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Further development of a cosmic veto gamma-spectrometer

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Abstract The Comprehensive Nuclear-Test-Ban Treaty (CTBT) is supported by a network of certified laboratories that perform high-resolution gamma-spectrometry on global air filter samples for the identification of 85 radionuclides. At the UK CTBT Radionuclide Laboratory (GBL15), a novel cosmic veto gamma-spectrometer has been developed to improve the sensitivity of measurements for treaty compliance. The system consists of plastic scintillation plates operated in time-stamp mode to detect coincident cosmic-ray interactions within an HPGe gamma-spectrometer. This provides a mean background reduction of 75.2 % with MDA improvements of 45.6 %. The CTBT requirement for a ^{140}Ba MDA is achievable after 1.5 days counting compared to 5–7 days using conventional systems. The system does not require dedicated coincidence electronics, and remains easily configurable with dual acquisition of unsuppressed and suppressed spectra. Performance has been significantly improved by complete processing of the cosmic-ray spectrum (0–25 MeV) combined with the Canberra Lynx$^{\mathrm{TM}}$ multi-channel analyser. The improved sensitivity has been demonstrated for a CTBT air filter sample collected after the Fukushima incident.

Keywords Gamma-spectrometry · Cosmic veto · List mode

Introduction

The Comprehensive Nuclear-Test-Ban Treaty (CTBT) bans any nuclear explosions, for military or civil purposes [1]. Until treaty ratification, a verification process is being undertaken for the establishment of a robust international monitoring system (IMS) for treaty compliance. This consists of a network of certified laboratories that perform high-resolution gamma-spectrometry on air filter samples collected from radionuclide monitoring stations distributed at 80 global locations. Measurement is undertaken for 85 radionuclides indicative of nuclear weapons tests and reactor incidents including ^{140}Ba, ^{95}Zr, ^{99}Mo, ^{141}Ce, ^{147}Nd, ^{131}I, ^{134}Cs and ^{137}Cs [2]. At the UK CTBT Radionuclide Laboratory (GBL15) situated at the Atomic Weapons Establishment (AWE), advanced gamma-spectrometry systems have been developed to improve the sensitivity of CTBT measurements [3–5]. This includes a novel cosmic veto device operated in time-stamp mode to detect coincident muon interactions within an HPGe gamma-spectrometer [3]. The system utilised a widely available Canberra Lynx$^{\mathrm{TM}}$ multichannel analyser (MCA) to provide comprehensive logging of all detector events with 100–200 ns time resolution [4]. Post-processing was applied to eliminate coincident events and provide a mean background reduction of 54.5 % with improved sensitivity for CTBT radionuclides by a mean of 29.5 %. Although this initial system demonstrated the potential for cosmic background reduction, the requirement for individual Lynx MCAs to control each plate and HPGe made the system unnecessary complex and expensive. This research discusses an improved design, whereby a single Lynx MCA controls all plates and operates in conjunction with the Lynx MCA controlling the HPGe. The simplified design is considered more suitable for implementation in radionuclide laboratories and monitoring stations.

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The cosmic veto gamma-spectrometer has been enhanced to consider cosmic-ray induced background within the range of 0–25 MeV. The earlier system was limited to muon interactions at ~10 MeV, with the predominantly terrestrial component (0–3 MeV) being removed due to the difficulties of processing high count rate time-stamp data. More advanced software now allows full processing of the cosmic-ray spectrum. This includes contributions from: (1) prompt continuously distributed background from charged particles penetrating the detector and producing a continuous background in coincidence with the primary particle; these particles also produce secondary particle showers (p, e^-, e^+) primarily in the detector shielding with bremsstrahlung and annihilation lines that strongly contribute towards lower energy background radiation, (2) fast neutron-induced prompt discrete gamma-rays are produced by the (n, n′) reaction in the HPGe or surrounding materials (e.g. Cu, Fe, Pb, Cd), with their Compton scattering adding further to the continuum and (3) delayed gamma-rays including bremsstrahlung and Compton scattering, from the de-excitation of isotopes formed in the crystal or surrounding materials by the capture of thermalized neutrons [6]. Unlike most existing cosmic veto devices [7–14], this system does not require dedicated coincidence electronics and utilises the Lynx MCA to provide an easy to configure system with simultaneous acquisition of suppressed and unsuppressed spectra.

Methodology

Instrumentation

The cosmic veto system included a Canberra (90 % relative efficiency) extended p-type HPGe detector (model GX8021) with Canberra Lynx MCA. High voltage was set at +4,500 V with a rise time of 8 µs and flat top of 1 µs. The detector was situated within a low-background graded shield of lead (125 mm), aged lead (<25 Bq/kg ^{210}Pb, 25 mm), tin (1 mm) and copper (1.6 mm). Ambient radon concentrations were minimised using high laboratory air flow. Surrounding the shield were five 55 × 55 cm Bicron BC408 plastic scintillation plates controlled by a single Canberra Lynx MCA with pre-amplifier (model 2007). Rise time was set at 1.2 µs and flat top at 0.6 µs. Each plate was connected to an individual Canberra HV power supply (model 3102D) set at +700 V. Each Lynx was connected to a computer using an Ethernet (10/100T) connection and a synchronisation cable run from unit-to-unit (Fig. 1). Initial configuration and calibration was undertaken using the Canberra Genie Gamma Analysis Software (version 3.2.3).

Data acquisition utilised the time-stamp functionality of the Lynx, with post-processing enabling the rejection of coincident detector–scintillator events. Collection of event data also provided a powerful tool for understanding cosmic interactions with the HPGe. It required the production of custom (C++) acquisition software and the designation of a master MCA (HPGe) and slave MCA (plates).When acquisition was started, an initialisation signal was sent from the master to slave unit using the synchronisation cable, exactly synchronising the start times for each MCA. All events interacting with the HPGe and plates were then logged to a comma-separated text file for data analysis after acquisition.

Data analysis

The time-stamped data was analysed using custom Microsoft Access 2003 software controlled using the Visual Basic for Applications language. The database was programmed to search for coincident events in the comma-separated text files produced using the MCA control software according to user specified parameters. This information was used to reconstruct suppressed (i.e. anti-coincident with plates) and

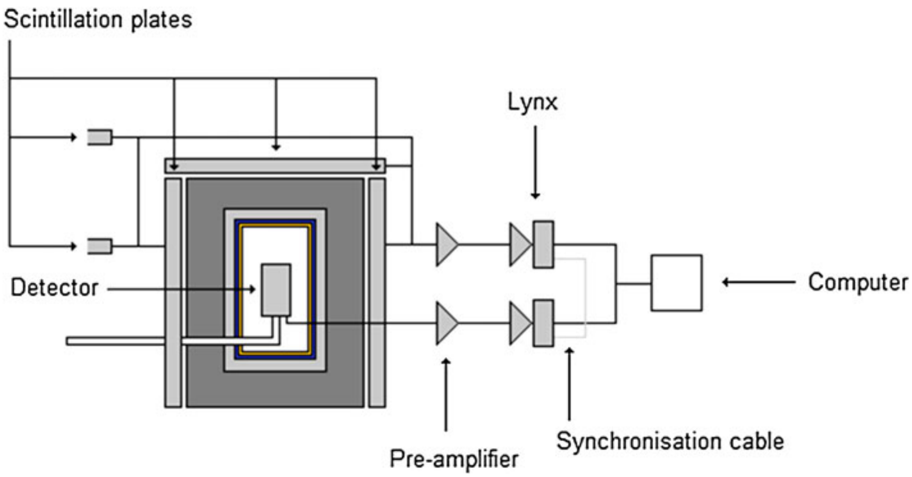

Fig. 1 Schematic of the experimental setup for cosmic veto system. Only three scintillation plates are shown

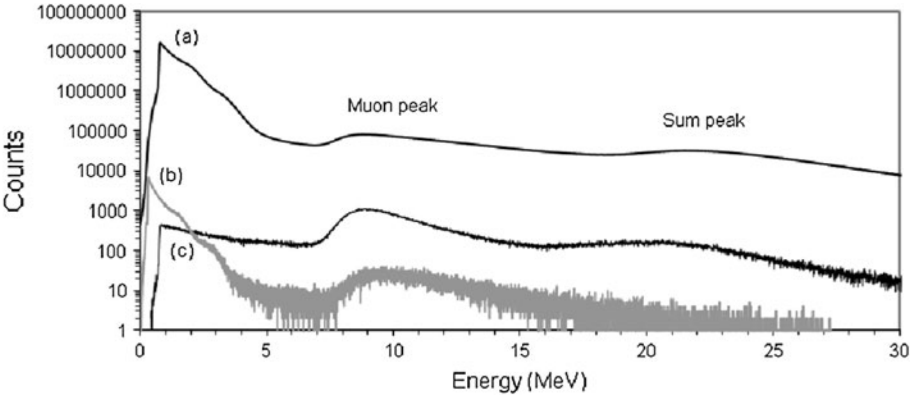

Fig. 2 Cosmic radiation spectra measured using (*a*) five and (*b*) one Bicron BC408 plastic scintillator plates. The signal of the muons crossing the plates is ~10 MeV. The spectrum collected using five plates also shows the presence of a muon sum peak at ~20 MeV. The coincidence cosmic radiation spectrum with the HPGe spectrum is also shown (*c*)

unsuppressed gamma-spectra. Processing of large datasets that exceeded the 2GB Microsoft Access file size restriction [15] were distributed across multiple databases and consolidated after processing. Further optional post-processing and charting was undertaken using custom Microsoft Excel 2003 software. The use of automated routines enabled all data analysis to be performed during acquisition in near real-time.

Samples

The system was tested using empty shield measurements and a CTBT sample collected on 19 August 2012 from radionuclide monitoring station JPP38 (Japan). This sample was contaminated with radionuclides from the Fukushima incident (11 March 2011).

Results and discussion

Optimisation

The time distribution of coincidence events between the HPGe and plates was examined within the range of ±100,000 ns. During a 7 day background measurement there were 1,560,852 coincident events recorded, of which 35.2 % occurred 7,000–8,000 ns in the HPGe after an initial plate event. The events within this timing window were found to significantly reduce the HPGe spectrum, and occurred as a prominent peak in the timing distribution. Their occurrence at 7,000–8,000 ns was a function of the HPGe rise time (8 μs) and flat top (1 μs) as the measured event time is taken at the signal pulse maxima (use of the leading-edge would remove the effect). The remainder of events (64.8 %) occurred as a continuum of noise across the ±100,000 ns range and were attributable to the plastic scintillator high count rates (~2,300 counts s^{-1}) producing random coincidence. Subsequent background suppression was undertaken using only coincidence events within the 7,000–8,000 ns range.

Connection of the five plastic scintillation plates to a single Canberra Lynx MCA produced a characteristic cosmic radiation spectrum (Fig. 2). This spectrum contrasted to earlier single plate measurements [3] due to the presence of a muon sum peak at ~20 MeV that was attributable to horizontal cosmic rays passing through multiple scintillation plates. These muons arrive at the Earth with a zenith angle (θ) slightly less than 90° and angular distribution of $\cos^n\theta$ where $n = 1.32$ [16, 17]. The ratio of the sum peak (28 %) to muon peak (72 %) was in agreement with the horizontal cosmic-ray component measured in the laboratory [18] and reported elsewhere [19]. Construction of the coincidence cosmic spectrum indicated that 10 MeV muons were the dominant cosmic interactions with the HPGe [17]. Further investigation of the event timing distribution allowed correlation of specific cosmic interactions with the HPGe, including the observation of the 444 ns half-life for ^{72}Ge [6].

Background reduction

Post-processing of event data collected during a 7 day background allows reconstruction of suppressed and unsuppressed gamma-spectra (Fig. 3). Removal of coincident events reduced the background spectra by a mean of 75.2 %. This was a significant improvement on the previous value of 54.5 % [3] and was attributable to the complete processing of the cosmic-ray spectrum (0–25 MeV) with improved single Lynx MCA electronics. Examination of the event data indicated that 12.6 % suppression was coincident with the cosmic-ray spectrum <5 MeV and 62.6 % suppression was >5 MeV. This represents respective contributions from fast neutron-induced prompt discrete gamma-rays and prompt continuously distributed background. Reduction of ^{72}Ge and ^{74}Ge peaks produced by the (n, n′) reaction ranged from 70.5–77.4 % (Table 1). These neutron capture gamma-rays are produced continuously by thermalised neutrons and inelastic scattering of fast neutrons from cosmic origin. The right asymmetric line shape is due to imperfect transformation

Fig. 3 Measurement of background gamma-spectra. The *chart* gives a comparison of the unsuppressed (*dark grey*) and suppressed (*light grey*) spectra, with magnification of the annihilation (511.0 keV), ^{72}Ge (691.0 keV), ^{65}Cu (962.1 keV) and ^{208}Tl (2,614.6 keV) peaks

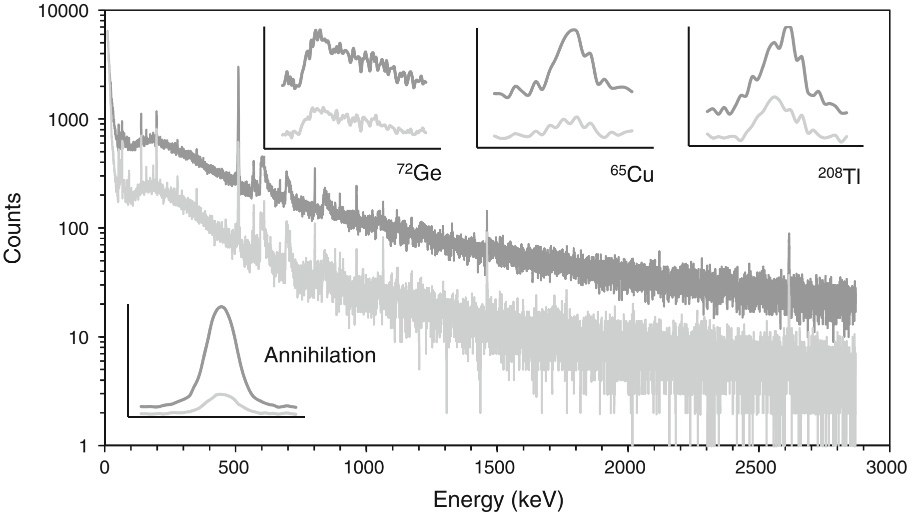

Table 1 Reduction of cosmic background lines

Energy (keV)	Isotope	Reaction	Reduction (%)
185.9	^{66}Cu	^{65}Cu(n, γ)^{66}Cu	12.1
278.3	^{64}Cu	^{63}Cu(n, γ)^{64}Cu	27.0
511.0		Annihilation	80.7
562.9	^{76}Ge	^{76}Ge(n, n′)^{76}Ge	72.5
569.8	^{207}Pb	^{207}(n, n′)^{207}Pb ^{206}Pb(n, γ)^{207}Pb	54.5
595.9	^{74}Ge	^{73}Ge(n, γ)^{74}Ge ^{74}Ge(n, n′)^{74}Ge	71.6
669.6	^{63}Cu	^{63}Cu(n, n′)^{63}Cu	39.8
691.0	^{72}Ge	^{72}Ge(n, n′)^{72}Ge	70.5
803.3	^{206}Pb	^{206}Pb(n, n′) ^{206}Pb^{210}Pb	58.5
834.0	^{72}Ge	^{72}Ge(n, n′)^{72}Ge	76.1
962.1	^{65}Cu	^{63}Cu(n, n′)^{63}Cu	73.7
1,039.5	^{70}Ge	^{70}Ge(n, n′)^{70}Ge	77.4
1,063.6	^{207}Pb	^{207}Pb(n, n′)^{207}Pb ^{206}Pb(n, γ)^{207}Pb	25.1
1,115.5	^{65}Cu	^{65}Cu(n, n′)^{65}Cu ^{65}Cu(p, n)^{65}Zn	52.1
1,204.1	^{74}Ge	^{74}Ge(n, n′)^{74}Ge	76.6
2,614.6	^{208}Pb	^{208}Pb(n, n′) ^{208}Pb^{208}Tl	57.2

of the recoil energy of the Ge atoms [5]. Prompt neutron capture of materials surrounding the detector including ^{64}Cu, ^{65}Cu, ^{66}Cu, ^{206}Pb, ^{207}Pb and ^{208}Pb were reduced by 12.1–73.7 %. A reduction of 57.2 % was achievable in the ^{208}Tl peak at 2,614.6 keV due to neutron capture of ^{208}Pb. The highest reduction of 80.7 % occurred for the annihilation peak due to pair production process induced by high energy gamma-rays of cosmic origin and muon-induced pair production. Measurement of a CTBT sample contaminated with radionuclides from the Fukushima incident had a background reduction of 66.6 % (Fig. 4). During the measurement there was no loss of counts for the detected radionuclides by coincident veto. This was demonstrated by calculation of the suppressed to unsuppressed count ratio for ^{210}Pb (1.00 ± 0.007), ^{7}Be (1.00 ± 0.002), ^{134}Cs (1.00 ± 0.006), ^{137}Cs (0.99 ± 0.004) and ^{40}K (0.97 ± 0.05).

MDA improvements

Calculation of the minimal detectable activity (MDA) for the 85 CTBT radionuclides during a background measurement gave a mean improvement of 45.6 %, with values of 19.6–64.8 %. This included the ^{140}Ba MDA which is a CTBT requirement to achieve 24 mBq for a compressed air filter with dimensions of 70 mm diameter and 6 mm height [20]. Using a conventional HPGe this is typically achieved after 5–7 days. The cosmic veto system allows an MDA of 19.2 mBq after 2 days, and 11.3 mBq after 7 days (Fig. 5). Unsuppressed MDA values during this time period are 35.9 and 21.7 mBq, respectively. The suppressed values are also an improvement on the earlier system design, which allowed an MDA of 22.3 mBq after 2 days [3]. Importantly the new system allows the ^{140}Ba MDA to be achieved after 1.5 days, giving increased sensitivity and sample throughput. Other key indicators of nuclear weapons tests include ^{95}Zr, ^{99}Mo, ^{141}Ce and ^{147}Nd. Over 7 days the average MDA improvements were 52.7, 37.3, 38.5 and 52.1 %. The system also provided increased sensitivity for radionuclides associated with a nuclear reactor incident, including ^{131}I, ^{134}Cs and ^{137}Cs with MDA improvements of 42.0, 46.6 and 49.5 %. For the CTBT sample collected after the Fukushima incident, that contained measurable amounts of ^{134}Cs and ^{137}Cs, the MDA was improved by 31.8 and 36.8 %.

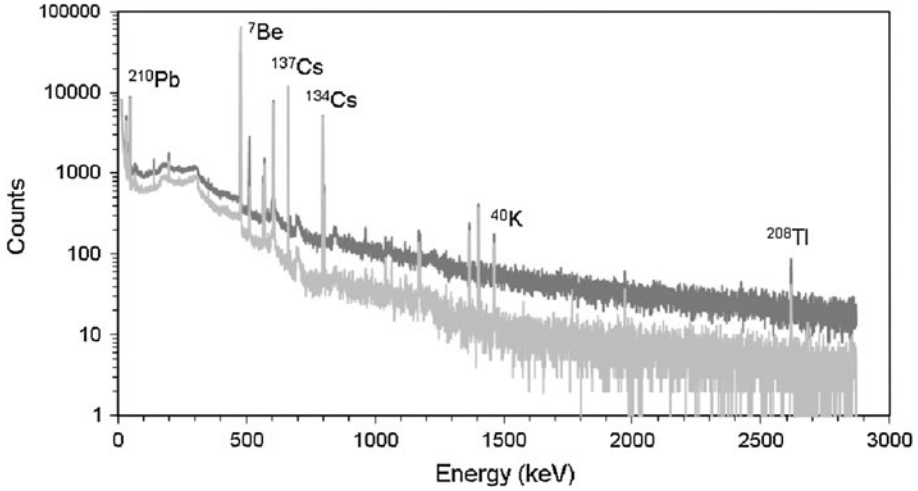

Fig. 4 Measurement of a CTBT air filter sample collected after the Fukushima incident. The *chart* gives a comparison of the unsuppressed (*dark grey*) and suppressed (*light grey*) spectra

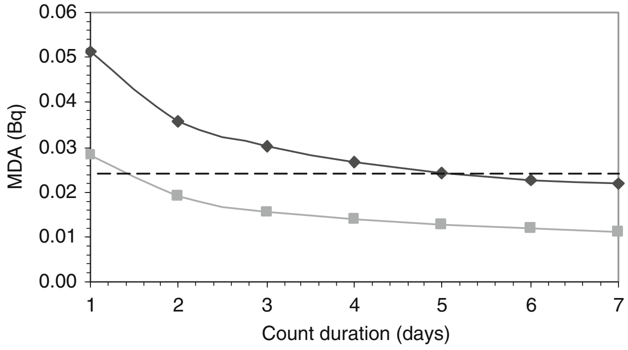

Fig. 5 Improvements in unsuppressed (*black*) and suppressed (*grey*) ^{140}Ba MDA with increased count duration. The CTBT requirement for a ^{140}Ba MDA of 24 mBq is shown by the *horizontal line*

Conclusions

The cosmic veto gamma-spectrometer provides improved sensitivity for radionuclides indicative of nuclear weapons tests and reactor incidents, including ^{140}Ba, ^{95}Zr, ^{99}Mo, ^{141}Ce, ^{147}Nd, ^{131}I, ^{134}Cs and ^{137}Cs. The complete processing of the cosmic-ray spectrum (0–25 MeV) combined with single Lynx MCA electronics provides improved background reduction by a mean of 75.2 %. The system does not require dedicated coincidence electronics, and remains easily configurable with dual acquisition of unsuppressed and suppressed spectra. This is advantageous for increasing the sensitivity of radionuclide laboratories and monitoring stations that support the CTBT. The requirement for a ^{140}Ba MDA of 24 mBq is now achievable after 1.5 days counting compared to 5–7 days using conventional systems. The improved sensitivity has been demonstrated for a CTBT air filter sample collected after the Fukushima incident with no compromise to other radionuclides within the sample including ^{210}Pb, ^{7}Be, ^{134}Cs, ^{137}Cs and ^{40}K.

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