

Development of Antic cosmic Shielded Ultra Low Background Gamma Spectrometer for Precise Measurement of Environmental Radioactivity

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(Received May 30, 2002)

Abstract

We developed an ultra low background gamma ray spectrometer particularly suitable for experiment which require lower detection limit. The background of a germanium spectrometer is suppressed by applying active and passive shielding technique at the same time. The active shielding devices consist of plastic scintillating plates of 50 mm thick and anti-coincidence electronic system. The shielding is made of 150 mm thick walls of very low activity lead, 20 mm with activity of <10 Bq/kg and 130 mm with activity of <50 Bq/kg. The observed background count rates are 1.2 s^{-1} and 0.36 s^{-1} without and with the active shielding, respectively, overall the energy regions from 30 keV to 3 MeV. The cosmic ray induced background is suppressed by a rate of 0.8 s^{-1} at the present work. The detection efficiency curve necessary to obtain the radioactivity of environmental samples has been precisely determined on the energy regions from 80 to 2000 keV with a 10^3 ml marinelli beaker sample, consisting of the calibrated radionuclides ^{109}Cd , ^{57}Co , ^{139}Ce , ^{203}Hg , ^{113}Sn , ^{85}Sr , ^{137}Cs , ^{60}Co and ^{88}Y . Virtues of the method are demonstrated by measuring the activity of ^{137}Cs contained in the powdered milk.

Key Words : cosmic rays, antic cosmic shield, germanium spectrometer, bulk environmental sample, MDA

1. Introduction

Recently, many user of gamma spectrometer are concerned with measuring low levels of radioactivity such as found in routine environmental

samples. For these, the background, thus the MDA (minimum detectable activity) value, should be as low as possible to provide lower detection limit. Usually in the low count rate systems, such parameters as the decision limit L_c or the detection

limit L_D or the determination limit L_Q can be of a criterion that estimate the capabilities of the measurement process itself [1,2]. These parameters are determined entirely by the error-structure arising from the backgrounds. The background on the gamma spectrometer will originates from; i) the intrinsic radioactivity of the detector and ii) the radioactivity of the materials surrounding the detector and iii) the cosmic radiation. The radiations due to former two cases can be reduced by rigorous selection of the materials with their low activities, or by maintaining the pressure of the inside slightly higher than outside. Then the cosmic ray induced background become predominant, but these can be suppressed if the spectrometer is installed in an underground laboratory [3-5]. Otherwise, active shield method, consisting of plastic scintillating plates surrounding the lead shield and the electronics to veto the coincidence events, can be of an alternative technique and so many researcher have contributed in developing the method to be suitable in the practical works [6-8]. We have been developing such a gamma ray spectrometer, using active and passive shield devices at the same time. We report on the results associated with the performance of these devices for practical works, especially in the analysis of very low-level environmental radionuclides. For these aims, we prepared a powdered milk sample in a 10³ ml minelli beaker. The decision limit, the detection limit and the MDA are investigated by measuring the activity of ¹³⁷Cs contained in the powdered milk.

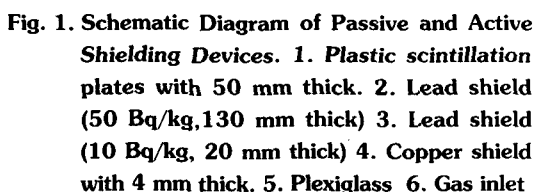
2. Cosmic Ray Induced Background

A cosmic ray is a high-speed particle, either an atomic nucleus or an electron, that travels throughout the Milky Way Galaxy, including the solar system. Some of these particles originate

from the Sun, but most come from sources outside the solar system and are known as galactic cosmic rays. The primary cosmic rays hitting the earth's atmosphere consists of very high energy protons and alpha particles (up to 10⁸-10¹⁰ GeV). These particles undergo collision in the stratosphere to produce secondary radiations of many sorts but largely pions. Down at the earth's surface, further reactions of the pions and decay have resulted in mainly high energy muons. Muons interact with the materials surrounding the crystal to produce many sorts of particles, which give rise to background pulses. The cosmic ray induced background on the germanium spectrometer can be summarized with the following three types (see reference [8] for more details); i) prompt background pulses ii) cosmic bremsstrahlung iii) neutron induced background. The former two cases can be almost suppressed by means of active shielding devices. Muon is both very penetrating and a prime source of fast neutrons. The fast neutrons generate discrete gamma lines through (n, n') reactions in the high Z materials (Ge, Pb, Cu) and, after sufficiently slow down, produces Ge isomers by means of the activation processes in the crystal itself [9,10]. Since the half-lives many of these isomers are significantly longer than the coincidence resolving times, the neutron induced background result in delayed pulses which are not in time coincidence with the primary cosmic particles. So only the few portions can be suppressed by the anti-coincidence shield.

3. Experimental Setup

Figure 1 shows the ultra low level gamma ray spectrometer developed at the present work. The shielding is made of 150 mm thick walls of low activity lead, 20 mm with very low activity of < 10 Bq/kg and 130 mm with activity of < 50 Bq/kg. Since the lead is a very poor neutron absorber,



additional lead shield will induces many thermal neutrons nearby the crystal.(the flux in the lead is an order of 10^{-3} - $3 \times 10^{-3} \text{ cm}^{-2} \text{ s}^{-1}$). Recommended thickness is 150 mm according to the earlier work of Nunez-Lagos and Virto [11]. An additional 4 mm thick copper shielding has been mounted around the detector to suppress the lead fluorescence X rays. The lead shielding is surrounded by scintillating plates made by Bicon Corp. They consist of seven pieces of 50 mm thick 700 mm x 350 mm and 700 mm x 700 mm, respectively, plastic scintillating plates (BC408). The germanium detector was made of ORTEC Corp. It is a p-type coaxial Ge crystal of 282 cm^3 sensitive volume. The relative efficiency is 60 % with resolution of 2.0 keV to the 1332 keV photopeak of ^{60}Co . The materials of the detector assembly are rigorously selected with their



Table 1. List of Radionuclides Prepared to Obtain the Detection Efficiency Curve for 10³ ml Marinelli Beaker Samples

| Nuclide | Energy (keV) | Half-life (day) | Emission Rate (s ⁻¹) | Detection Efficiency | Fitting Error (%) |
|-------------------|-----------------|--------------------|-------------------------------------|-------------------------|----------------------|
| ¹⁰⁹ Cd | 88.03 | 462.6 | 199.25 | 0.013 | 0.01 |
| ⁵⁷ Co | 122.06 | 271.8 | 121.78 | 0.027 | 0.34 |
| ⁵⁷ Co | 136.47 | 271.8 | 15.24 | 0.031 | 0.60 |
| ¹³⁹ Ce | 165.85 | 137.6 | 59.99 | 0.035 | 0.27 |
| ²⁰³ Hg | 279.19 | 46.6 | 4.12 | 0.035 | 0.63 |
| ¹¹³ Sn | 391.69 | 115.1 | 119.04 | 0.029 | 2.20 |
| ⁸⁵ Sr | 514.00 | 64.9 | 40.01 | 0.023 | 2.80 |
| ¹³⁷ Cs | 661.66 | 11020 | 1306.04 | 0.019 | 1.33 |
| ⁸⁸ Y | 898.07 | 106.6 | 300.79 | 0.015 | 0.51 |
| ⁶⁰ Co | 1173.24 | 1925.5 | 1573.64 | 0.012 | 0.03 |
| ⁶⁰ Co | 1332.50 | 1925.5 | 1575.05 | 0.011 | 0.41 |
| ⁸⁸ Y | 1836.08 | 106.6 | 317.98 | 0.010 | 0.18 |

low activity. For example, the end-caps are made of magnesium. the detector holder, clamps and cooling rod are OFHC (Oxygen Free High Conductivity) copper. The crystal is mounted in a U-style cryostat to away from the radioactivity of the preamplifier and the dewar. The inside of the shield is always filled with nitrogen gases, so that the contaminated air can not enter. The flow rate was 50 cc/min. Figure 2 shows the electronic circuit applied to analyze the coincidence events. The pulses from the scintillation plates and Ge detectors are converted to fast logic signals at CFD prepared for each counting channel. The TAC measures the time interval of each start and stop pulse to provide a logic signal if their difference is within the adjusted value of 2μs at the present work. This signal is extended to 50μs width, sufficient to content the shaped linear pulses with 6μs shaping time. It plays a role of gate signal to veto the coincident signal with the ADC input. The detection efficiency curve, available for bulk environmental samples, has measured for 2 ×

10⁵ s with a 10³ ml marinelli beaker sample which contains calibrated mixed sources, ¹⁰⁹Cd, ⁵⁷Co, ¹³⁹Ce, ²⁰³Hg, ¹¹³Sn, ⁸⁵Sr, ¹³⁷Cs, ⁶⁰Co and ⁸⁸Y. In table 1, we list their calibrated activities to the reference date and to the measurement date together with their half-lives. Since each nucleus listed in table decays to their daughter by emitting at most two gamma lines, coincidence sum effect, normally encountered for bulk samples, can be negligible except the cases of ⁶⁰Co and ⁸⁸Y. The powdered milk sample of 582 g was prepared without any pretreatment in a 10³ ml marinelli beaker and the photopeak emitted in the decay of ¹³⁷Cs was measured until the net area is comparable to the determination limit L_Q [1,2], so that the standard deviations of the net area become 10 %. The measurement time was 2 × 10⁵ s.

4. Analysis and Results

In figure 3, we compare the observed background spectra obtained with and without the

active shielding devices. Integral counting rates of the background on the region of 30 keV to 3 MeV, are 1.20 and 0.36 s^{-1} , respectively. The background count rate due to contaminated air was 0.04 s^{-1} . Thus the cosmic ray induced background is reduced by a rate of 0.8 s^{-1} at the present work. It was observed as 0.71 s^{-1} at the earlier work of ref.[8]. They used 50 % efficiency Ge detector which is 10 % lower than ours. Thus we can expect larger suppression rate.

Since the fast neutrons generate gamma lines by means of neutron-neutron reactions in the lead and, after sufficiently slow down, they provides isomers through the activation processes in the detector. The gamma lines emitted in the decay of these isomers are observed on the spectrum. For example in figure 3, the photopeaks of 139.7 and 198.3 keV, which correspond to gamma emission of isomeric level $^{75\text{m}}\text{Ge}$ ($T_{1/2} = 48 \text{ s}$) and

$^{71\text{m}}\text{Ge}$ ($T_{1/2} = 22 \text{ ms}$), are not suppressed. But the gamma line of 174.9 keV populated from the decay of another isomeric state of $^{71\text{m}}\text{Ge}$, $T_{1/2} = 73 \text{ ns}$, is almost suppressed. Besides, the photopeak of 159.1 keV from $^{77\text{m}}\text{Ge}$ and the photopeak of 595.8 keV from $^{74\text{m}}\text{Ge}$ were observed. When the muon collides with high Z materials, it produces many kinds of secondaries such as electrons, positrons and protons. These secondaries are absorbed in the lead and generates gamma rays by means of bremsstrahlung or annihilation effects. The 511 keV photopeak shown in figure 3 is mainly due to cosmic bremsstrahlung. It is suppressed by a factor of 6 by the anti-coincidence shield. Especially the observed background rate of the 662 keV photopeak from ^{137}Cs and the 1461 keV from ^{40}K are an order of $\sim 5 \times 10^{-4} \text{ s}^{-1}$. The peaks at 295.2, 351.9 and 609.3 keV, mostly originated

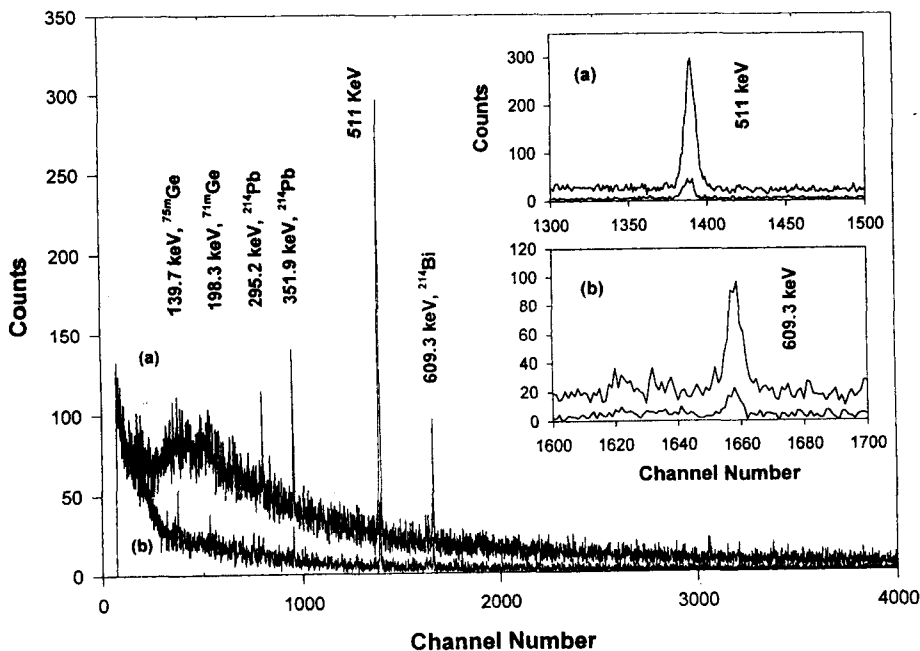


Fig. 3. Comparing the Background Spectra Obtained Without (a) and with (b) the Antic cosmic Device
Inset (a) is the 511 keV Photopeak Obtained with Each Case. Inset (b) is to Show the Reduction of the Air Contamination

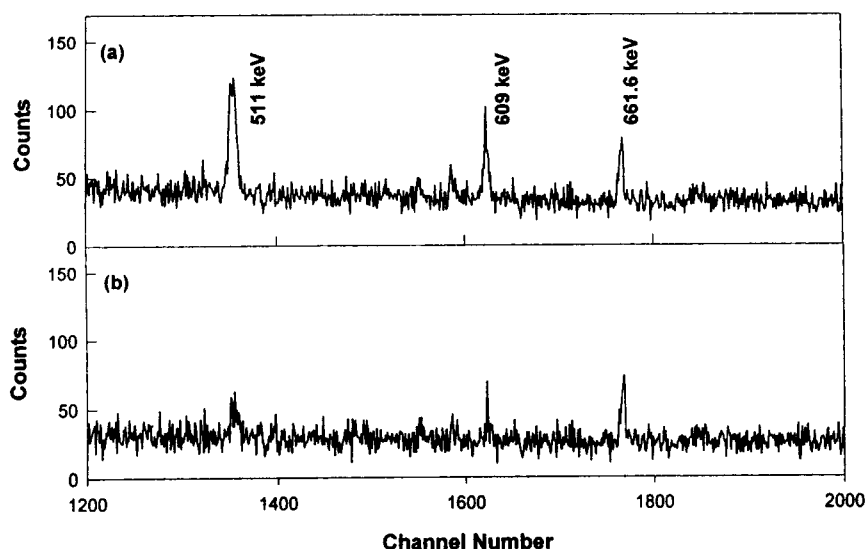


Fig. 4(a). Singles Gamma Ray Spectrum Resulted with 2×10^5 s Measurement for Powered Milk of 10^3 ml Marinelli Beaker Sample. The MDA of 661.6keV Photopeak is 0.018Bq/kg. Fig 4(b) is the Derived one Obtained by Subtracting the Backgrounds

Table 1. Comparing the Background Rates Observed on the Various Energy Ranges Obtained with and without Active Shield

| Energy Range | Count Rates (s^{-1}) | |
|------------------------------------------|--------------------------|------------------|
| | Passive | Passive + Active |
| $30 \text{ keV} \leq E \leq 3\text{MeV}$ | 1.2 | 0.36 |
| $E \leq 500 \text{ keV}$ | 0.75 | 0.28 |
| $E \geq 300 \text{ keV}$ | 0.66 | $0.1 <$ |
| $E \geq 500 \text{ keV}$ | 0.45 | $0.08 <$ |

from the contaminated air in the detection inside, are suppressed by a factor of 6 by flowing the nitrogen gases into the crystal assembly. Inset (b) is to demonstrate the performance in the reduction of activity coming from air contamination.

Consequently, cosmic ray induced background is suppressed by a factor of about 3 overall the energy ranges, by a factor of about 6-7 for $E > 300 \text{ keV}$, and a factor of about 6 for $E > 500 \text{ keV}$. The results are summarized in table 2. The detection efficiency curve was obtained with the

10^3 ml marinelli beaker sample containing the calibrated radionuclides shown in table 1. The measurement time was $2 \times 10^5 \text{ s}$. The efficiency curve was derived by fit the data to the least squared linear function. The fitting error is negligible on the regions of $80 \text{ keV} < E < 210 \text{ keV}$ according to our data. It shows a relatively large value, maximally 2.8 % for 514 keV photopeak, for $210 \text{ keV} < E < 2000 \text{ keV}$. The results are tabulated in table 1 together with corresponding fitting errors.

Finally we applied the method to determining the activity of ^{137}Cs in powdered milk of 10^3 ml marinelli beaker sample. Fig. 4 (a) shows the obtained spectrum resulted in $2 \times 10^5 \text{ s}$ measurement and fig.4(b) is the one derived by subtracting the background events. The activity of ^{137}Cs contained in the sample is determined by using the efficiency data shown in table 1 and found as 0.14 Bq / kg with combined uncertainty of 10.1 %. Concerning the uncertainty estimation,

it was calculated from the square root of the summed squares of each standard uncertainty of most probable component in the measurement, i.e., 1.3 % for efficiency fitting error, 10 % for standard deviation of net area and 0.9 % for decay parameter error.

The net area of 661.7 keV photopeak showing in figure 4(a) is five times larger than the value of decision limit $L_C (= 2.33 B^{1/2})$, where B is background counts, and over the two times than that of detection limit $L_D (= 2.71 + 4.65 B^{1/2})$ [1,2]. The MDA for 661.7 keV photopeak is observed as 1.8×10^{-2} Bq / kg at the present work.

5. Conclusions

We have successfully developed an ultra low-level background germanium spectrometer particularly suitable to determining the activity of environmental samples. The cosmic ray induced background is well suppressed by means of active shield devices, which consists of plastic scintillators and anticoincidence electronics. The observed integral count rates is 0.36 s^{-1} and 1.2 s^{-1} with and without the anti-coincidence devices, respectively, on the energy regions from 50 to 3000 keV. The photopeaks of 511 keV, mainly arising from cosmic bremsstrahlung, is suppressed by a factor of about 6. In addition, the MDA limit is at most 1.8×10^{-2} Bq/kg (= 0.55 pCi/kg) in case for the 661.7 keV photopeak. It is appeared to be a factor of 10 times below than that rates resulted with typical cryostat assembly.[12]

The detection efficiency curve, available for 10^3 ml marinelli beaker sample, has precisely determined on the energy regions from 80 to 2000 keV. Virtues of the method have been demonstrated by measuring the activity of ^{137}Cs presented in powdered milk. Its activity was too small, 0.14 Bq/kg (= 3.8 pCi), to measure by

using the conventional method. Since the activity of the environmental samples are usually far below the MDA limit, one should carries out additional works in sample preparation to achieve the activity exceed that limit, for examples radiochemical concentration or extraction procedures. But the method developed at the present work enables to reduces the MDA limit to very low-level by applying the passive and active shield devices at the same time, thus one can obtains the data with good precisions even with relatively short counting time with small amount of samples prepared without any pretreatment.

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Acknowledgement

This work was supported by Korea Research Foundation Grant No. krf-99-041-D00137D2003