Scintillators and Cherenkov detectors for the registration of 10.8 MeV gamma rays

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Scintillators and Cherenkov detectors for the registration of 10.8 MeV gamma rays

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Abstract. The identification of nitrogen by neutron activation has been utilized in both explosive detection and in-vivo metabolic analysis. The 10.8 MeV gamma ray line emitted by thermal neutron capture provides a unique signature, however, due to its high energy its registration is non-trivial. Conventional approaches have used large dense inorganic scintillators which inevitably entail considerable expense. We examine the capabilities of arrays of smaller scintillation detectors and the use of glass Cherenkov detectors as an alternative.

1. Introduction

High energy gamma rays are produced as a result of neutron activation of materials. There is particular interest in detecting the 10.8 MeV gamma-ray line produced as a result of thermal capture on nitrogen. The cross-section for the capture reaction on nitrogen is $\sigma_{\gamma,\mathrm{all}} = 79.0$mb while the component generating 10.8 MeV gamma-rays is $\sigma_{\gamma,10.8} = 11.3$mb. This reaction has the potential for use as an indicator of the presence of explosives, since all commercial and most improvised explosives are rich in nitrogen [1]. There has also been some work on in-vivo nitrogen/carbon ratios as a diagnostic of human metabolic disorders [2].

Few elements, and certainly no other common element, has significant gamma emission at 10.8 MeV or higher [2]. While the detection of these high energy gamma-rays offers a unique signature, actual detection is more problematic, due to the size and density of detectors required for total absorption. The task is made more difficult by the fact that the gamma detectors are inevitably operated in a mixed field also containing neutrons of all energies from production down to thermal.

High-purity germanium (HPGe) detectors are ill-suited to the task, having low efficiency and requiring cryogenic cooling. Germanium suffers from activation problems [3], while the expense of HPGe and other semiconductor detectors rules them out for deployment in field deployed explosive detector systems.

Conventional wisdom is to use large dense inorganic scintillators, and the obvious choice is sodium iodide (NaI(Tl)). Data from Saint-Gobain [4] indicate that at 10.8 MeV, 50mm NaI(Tl) crystals have an absorption efficiency of 50% and that 120mm crystals have an efficiency of around 80%. The studies by Stamatelatos et al.[3], McFee et al.[5] and Gierlik et al.[6] show that there is little to be gained by using recent dense inorganic scintillators such as bismuth germanate (BGO) or lanthanum bromide ($LaBr_3$). These materials have activation problems

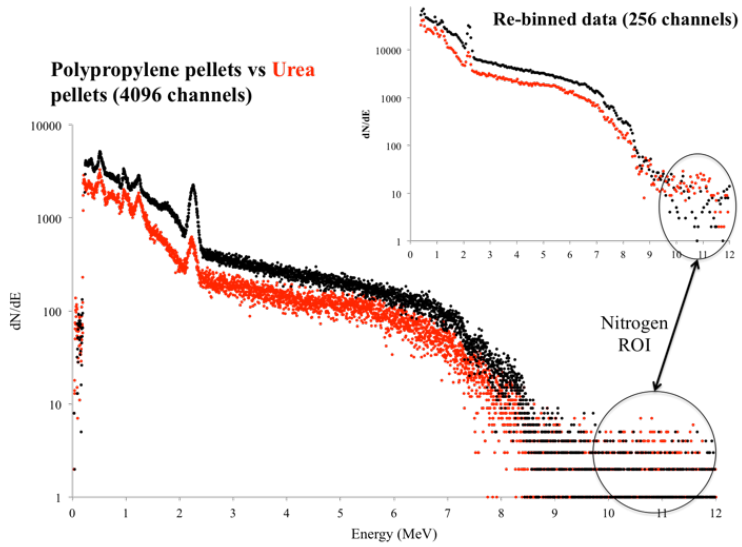

Figure 1. Gamma spectrum produced on 10kg urea sample in 300 minutes.

and the improvements in detection efficiency are outweighed by the huge increase in cost over NaI(Tl). Overall detection efficiency can be improved more cost-effectively by deploying a larger area of NaI(Tl).

Stamatelatos et al.[3] emphasize that the measurement is a signal-to-noise problem and that reducing background is more important than increasing signal. They stress that, in the environment of a neutron source, it is necessary to shield the detectors adequately from both gamma-rays coming from directions other than the target and from neutrons. They suggest the use of bismuth for the gamma shielding as it activates to a much lesser extent than lead and the resulting activation products are not gamma emitters. Further they suggest a layered shielding approach with both neutron and gamma absorbing materials. Closest to the crystal, they used a layer of resin loaded with ^{6}LiF, to absorb thermal neutrons without the emission of gamma-rays. ^{6}LiF is now too expensive to be considered for shielding, but we have found that a 10mm layer of a 40/60 mixture of wax and unenriched lithium carbonate intended for ceramic glazes is similarly effective at suppressing thermal neutrons.

Stamatelatos et al.[3] also note that significant background is caused by pulse pile-up in which two lower energy gamma-rays interact in the crystal within the decay time of 250ns. They point out that this can be reduced by using a number of crystals 50mm diameter by 100mm long, each with its own readout, rather than a large single crystal.

2. Measurements with DT generator

Using an NSD-Fusion 14 MeV DT neutron generator and pelleted urea as an explosives surrogate, we have demonstrated this technique [7]. The emitted gamma-rays were recorded with a single $4'' \times 4''$ NaI crystal fed to a pulse height analyzer. The crystal was lead shielded in all directions but the forward. The pulses were time gated in order to be optimally sensitive to activation caused by thermal neutrons. Typically three hour exposures were employed on 10kg urea samples which provided a sufficiently detailed spectrum to understand the processes.

A typical observed spectrum is shown in figure 1. The background caused by pulse pile-up is particularly severe in the region 4–7 MeV. By subtracting the background polypropylene spectrum from the urea spectrum, the 10.8 MeV emission can clearly be seen as in figure 2. Despite the pulse pile-up and broadening which it causes, a statistically significant peak could

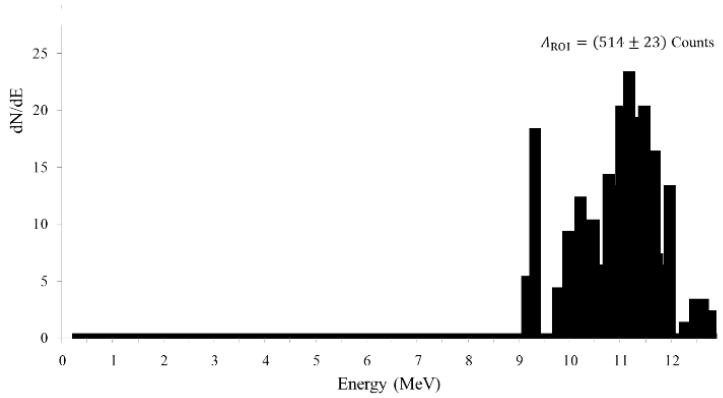

Figure 2. 10.8MeV peak obtained from 10kg urea sample in 300 minutes

be obtained in around ten minutes. Further details of these measurements can be found in Cole [7].

3. Cherenkov detectors

To achieve similar results in shorter time, it is clear that a larger area of detector must be used. As arrays of NaI(Tl) scintillators are likely to remain expensive, it was decided to investigate whether much cheaper Cherenkov detectors could be used.

Cherenkov detectors with dense radiators have found extensive use as charged particle detectors but are also useful for gamma-ray detection where fast timing is required and where good energy resolution is not important. Sowerby [8] examined acrylic Cherenkov detectors for use over the range 1.1–2.6 MeV and Sober et al. [9] consider lead-glass detectors of 153mm depth for the region 10-20 MeV. They show there is a threshold below which no signal is produced, while at 10.8 MeV the efficiency can be expected to be around 60%. Grannis et al. [10] give indications regarding low-cost production of these detectors. Ayaz-Maierhafer et al. [11] have recently demonstrated the utility of glass Cherenkov detectors in the detection of fission gammas. Ayaz-Maierhafer et al. [12] have also performed experimental measurements on neutron activation of Cherenkov glasses which suggest that their long term operation in mixed neutron-gamma fields is not likely to be problematic. Hayward et al. [13] provide a detailed simulation of the response of glass Cherenkov detectors to 6 MeV gamma rays.

It is evident from these studies, that 10.8 MeV is at the lower end of the useful energy range of Cherenkov detectors. However, they offer large size, high density and consequently high efficiency at low cost. Lead-glass is inherently cheaper than NaI(Tl), as an indication, Schott F2 glass was quoted as £56/kg [14]. Being non-hygroscopic, no special precautions are needed in construction and the resulting detectors are robust. Their time response is limited by the risetime of the photomultiplier used and is typically 10ns, so problems with pulse pile-up are much reduced.

The obvious disadvantage of Cherenkov detectors derives from the low light output and the subsequent poor energy resolution at 10.8MeV. In the absence of distinguishable peaks in the output spectra, the calibration of Cherenkov detectors is also problematic. This is compounded by the unavailability of sources with suitable high-energy emission. Calibration has been considered in detail by Rose and Erickson [15].

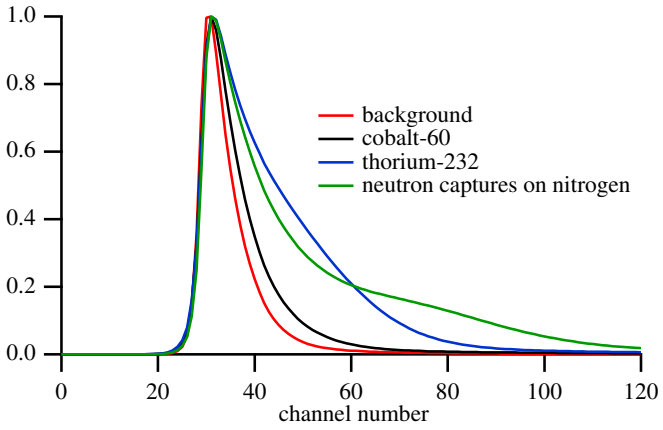

Figure 3. Normalized pulse height spectra obtained from lead-glass detector

3.1. Experimental detector

To test these ideas, an experimental lead-glass Cherenkov detector was constructed. A block of Schott F2 lead-glass 80mm×80mm×80mm was obtained. The surfaces of this were shot-blasted, with the exception of a circular output window on one face. The block was then painted with Edmund 53-697 $BaSO_4$ high reflectance paint, which provides good diffuse reflectance, even in the ultraviolet. The block was then coupled to an ET9265K 70mm diameter photomultiplier using a transparent silicone pad. The whole assembly was made light-tight with aluminium foil and black tape.

The resulting detector worked largely as expected. The 9265K has a rise-time of 7.5ns, so the output pulses are around 15ns FWHM. Figure 3 shows normalized pulse height spectra obtained in response to different sources. There is clearly an offset of some 20 channels on the x-axis, which should be ignored.

The spectrum taken with ^{60}Co, having peaks at 1.1 and 1.3 MeV shows little difference from background. The ^{232}Th spectrum was taken with an old sample of ThO, so had emissions from all the progeny, of which ^{208}Tl is the most important as it decays with a major branch emitting a gamma of 2.62 MeV. The neutron captures on nitrogen spectrum was obtained by placing a few kilograms of urea in front of the detector and exposing it to thermal neutrons obtained from an AmBe source in a mass of polypropylene moderator. It can be seen that by channel 60, background and low-energy gammas are negligible. Any events registered with a height greater than channel 75 are interesting and indicative of high-energy gamma emission.

Further to the spectra shown, there was a population of much larger pulses, with a peak at around channel 1200 and a high energy tail. The peak was attributed to single cosmic muons traversing the block, while the higher energy events are probably showers and residual gas events in the photomultiplier. As the rate of these events was many orders of magnitude less than that expected in a screening scenario, they can be ignored.

These results are broadly similar to those obtained by Sober et al. [9] and Rose and Erickson [15]. While detailed measurements have not yet been made, it appears that our experimental detector has an electron detection threshold at ∼138keV and at 10.8 MeV, a detection efficiency of ∼50% and an energy resolution of ∼50%.

4. Detector arrays

The object of the proposed measurements is not to produce a detailed emission spectrum of the sample being irradiated, but simply to establish whether the emission is consistent with the presence of emission at 10.8 MeV. Consequently the analysis of peaks in regions-of-interest can be replaced with the simple registration of Cherenkov signals exceeding a threshold.

A more convincing result may be obtained by combining the signals from a small area of NaI(Tl) detectors with a larger area of Cherenkov detectors. An array containing a mixture of the two types of detectors is envisaged. In the presence of 10.8 MeV emission the NaI(Tl) would record a small number in the relevant peak region-of-interest, while the Cherenkov detectors would record a large number of high-energy events. The most obvious conclusion would be that the Cherenkov events have the same energy distribution as those recorded by the NaI(Tl) detector. The presence of high-energy Cherenkov events with no signal in the NaI(Tl) region of interest would be indicative of neutron activation of some other element than nitrogen.

5. Combining datasets

The combination of data from detectors with different resolutions and efficiencies and the determination of the presence of 10.8 MeV gamma emission can be examined using objective Bayesian analysis [16, 17]. An algorithm using Bayes Factors calculated from count rates over specified periods for the two types of detector is proposed. In the case of the NaI(Tl) detector there is a region of interest (ROI) which includes the 10.8 MeV peak, while in the case of the Cherenkov detector, the high-energy region of interest is everything above a discrimination threshold. The Bayes factors B_{01} are calculated as per equation 3.1 of [17]. Similarly the threshold is set as a Bayesian z-value as per equation 3.3 of [17].

Record background for a certain time t_{off}:
- Record background with NaI detector,
  - number of events in the 10.8MeV RoI is $N_{off,\mathrm{NaI}}$
- Record background with Cherenkov detector,
  - number of events in the high-energy RoI is $N_{off,\mathrm{Che}}$

Record sample for a certain time t_{on}:
- Record sample with NaI detector,
  - number of events in the 10.8MeV RoI is $N_{on,\mathrm{NaI}}$
- Record sample with Cherenkov detector,
  - number of events in the high-energy RoI is $N_{on,\mathrm{Che}}$

Calculate individual Bayes Factors that events are due to background only:
- Calculate Bayes Factor for NaI detector with:
  - $B_{01,\mathrm{NaI}}(N_{on,\mathrm{NaI}}, N_{off,\mathrm{NaI}}, t_{on}/t_{off})$
- Calculate Bayes Factor for Cherenkov detector with:
  - $B_{01,\mathrm{Che}}(N_{on,\mathrm{Che}}, N_{off,\mathrm{Che}}, t_{on}/t_{off})$

As Bayes Factors are commutative, the experiments can be performed in any order.
The new odds result from previous odds by multiplication:

Resulting odds of the background (H_0) over signal (H_1) hypothesis:
- $P(H_0)/P(H_1) = B_{01,\mathrm{NaI}} * B_{01,\mathrm{Che}}$

Set a threshold. If $P(H_0)/P(H_1) < 5.7 \times 10^{-7}$ then there is 5σ detection

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6. Current status

The shielding of both the NaI(Tl) and Cherenkov detector needs improvement and more detailed calibration runs of the Cherenkov detector are planned. The Bayesian algorithm is being tested with simulated data. If the initial experiments look fruitful, a larger detector array will be built.

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