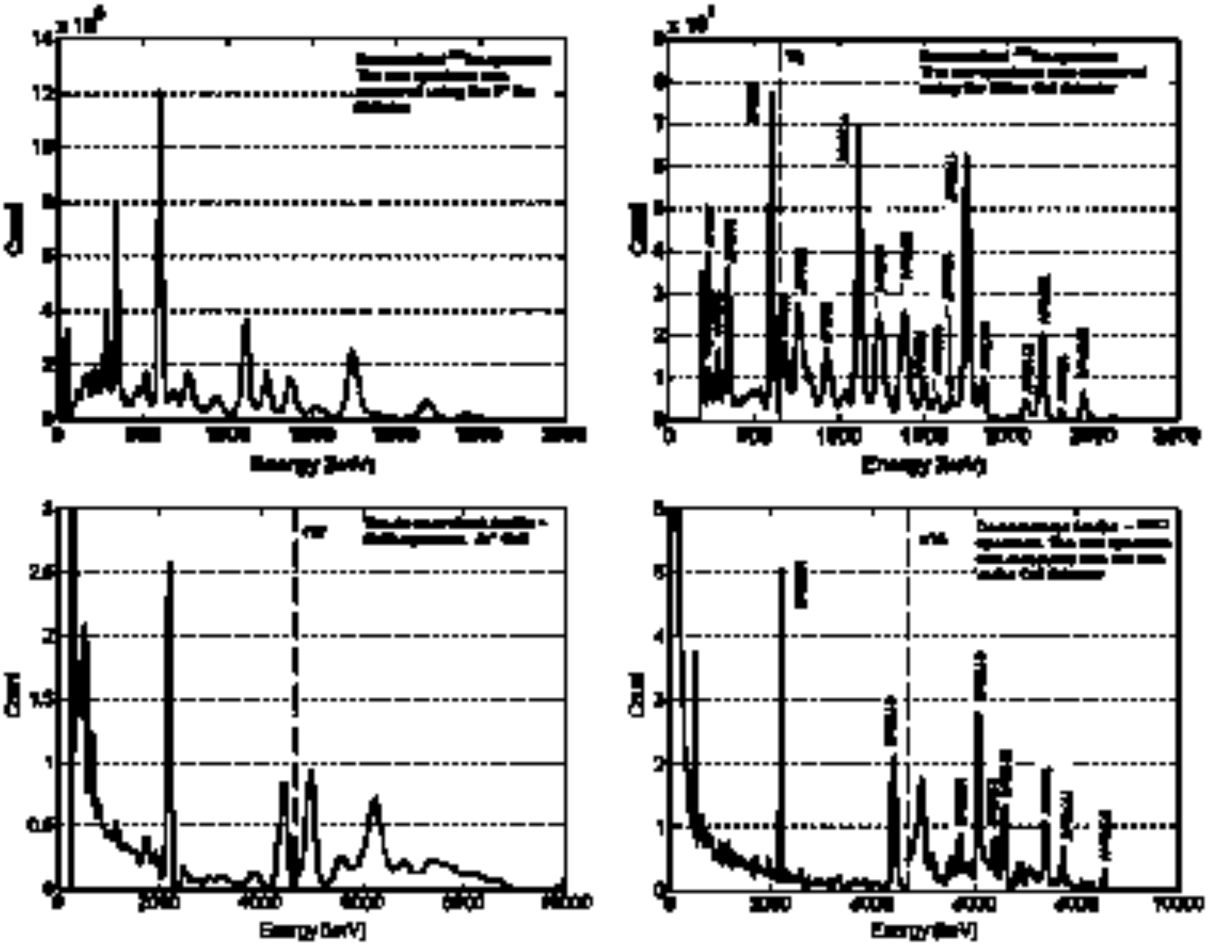

Fig. 6. Comparison of the deconvolved spectra based on the 3 in. NaI detector and 100 cm^3 CsI detectors. The spectra shown on the bottom two panels are based on the use of a water-moderated Am/Be neutron source.

4. Conclusions and discussions

This paper described the design and performance of a large-volume spherical scintillation detector. The results presented have clearly demonstrated the benefits achieved using this unique detector design. A number of conclusions drawn from this study are listed below:

- The spherical crystal enables one to use a relatively small photo-detector, whilst still providing a good light-collection efficiency. This feature helps to provide an optimum compromise between the light collection efficiency and readout electronic noise.
- The use of a small, uniform PIN diode, minimises the contribution made by spatial variations in the light-collection efficiency to the overall spectral broadening.
- Compared with a standard 3 in. NaI detector, this design results in a more "predictable" detector, in the sense that an accurate DRF can be calculated. This improves the accuracy of the post-processing algorithms.
- The main limitation in the current detector design is that the electronic noise introduced by the photo-diode readout severely degrades its performance below 500 keV. This can be overcome by the use of either a large area avalanche photo-diode or a photo-multiplier.

Clearly, given the particular scintillation material and the volume required, the spherical detector design [14] provides a near-optimum solution for gamma-ray scintillation spectrometer. A range of detectors based on this SCINTISPHERE® concept are currently under development, using APD and PMT readouts. These detectors, combined with post-processing methods, should provide attractive alternatives to the standard 3 in. NaI detectors and HPGe detectors in a variety of applications.

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