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A system for low-level the cosmogenic ^{22}Na radionuclide measurement by gamma–gamma coincidence method using BGO detectors

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Abstract In this study, a gamma–gamma coincidence spectrometry was developed and examined for environmental low-level cosmogenic ^{22}Na monitoring purposes. The spectrometry consists of two bismuth germanate scintillators (BGO) and XIA LLC Digital Gamma Finder (DGF)/Pixie-4 software and card package. The developed spectrometry was optimized according to the considerations of output count rate and gamma peak energy resolution. This spectrometry provides a more sensitive and effective way to quantify even trace amounts of ^{22}Na with critical detection limit of 9 mBq. A sophisticated computer simulation was also created with the goal of obtaining a better understanding of the experimental results and gamma–gamma coincidence efficiencies at different sample geometries.

Keywords Gamma–gamma coincidence · ^{22}Na · BGO · Aerosol sample

Introduction

The cosmogenic radionuclide ^{22}Na (half-life: 2.602 years) is generated in the upper atmosphere by spallation between argon in air and protons or secondary neutrons from cosmic rays. The activity ratio of ^{22}Na and another cosmogenic radionuclide, namely ^{7}Be (half-life: 53.3 days), is often used as tracer of stratosphere-troposphere vertical exchange, in global aerosol radionuclide monitoring networks [1–4]. These two radionuclides are simultaneously generated in the upper atmosphere and have similar behaviour after production. The gamma-rays of 1274.5 keV and 477.8 keV emitted by ^{22}Na and ^{7}Be are often used to determine their activities by HPGe single-gamma spectrometry. The problem is that the activity level of ^{22}Na is extremely low, which is about four orders of magnitude lower than that of ^{7}Be. In addition, the low full-energy peak efficiency and the rise of background by Compton scattering of 1460.8 keV from ^{40}K make the 1274.5 keV gamma-ray more difficult to be determined with single-gamma spectrometry.

^{22}Na is a proton rich nuclide and 90% of its decays to yield positrons. The positron travels only a short distance before it annihilates with an electron. As a result of the annihilation process, the two 511.0 keV annihilation photons emit with an angle of 180^{o} between each other. Based on this useful decay property, a BGO gamma–gamma coincidence spectrometry has been developed in this work. The purpose of this experiment is to determine ^{22}Na with lower detection limits by using gamma–gamma coincidence method to detect the gamma-rays emitted only when the positron is captured. The method has three major advantages over the most commonly used single-gamma spectrometry for ^{22}Na measurement. First, the background continuum near coincident signatures is reduced by simultaneously detecting two 511.0 keV annihilation photons in two detectors, which significantly lowers the minimum detectable activity of ^{22}Na such that its activity becomes the dominant signal in the detection system. Second, the gamma–gamma coincident counting eliminates many of the interferences that are presented in single-gamma spectrometry. Finally, the study has demonstrated that the coincidence efficiency of 511.0 keV by both detectors is even higher than that of 1274.5 keV gamma-ray by the single HPGe detector measurement. This is because of

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the detector efficiency differences at the two energy levels, the specific decay signatures of ^{22}Na and high gamma–gamma coincident emissions of the annihilation process.

Experimental

System descriptions and calibrations

The gamma–gamma coincidence spectrometry consists of two 7.62 × 7.62 cm BGO scintillators with their endcaps facing each other and the endcap-to-endcap distance is continuously adjustable. A sample can be positioned directly on the endcaps of two detectors for counting. This orientation provides a large geometric efficiency when the two detector distance is small. A known drawback to this configuration is detector cross-talk. However, these cross-talk events, resulting from coincident backscattered gamma-rays, are mostly located on diagonal lines, which affect very little ^{22}Na 511.0 keV coincident counting. The BGO was selected because of its high stopping power of 465 keV/cm at 511.0 keV, which is about three times higher than that of NaI(Tl) scintillator. Each BGO detector is a combination of scintillator/photomultipier (PMT) and preamplifier manufactured by Bicron (Saint Gobain), Newbury. The PMT high-voltage power supply is a programmable DC power supply from National Instruments, Austin, model number NI PXI-4110. The preamplifier is powered by an ORTEC portable power supply, model number 4002P.

The data-acquisition system for the spectrometer utilizes all-digital electronics, based on the X-Ray Instrumentation Associates (XIA LLC) Digital Gamma Finder (DGF)/Pixie-4 software and card package [5]. The Pixie-4 card is a four channel digital pulse-processing module deployed on a chassis with CompactPCI/PXI backplane from National Instrument. The waveform of an input signal, taken directly from BGO preamplifier output, is continuously sampled and digitized by a 14-bit analog-to-digital converter (ADC) at a rate of 7.5×10^7 samples/s. The signal pulse height is determined by a programmable, digital trapezoidal energy filter implemented in a field-programmable gate array (FPGA). Preamplifier pulse heights are determined to 16-bit resolution. Event timing and pulse-pileup inspection is also carried out in the FPGA by a fast programmable trapezoidal trigger filter. Events are time-stamped at the full ADC rate of 75 MHz. The Pixie-4 card resides in a 4-unit PXI crate. A host desktop PC controls the pulse processing module and performs data readout. All operating parameters, including the filter values, are user adjustable in the software on the host PC. The coincidence time window is also set in the software with a step of 13.33 ns and a lower limit of 79.33 ns, thus allowing accurate reconstruction of coincident interactions in two BGO detectors. The parameters of trapezoidal trigger and energy filter, such as trigger threshold, filter rise and flat top time, are optimized by maximizing the output and input count rate ratio and minimizing the 661.7 keV peak resolution of ^{137}Cs. Whenever a valid event is detected, a digital signal processor (DSP) reads out the energy filter values, reconstructs the pulse height, and bins the energy. In the case of coincidence counting, the DSP obtains the coincidence pattern of active channels in a given event. The simplicity of the present coincidence system is especially apparent in contrast to the systems associated with many NIM-based analog processing modules.

Three certified standard point sources (^{137}Cs, ^{22}Na and ^{60}Co) from Eckert & Ziegler Analytics were used for the energy, energy resolution and full-energy peak efficiency calibration of each BGO scintillator. The calibration data are listed in Table 1. By tuning each PMT's high-voltage supply, and gain, and offset of each Pixie-4 card input channel, the same full-energy gamma ray appears in almost the same center channel for both BGO detectors, as shown in Table 1. The energy resolution of the two BGO detectors is about 11% at 661.7 keV, which is very close each other. It should be noted that the full-energy peak efficiencies listed in Table 1 were not corrected by cascade coincidence summation effect, such that they can be used to estimate the gamma–gamma coincidence counting efficiency.

Table 1 Energy, resolution and efficiency calibrations of each single BGO detector

Gamma-ray (keV)	Peak centre, channel		Energy resolution (FWHM) (%)		Full-energy peak efficiency (%)	
	BGO (A)	BGO (B)	BGO (A)	BGO (B)	BGO (A)	BGO (B)
511.0	623	622	14.2	13.7	30.2	30.5
661.7	807	804	11.4	11.0	30.3	30.8
1173.2	1455	1456	9.0	8.6	11.8	12.4
1274.5	1579	1583	8.4	8.1	7.2	7.3
1332.5	1654	1656	8.2	7.9	10.9	11.3

System modeling

Simulations of the system detection efficiency performance and measurement of ^{137}Cs, ^{22}Na and ^{60}Co were conducted using the Geant4 [6]. Two principle simulations were created. First, a simulation of a single BGO scintillator was created. This simulation was experimentally verified before the creation of the second simulation. The second simulation consists of two BGO scintillators which will be used for coincidence counting.

The BGO scintillator is composed of two main parts: bismuth germanate crystal of 7.62 cm in diameter and 7.62 cm long, and a PMT tube. These two parts are optically coupled and shielded as well. Around this crystal is a layer of 0.315 cm thick aluminum shielding. On the top of the crystal is a layer of 0.05 cm thick aluminum shielding. The inner surface of the shielding is coated with a white reflector. The reflectivity of this surface has been set at 95%. The PMT optically coupled to the bottom of the BGO crystal has a borosilicate glass window. Under the 0.1 cm optical window of the PMT is a bialkali photocathode. The effective radius of the photocathode is 3.5 cm.

The Geant4 Toolkit requires the user to select the physics processes which will be incorporated into the simulation. For this simulation, the standard electromagnetic (EM) package was used. Along with the EM package the standard decay and radioactive decay physics process were also included. Finally, since the simulation handles the modeling of light, the following optical photon processes were included, such as Cerenkov, scintillation, Rayleigh scattering, absorption, and boundary processes. The indexes of refraction of bismuth germanate and borosilicate glasses are defined as 2.15 and 1.50 respectively. The attenuation length of bismuth germanate is defined as 280 cm. No attenuation length was defined for borosilicate glass, as the amount of photons lost to absorption is negligible. The BGO crystal maximum emission wavelength was defined as 480 nm and the decay constant was set at 300 ns. The light yield was defined as 10,000 photons/MeV.

During an event, the energy deposited in the scintillation crystal results in the production of light, with the intensity proportional to the energy. This light then travels around inside the detector, until it is either lost (due to absorption, etc.) or is detected by the photocathode. Every time a photon is detected by the photocathode, a count is created. The result at the end of the event is the total number of photons that were detected by the photocathode, which gives the intensity of light at the photocathode. Also during an event, the BGO crystal detector records the absolute energy deposition that occurs. At the end of the event the absolute energy deposition in the crystal is recorded for that event.

Results and discussion

Four ^{22}Na standard point sources purchased from Eckert & Ziegler Analytics were measured by the system. The certified activities of the four point sources ranging from 37,000 to 37 Bq with ±1.4% relative uncertainty (at 95% confidence level). For each standard, list-mode data were collected by the gamma–gamma coincidence counting system over 120 s at a 119.33 ns coincidence window. The data acquired in list-mode include coincidence events consisting of records of energy in DSP units and timestamp from BGO detector "A" and "B". The list-mode data were stored in a text file for offline analysis. During offline analysis, the DSP units of coincidence events from both BGO detector "A" and "B" were divided by detector's calibration factor correspondingly, then histogrammed to a three-dimensional (3D) gamma–gamma coincident energy spectrum. An example is illustrated in Fig. 1.

As shown in Fig. 1, the most intensive peak results from 511.0 to 511.0 keV coincidences in both detectors; then followed by 511.0 and 1785.5 keV sum peak (511.0 + 1274.5 keV) coincidences. The coincidence of 511.0 keV in one detector and 1274.5 keV in other detector has the lowest probability. The vertical and horizontal lines trailing from each peak are due to random coincidences with Compton-scattered gamma-rays; the diagonal lines result from detector cross-talk. A rectangular region of interest (ROI) is defined to determine the numbers of counts associated with the strongest coincident peak of 511.0 and 511.0 keV. The ROI boundary definitions are based on corresponding peak energy resolutions of detector "A" and "B". The background coincidence count rate in the ROI of 511.0 keV is 0.015 (count/s). It has been

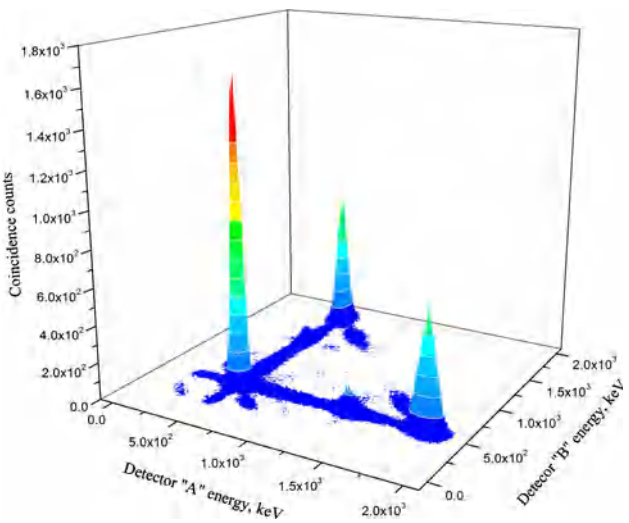

Fig. 1 A 3D histogram gamma–gamma coincidence spectrum obtained by a ^{22}Na standard point source

demonstrated that the background counts are mainly contributed by activity of ^{40}K contained in the borosilicate glass of the PMT window and bialkali photocathode. The sample gross coincidence counts in the ROI are calculated by summing the counts per channel within the region. The net counts are calculated by correcting the gross counts for the effects of detector background. The gamma–gamma coincidence detection efficiency for the ^{22}Na whose signature is located within the ROI bounds can be calculated by Eq. 1.

$$\varepsilon_{\gamma\gamma} = \frac{C_{\mathrm{ROI}}}{A \times T \times \gamma\gamma} \times 100 \quad (1)$$

Where: C_{ROI} is the number of net coincidence counts in the ROI, A is the activity of the standard source, T is total counting time, $\gamma\gamma$ is the gamma–gamma branching ratio of 511.0 keV gamma-rays.

Using the equation, gamma–gamma coincidence efficiency of ^{22}Na was calculated as (9.98 ± 0.59) % at 511.0 and 511.0 keV coincident peak for the point source close counting geometry. The critical limit (L_C) [7] of this coincident peak is estimated about 9 mBq by its coincidence efficiency and background coincidence count rate in the ROI. The efficiency of second strong coincident peak of 511.0 and 1875.5 keV was also estimated as (6.75 ± 0.42) % by the ratio of net coincidence counts in ROI of 511.0 and 511.0 keV to that of 511.0 and 1875.5 keV because of an unknown $\gamma\gamma$ for this coincidence pair. The background coincidence count rate is 0.005 (counts/s) in the ROI of 511.0 and 1875.5 keV. The L_C is calculated about 6 mBq, which is about two times lower than that of 511. keV coincidence peak.

To verify the accuracy of the system modeling, a simulation for ^{137}Cs was performed on one single detector. The simulated spectrum is illustrated in Fig. 2, together with the experimentally measured one. The *y*-axis of Fig. 2 is normalized to the top of the 661.7 keV energy peak, since the two spectra had different numbers of counts.

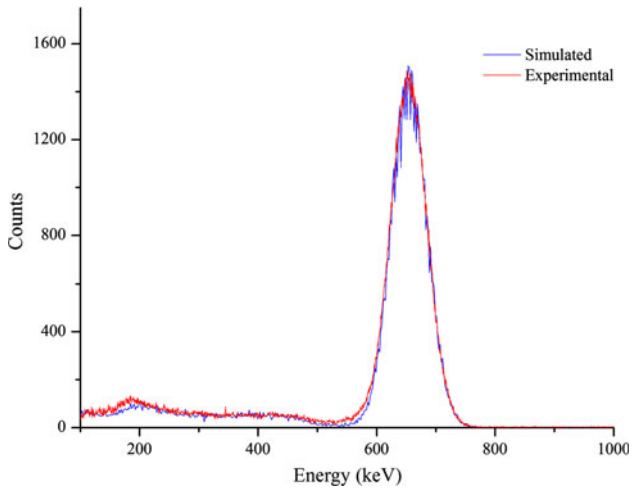

Fig. 2 Simulated and experimentally measured single detector energy spectrum of BGO "A" by ^{137}Cs point source

As shown in Fig. 2, the simulated and experimentally determined spectra are well matched both at full-energy peak and at the Compton plateau. The simulated detector efficiency and energy resolution at 661.7 keV are 29.9 and 11.2% respectively, which have good agreement with the experimental data of 30.4 and 11.4% in Table 1. The results indicate that the BGO detector modeling closely matched the physical one.

A coincidence simulation was also performed for ^{22}Na point source. Figure 3a shows the simulated 3D gamma–gamma coincidence histogram of ^{22}Na. Comparison with a experimental gamma–gamma coincidence 3D histogram of ^{22}Na, as illustrated in Fig. 3b, shows good agreement both in the gamma–gamma coincidence features and energy regions.

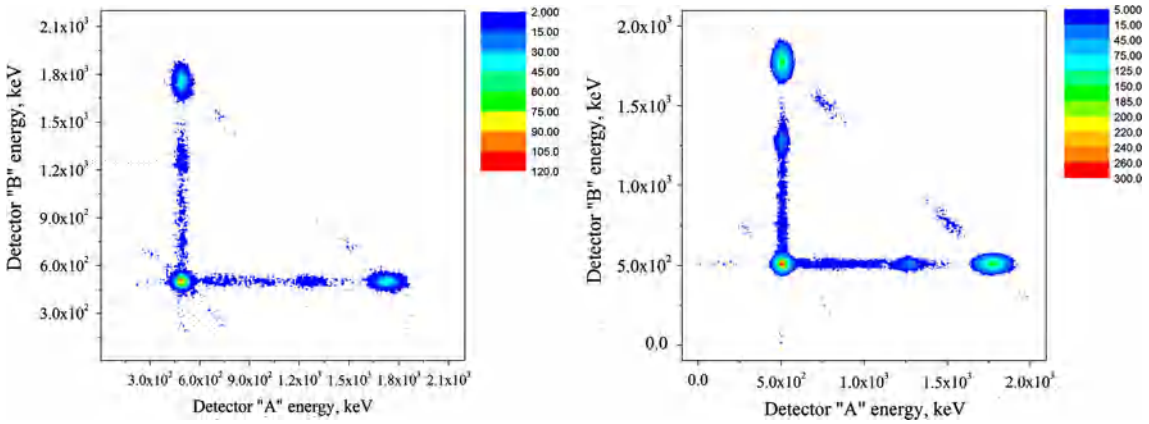

Fig. 3 Top view of the simulated 3D histogram of ^{22}Na by 150,000 decays (*right*), *top view* of the experimental 3D histogram by 370 Bq ^{22}Na point source counted for 120 s (*left*)

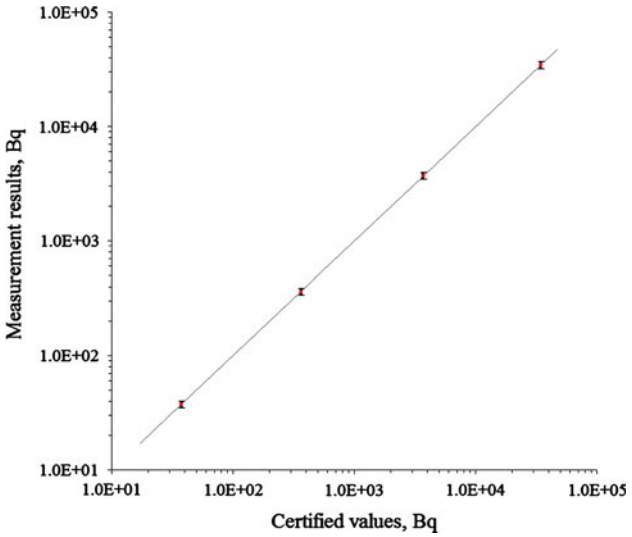

Fig. 4 Comparison of ^{22}Na activity between measurement results and the values from certificate

The 511.0 keV gamma–gamma coincidence efficiency calculated with the simulation is about 9.82%, which agrees well with the experimentally determined results. Using the efficiencies and the procedure of ^{22}Na gamma–gamma coincidence quantitative analysis, four ^{22}Na standard point sources were analyzed. The activities determined are plotted in Fig. 4 together with the certified values. The results are in very good consistency with the certified values of each standard, showing good accuracy and reproducibility at the activities ranging from 37 to 37000 Bq. Two aerosol samples were also analysed by the gamma–gamma coincidence counting system. They were obtained from regional laboratory in northern Finland of Radiation and Nuclear Safety Authority. The collection location for both samples was Rovaniemi, Finland. One was collected from May 10–17, 2010, air volume 104713 m^3; the other was collected from May 17–24, 2010, air volume 102993 m^3. The reason for using these samples is that their ^{22}Na activities have been analysed at the regional laboratory in northern Finland using conventional HPGe detector; and they are collected in a real environment where typical radon progenies and ^{40}K are present in the background. The ^{22}Na activities, independently analysed by HPGe and the gamma–gamma coincidence counting system, are 50.3 ± 5.1 and 49.7 ± 4.1 mBq in one sample; and 60.2 ± 6.1 and 60.3 ± 4.2 mBq in the other. The data shows an excellent agreement. It should be noted that dead-time correction is necessary on coincidence count rate when the filter dead time of Pixie-4 card input channel is over 5% of live time.

Conclusions and future work

In this study, a gamma–gamma coincidence spectrometer has been developed, using two BGO detectors and a DGF Pixie-4 digital multichannel data acquisition system, for the purpose of exploring environmental low-level cosmogenic ^{22}Na coincidence measurements. The study demonstrates that the advanced Pixie-4 digital data acquisition and signal processing system can be used successfully to replace the conventional analog electronics system in quantitative gamma–gamma coincidence spectrometry. An initial evaluation of quantitative coincidence counting has been performed based on four certified ^{22}Na point sources and aerosol samples, which corroborated the feasibility of this approach. Future improvement to the system would be background reduction by using a passive shield and modifying the BGO detector with a low potassium PMT. The background counts due to ^{40}K are expected to be minimized.

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