

Construction of a Ge-BGO Compton Suppression Gamma-ray Spectrometer and  
Its Application to Environmental Samples  
Part 1: The System of the Spectrometer and  
Its Basic Performance

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**Abstract.** Construction of a Ge-BGO Compton Suppression Gamma-ray Spectrometer and Its Application to Environmental Samples, Part 1: The System of the Spectrometer and Its Basic Performance. In order to develop a very high sensitive Ge gamma-ray spectrometer, a Compton suppression gamma-ray spectrometer with a BGO guard detector system has been constructed. The BGO detector system detects 88% of the gamma-rays from  $^{60}\text{Co}$  that are committing to the principle of the suppression system. Peak-to-Compton ratio for the 661 keV gamma-rays from  $^{137}\text{Cs}$  has been improved from 128 to 786 with the Compton suppression spectroscopy. By the development of the spectrometer the minimum acceptable radioactivity of  $^{137}\text{Cs}$  has been improved from 0.091 Bq to 0.044 Bq with the coefficient of variation of 30% and measurement time of 8 hours.

**Abstrak.** Construction of a Ge-BGO Compton Suppression Gamma-ray Spectrometer and Its Application to Environmental Samples, Part 1: The System of the Spectrometer and Its Basic Performance. Untuk mengembangkan spektrometer sinar-x Ge dengan kepekaan yang sangat tinggi, telah dibuat sebuah sistem spektrometer sinar-x penindas Compton dengan menggunakan detektor pelindung BGO. Sistem detektor BGO mendeteksi 88% sinar gamma dari  $^{60}\text{Co}$  yang digunakan dalam prinsip sistem penindas. *Peak-to-Compton ratio* untuk sinar gamma berenergi 661 keV dari  $^{137}\text{Cs}$  telah diperbaiki dari 128 menjadi 786 dengan spektroskopi penindas Compton. Melalui pengembangan spektrometer ini radioaktivitas  $^{137}\text{Cs}$  minimum yang dapat diterima meningkat dari 0.091 Bq menjadi 0.044 Bq dengan koefisien variasi 30% dan waktu pengukuran 8 jam..

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## Introduction

A research, development and utilization of nuclear energy should not be accepted without any programs to ensure and maintain nuclear safety. Nuclear spectrometry technique is widely used as the one of the useful techniques to maintain natural environment and to assure safe normal operation of nuclear facilities.

High sensitive gamma-ray spectrometers have been applied to nuclear laboratories from the standpoint of radiation protection, and to diagnoses of nuclear facilities such as nuclear power plants and radioactive-waste management facilities from standpoint of nuclear safety taken the maintenance of natural environment into consideration.

The deeply analyzed measurements results can be fed back to improve the facilities and natural environment, and they will contribute in getting acceptance of nuclear utilization by the general publics, and to promote development of nuclear technologies.

It is essential to reduce background level for improving instrumental sensitivity of radioactivity, especially in low level counting.

Suppression of Compton continuum will be the most effective method to reduce the background in the region of full-energy peaks in multichannel analyzer spectra.

With a view toward development of a high sensitive Ge gamma-ray spectrometer, mainly intending for the detection of  $^{137}\text{Cs}$  in natural environment, and for the leachability test of  $^{137}\text{Cs}$  in radwaste management facilities, a Compton suppression Ge gamma-ray spectrometer has been constructed. A BGO (Bismuth Germanate) of the composition  $\text{Bi}_2\text{Ge}_2\text{O}_{10}$  scintillation detector system has been used as a Compton suppression detector.

Due to the high atomic number (Bi: 83) and high density (7.13 g/cm<sup>3</sup>), BGO scintillation detectors are the most efficient gamma-ray detectors now in use. They are recommended for use as a Compton suppression detectors.

Therefore, it is expected that this spectrometer system achieves an appreciable suppression of Compton background, i. e. makes possible to detect further lower level of radioactivity of  $^{137}\text{Cs}$  compared with the

achievable level by the usual (single) mode of this spectrometer.

## 1. Configuration of the Ge-BGO Compton Suppression Gamma-ray Spectrometer

In the Compton suppression Ge gamma-ray spectrometer constructed in this work, the Ge detector is surrounded by BGO detector system. The signals from the Ge detector have been rejected at a linear gate circuit in anti-coincidence mode by the signals which are generated in the Ge detector and tile BGO detector system at the same time. In other words, the linear pulses of the Ge detector are rejected at a linear gate circuit by the coincidence pulses of the outputs of two timing single channel analyzers for the Ge and BGO detectors.

### 1.1. Configuration of the Ge-BGO Detector System

Simplified drawing of the arrangement of the Ge detector and Compton suppression BGO detectors is shown in Fig. 1.

#### 1.1.1 HP Ge Detector

Manufacturer: Princeton Gamma-Tec, Inc. (USA)  
 Model: IGC 40195, SeT.No.: 2631  
 Detector Geometry: p-type Coaxial  
 Crystal Size: Ø3 nun in diam., 59 nun in length  
 Active Volume: 176 cm<sup>3</sup>  
 Dead layer: 1 nun  
 Detector/Window Distance: 5 mm  
 Relative Efficiency: 40%,  
 FWHM: 0.4 keV at 1332 keV ( $^{60}\text{Co}$ ),  
 X60 eV at 122 keV ( $^{57}\text{Co}$ )

#### 1.1.2 HGD Detector System

For synthesizing low-background BGO crystals, bismuth compound (Bi<sub>2</sub>O<sub>3</sub>) of the lowest radioactive contamination has been chosen, especially avoiding the contamination of  $^{210}\text{Bi}$  that is the one of fallout-nuclides caused by nuclear testing. The radioactive concentration of  $^{210}\text{Bi}$  in the Bi<sub>2</sub>O<sub>3</sub> used for the crystals is less than

0.03 Bq/kg.

The background count-rate of the BGO detector system resulting from the contamination is less than 2 counts per second, which is less than 1% compared to the most contaminated one.

(a) Main BGO Detector

The main BGO detector is annulus-type crystal for detecting side- and back-scattered gamma-rays from the Ge detector, into which the Ge detector is inserted.

Manufacturer: Crismatec (France)  
 Model: EES-Model V  
 Crystal Shape: Annulus  
 Crystal Size: External Diam.: 111 mm  
 Internal Diam.: 83.2 mm  
 Length: 200 mm  
 Crystal Thickness: 50 mm  
 Annulus Diameter: 80.2 mm  
 Crystal Weight: 29.78 kg  
 Housing Thickness: 1.5 mm aluminum alloy  
 FWHM: Better than 20% (661 keV, 137CS)  
 Notes: Seven optically independent side element BGO crystals with seven photomultipliers of 2 inch diameter

(b) Plug BGO Detector

The plug BGO detector is solid cylindrical crystal for detecting back-scattered gamma-rays from the Ge detector. The plug BGO detector is inserted into the annulus of the main BGO detector in usual uses.

Manufacturer: Crismatec (France)  
 Model: EEP-Model V  
 Crystal Shape: Solid Cylinder  
 Crystal Size: 70.2 mm diam. x 76.2 mm  
 Crystal Weight: 2.45 kg  
 Housing Thickness: 0.8 mm aluminum  
 FWHM: Better than 13% (661 keV, 137CS)  
 Notes: One photomultiplier of 3 inch diameter

(c) Back-catcher BGO Detector

Manufacturer: Crismatec (France)  
 Model: EEC-Model V  
 Crystal Shape: Annulus separated into 1st half and 2nd half for surrounding the dip-stick of the Ge detector  
 Crystal Size: External Diam.: 111 mm

Internal Diam.: 28.4 mm

Length: 50 mm

Crystal Thickness: 26 mm

Annulus Diameter: 25.4 mm

Crystal Weight: 1.32 kg

Housing Thickness: 0.8 mm aluminum

FWHM: Better than 25% (661 keV, 137CS)

Notes: The annulus consists of two independent BGO detectors; the half detector consists of two BGO segments, and each segment is equipped with one photomultiplier of one-inch diameter.

### 1.1.3 Shielding of the Ge-BGO Detector System

The Ge-BGO detector system is shielded by low-background lead in thickness of 10 cm.

The inside of the lead shield is lined with low-background oxygen-free copper in thickness of 5 mm and Lucite in thickness of 2 mm.

## 1.2 Arrangement of the Electronic Units

Block diagram of the electronic system of the Ge-BGO Compton suppression gamma-ray spectrometer is presented in Fig. 2.

The input pulse width and anti-coincidence gate pulse width of the anti-coincidence circuit is 0.5  $\mu$ s and 50 ns, respectively.

Shaping time constant of 50 ns has been set for the spectroscopy amplifier of HP Ge detector.

### 1.2.1 Linear/Logic Pulse System of the Ge Detector

Preamplifier: PGT RG IIB/C  
 Spectroscopy Amplifier: Canberra 1413  
 Linear Delay: ORTEC 427 A  
 Pulse Height Adjuster (No. 1 Input of SU111 Amplifier): Canberra 1465A  
 Timing Single Channel Analyzer: ORTEC 551



### 1.2.2 Linear Logic Pulse System or the BGO Detector System

Sum for Each BGO: Three Channel Preamplifier/Line Drivers: Cyberstar (France) EES-VIEEC-V  
Sum Amplifier: Three Channel Sum-Shaping-Base Line Restorer Amplifier (Gaussian 1  $\mu$ s fixed peaking time). Cyberstar CSY-ISA  
Timing Single Channel Analyzer: ORTEC 455  
Gate/Delay Generator: ORTEC 41 6A

### 1.2.3 Logic System for HP Ge and EGO Systems

Anti-Coincidence: ORTEC -114A  
Gate/Delay: ORTEC -110A  
Linear Gate/Stretch: ORTEC 542

### 1.2.4 Multichannel Analyzer and Data Processor

Multichannel Analyzer: Seiko EG&G 7x00 with two Analog-to-Digital Converters (Seiko IX20)  
Data Processor and Output Machines: NEC PC YX01 YX with a Software/Data Base by Seiko. Plotter (Roland) and Line Printer (NEC)

## 2. Measurements of Basic Characteristics of the Ge-BGO Compton Suppression Gamma-ray Spectrometer

The performance of the Ge-BGO Compton suppression gamma-ray spectrometer has been examined by the measurements with the representative parameters characterizing gamma-ray spectrometers such as energy resolution, detection efficiency, peak-to-Compton ratio, peak-to-total ratio, Compton suppression factor, natural background level, etc.

Minimum acceptable radioactivities of the Ge-BGO Compton suppression gamma-ray spectrometer has been evaluated for  $^{137}\text{Cs}$  in terms of the coefficient of variation and measurement time. The change of background level into is taken into account.

## 2.1 Performance of the BGO Detector System

Pulse height distribution of the main, plug and back-catcher BGO detectors have been measured with use of a point source of  $^{137}\text{Cs}$ . In the measurements, the output amplitude of each detector has been adjusted to give almost the same pulse height by controlling high voltage power supply, which is equipped with each detector.

The pulse height distributions of  $^{137}\text{Cs}$  of the main, plug and back-catcher BGO detectors are shown in Fig. 3.

The pulse height distributions of the BGO detector system have been measured by using point sources of  $^{137}\text{Cs}$  and  $^{60}\text{Co}$  as shown in Figs. 4 and 5, and the results of FWHM and peak-to-total ratio are tabled in Table I, together with the results shown in Fig. 3.

Errors in Table I are statistical standard deviation.

The result of Table I clarifies that the peak-to-total ratio of 3 inch diam, x3 inch BGO detector is 1.21 times or that of the same size NaI(Tl) crystal of the same sizes, resulting from its high atomic number and density.

Energy resolution of the BGO detector system becomes poor when the source position in the annulus becomes far from the photomultipliers of main BGO, which is mainly due to the decrease of light collection efficiency at the photocathodes and the increase of attenuation of light in BGO crystal.

## 2.2 Performance of the HP Ge Detector

The certified values of the HP Ge detector by the manufacturer, 0.18 keV of FWHM and 46.1%, of relative efficiency have been satisfied at the initial measurement without meaningful differences.

Three years after the initial measurement, measurements of pulse height distributions with the HP Ge detector as the Ge-BGO system shown in Fig. 2 have been done.

iii the measurements, point sources of

$^{137}\text{Cs}$ ,  $^{60}\text{Co}$  and mixed nuclides of  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$  and  $^{232}\text{Th}$  have been used in both spectroscopy modes of the single (normal) and Compton suppression. The results of the measurements are shown in Figs. (1A, 7A and X). It is apparent that the pulse height distributions of Figs. (1A and 7A are recording the events of scattered gamma-rays from the BGO detector system, which are seen as the increase of the Compton continua like a steps at around the energy of  $U_0$  back scattered gamma-rays.

The pulse height distributions featuring Compton spectra of the Figs. (1A and 7A are shown in Figs. (1B and 7B), in which the energy regions used for averaging Compton continuum needed to calculate peak-to-total ratios are indicated. In particular, Fig. 7B illustrates apparently the single and double escape peaks of the 1332 keV from  $^{60}\text{Co}$  as well as the effective reduction of Compton continuum.

### 2.2.1 Energy Resolution

Measurements of energy resolution of the HP Ge detector for 1332 keV ( $^{60}\text{Co}$ ) and 661 keV ( $^{137}\text{Cs}$ ) have been made with the ADC of conversion gain of 100 channels. The results are shown in Table 2, together with the ratio of FWTM/FWHM.

Any observable difference of FWHM has not been observed between the single and Compton suppression modes.

The result of Table 2 shows that FWHM for 1332 keV is broadened by 1.4 eV compared to the initial value with the simplest electronic arrangement.

The ratios, FWTM/FWHM indicate that the profile of full-energy peak can be fitted with Gaussian distribution quite well, because of that the ratios agree with the theoretical value of  $1.75$  ( $\text{FWTM}/\text{FWHM} = 1.75$ ), where  $\sigma$  is standard deviation).

### 2.2.2 Detection Efficiency

The 25 cm relative efficiency of the HP Ge has been tested, and no appreciable change, i.e. more than 1%, in the efficiency has been observed.

Intending to obtain full-energy peak

efficiency for volume samples, three volume sources containing  $^{137}\text{Cs}$ ,  $^{60}\text{Co}$ ,  $^{40}\text{K}$  and/or  $^{232}\text{Th}$  have been prepared with use of induced and natural radioactivity.

Density of the sample ranges widely from 0.347 g/cm<sup>3</sup> to 1.50 g/cm<sup>3</sup>, depending on the sample.

The diameter and the thickness of the sample is 75 mm and  $34 \pm 1$  mm, respectively depending on the sample. The sample container is made of Lucite of 1 mm in wall thickness.

In the measurements, the samples are attached directly to the detector window (sample-to-window distance, SD: 3 mm) of horizontal type dipstick. Full-energy efficiencies obtained by the measurements with single and Compton suppression modes are listed in Table 3, and the result measured with the single mode is shown in Fig. 1) as a function of gamma-ray energy, in which the correction for coincidence sum effect has not been applied.

In Fig. 9 the error bars are fixed at 11.4%, which are three standard deviations of  $^{137}\text{Cs}$ .

It is seemed to be that the efficiencies of 661 keV and 795 keV from  $^{137}\text{Cs}$  is suppressed, more or less, by coincidence sum effect.

In Table 3 the ratio of the full-energy peak efficiency in the single mode ( $\epsilon_s$ ) to the one in the Compton suppression mode ( $\epsilon_{cs}$ ) is listed.

The ratio for single gamma-ray from  $^{137}\text{Cs}$  and  $^{40}\text{K}$  is unity in both mode, i.e. any change in the efficiency has not been observed.

Contrary to single gamma-ray emitters, the ratios for the gamma-ray emitted from multiple gamma-ray emitters,  $^{137}\text{Cs}$ ,  $^{60}\text{Co}$  and  $^{232}\text{Th}$ , increase drastically far beyond unity, i.e. the full energy peak efficiencies decrease drastically in the Compton suppression mode by the cascade-gamma effect.

The large value of these ratios proves that the sum of the total detection efficiency of the BGO detector system for the

associated gamma-rays with the gamma-ray concerned, i. e. the gamma-rays belonging to the same cascade as the gamma-ray concerned is very high.

From the ratios listed in Table 3, the sum of the total detection efficiency of the BGO detector system for the associated gamma-rays including the scattered gamma-rays of the gamma-rays concerned in the Ge detector can be calculated as 95.1% for  $^{134}\text{Cs}$ , 87.5% for  $^{60}\text{Co}$  and 94.7% for  $^{137}\text{Cs}$  from the following relationship [4]:

$$\epsilon_{\text{BGO}} = 1 - (\epsilon_{\text{S}} / \epsilon_{\text{C}})^{-1}$$

Energy and its abundance of gamma-ray concerned and its associated gamma-rays are summarized in Table 1.

It is clear that the tendency of the degree of the suppression of full-energy peaks in the Compton suppression mode that is represented by  $I_{\text{S}}/I_{\text{C}}$  is the reflection of the gamma-ray energies related to the detection efficiency of the BGO detector system and abundance for each nuclide listed in Table 1.

Dependence of full-energy peak efficiency of the 661 keV gamma-rays from  $^{137}\text{Cs}$  is shown in Fig. 10. In the figure error bars are fixed at the average value of 6.4%, which ranges actually from 5.3% at 0.674 g/cnr to 9% at 1.7X g/cnr.

In this work the efficiency of  $0.020 \pm 5\%$  at 1 g/cnr is employed to evaluate the minimum acceptable activity of  $^{137}\text{Cs}$ .

### 3.3 Peak-to-total Ratio

Peak-to-total ratio of the Ge detector has been measured with a set of point sources prepared by Amersham,  $^{203}\text{Hg}$ ,  $^{54}\text{Mn}$ ,  $^{137}\text{Cs}$ ,  $^{22}\text{Na}$  and  $^{60}\text{Co}$ .

Typical source distance in the measurement is 5cm on the axis of the Ge crystal.

A simple method of extrapolation or interpolation has been used for stripping the peaks generated by K $\alpha$ -rays, annihilation radiation as well as the associated Compton continuum.

The result of Peak-to-total ratio is shown as a function of gamma-ray energy in Fig. 11

in which statistical standard deviation is 0.5% at the maximum.

The result of Fig. 11 shows that peak-to-total ratio of 661 keV-gamma-rays and 835 keV-gamma-rays, both from single gamma-ray emitter, is  $0.281 \pm 0.2\%$  and  $0.250 \pm 0.1\%$ , respectively.

### 2.2. Peak-to-Compton Ratio

The peak-to-Compton ratio is defined as the ratio of the counts/channel in the highest full-energy peak to the counts/channel in a typical channel of the Compton continuum associated with the full-energy peak.

The averaged counts/channel of the energy region of the Compton continuum ranging from 358 keV to 382 keV for the 661 keV gamma-rays from  $^{137}\text{Cs}$  and of the region 1040 keV to 10% for the 1332 keV gamma-rays from  $^{60}\text{Co}$  has been used, respectively.

The energy region for  $^{137}\text{Cs}$  and  $^{60}\text{Co}$  is shown in Fig. 6B and 7B, respectively.

The peak-to-Compton ratios for the gamma-rays of 661 keV ( $^{137}\text{Cs}$ ) and 1332 keV ( $^{60}\text{Co}$ ) have been calculated from the results of Fig. 6A and 7A.

The results with the single and the Compton suppression mode are presented in Table 5.

### 2.1.5 Compton Suppression Factor

The Compton suppression factor of the Ge-BGO gamma-ray spectrometer has been determined from the results of Figs. 6A and 7A.

The Compton suppression factor is defined as: counts/channel of the single mode divided by counts/channel of the Compton suppression mode.

The results are shown in Figs. 12 and 13, in which the method of moving averages of six data points has been applied, and the factor in the region of full-energy peaks has been omitted.

Figure 12 shows that the Compton suppression factor at the Compton edge of the 661 keV gamma-rays from  $^{137}\text{Cs}$  is 10.4 and the factor averaged over the energy



region used for determining Compton counts for the peak-to-total ratio is 0.2 on the average.

Scattering angle and the corresponding energy of scattered gamma-rays in the Ge detector have been inserted in Fig. 12.

Figure 13 shows that the suppression factor at the Compton edge of the 1332 keV gamma-rays from  $^{60}\text{Co}$  and the factor in the energy region for the average Compton counts is almost the same value of 10.1. The factor at the Compton edge of the 1120 keV gamma-rays is the maximum value of 10.7.

Behavior on Compton suppression factor of the Ge-BGO spectrometer can be summarized as the followings:

- Appreciably large value of the Compton suppression factor around Compton edge proves that the plug and main BGO detectors work effectively for back-scattered gamma-rays from the Ge detector. Compton suppression factor around Compton edge in Fig. 12 proves that the detection efficiency of the BGO detector system for scattered gamma-rays of energy around 200 keV is more than 100%.
- The suppression factor at around 100 keV back scattered gamma-rays from the plug BGO detector is smaller compared to other region. This depletion of the factor is seemed to be caused by the back scattered gamma-rays by housing material of the BGO detector, which can not be detected by the plug BGO detector.
- The suppression factor of a little more than four at a smaller scattered angle proves that the back-catcher BGO detector seems to be quite effective. This fact becomes more clearly in Fig. 13 obtained with  $^{60}\text{Co}$ . In this figure the energies of scattered gamma-rays from  $^{60}\text{Co}$  become higher than that from  $^{137}\text{Cs}$ .
- It is noticeable in Fig. 12 and 13 that the suppression factor of the region between the full-energy peak and the Compton edge that are resulted mainly in coincidence sum effect have also been suppressed rather effectively. This fact suggests that the BGO detector system has

an appreciable detection efficiency for multiple-scattered gamma-rays. The suppression factor in the pulse height region decreases with increasing the pulse height. Because of that the energy of multiple-scattered gamma-rays decreases with increasing the pulse height of coincidence-sum events. In other words, the suppression factor in the region of pulse height decreases with increasing the pulse height of coincidence-sum events, since the shielding effect by detector housing against multiple-scattered gamma-rays increases with increasing the pulse height of coincidence-sum events.

### 3. Natural Background Level of the Ge-BGO Compton Suppression Gamma-ray Spectrometer

Natural background level of the gamma-ray spectrometer has been measured with the single and the Compton suppression mode, and the results are shown in Fig. 14.

The pulse height distribution with the single mode in Fig. 14 is multiplied by 100 for distinguishing both spectra easily.

Peak area of the main peaks observed in Fig. 1-1 is listed in Table ().

The result of Table () can be summarized as follow:

- In spite of full-energy peaks formed by cascaded gamma-rays such as  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$ , the suppression of peak area is not appreciable, because of that the solid angle of naturally occurring radionuclides subtended by the Ge detector is very small.
- Contrary to the above, the full-energy peaks of  $^{207}\text{Pb}$  are strongly suppressed with Compton suppression mode, because of that  $^{207}\text{Pb}$  is contaminant of the BGO crystal and the total detection efficiency of the BGO detector system is very high. The ratio of the peak area of the single mode to the Compton suppression mode, 10.5 indicates that the peak area of the 100 keV is reduced to 0.095 by 10.5, of the total detection efficiency of the BGO detector

system for the main associated gamma-rays of 569.7 keV.

- The ratio of peak area of the doublet of the 511 keV, 7.86 is considerably larger compared to 1.77 of the 261.4 keV gamma-rays from <sup>214</sup>Pb. This result shows that the peak area of 511 keV with the single mode is formed mostly by annihilation radiation generated in/or near the BGO detector. The 511 keV peak and the single/double escape peaks corresponding to the 261.4 keV from <sup>214</sup>Pb have not been observed at all by the measurement with <sup>228</sup>Th source in the Compton suppression mode as shown in Fig. 8. Because of that the detection efficiency of the BGO detector system surrounding the Ge detector with almost 4π is very high (>95%) for annihilation radiation.

Compton suppression factor for natural background obtained from the result of Fig. 14 is shown in Fig. 15, in which the method of moving averages of six data points has been applied and no interpolation in full-energy peaks has been applied for estimating the suppression factor.

The Compton suppression factor shown in Fig. 15 is divided into two groups bordering the energy of the 140 keV from <sup>137</sup>Cs.

In the lower pulse height group, the maximum and the minimum suppression factor is expected to be about 7 at around 1250 keV (830 channel) and about 3 at around 250 keV (250 channel), respectively.

In the higher pulse height group, the maximum and the minimum is about 10 at around 1500 keV (2650 channel) and about 4 at around 1460 keV (2110 channel), respectively.

Background levels of the energy region of the 661 keV from <sup>137</sup>Cs have been obtained from the results of Fig. 14 as 7.83 ± 0.1 (cps/keV) ± 3.4%, in the single mode and 1.31 ± 0.1 cps/keV ± 8.4% in the Compton suppression mode, resulting in the suppression factor of (0.0 ± 6.1%).

### 1. Evaluation of the Minimum Acceptable Radioactivity of <sup>137</sup>Cs

In the measurement of very weak radioactive samples like environmental samples, it is principal to grasp minimum acceptable radioactivity of spectrometers.

If minimum acceptable radioactivity of spectrometers is known in terms of main parameters characterizing spectrometers (energy resolution and detection efficiency, etc.) and in terms of measurement conditions (measurement time, background level and an acceptable statistical accuracy, etc.), it is very useful to design experiments and to improve the quality of measurement results.

In this work minimum acceptable radioactivity of <sup>137</sup>Cs, AM has been evaluated by the method proposed by G. Y. Walford et al. (6) and I. A. Cooper et al.

The evaluation method expresses the AM as the following relationship:

$$A_{min} = \frac{1}{2 \cdot t \cdot (E) P_r t} \left( 1 + \frac{1}{1 + 8C^2 hR(E) n n_1 + 4C^2 hR(E) n_2} \right)$$

where the C: the acceptable coefficient of variation, which defines the precision of peak area:

$$C = \frac{\text{standard deviation} (\sim \text{net peak area})}{\text{net peak area}}$$

the  $\epsilon(E)$ : full-energy peak efficiency at a gamma-ray energy E [keV].

the  $P_r$  gamma-ray emission probability (abundance) [y/decay].

the t: measurement time [second].

the  $R(E)$ : energy resolution, FWHM [keV].

the  $\mu_{ROI}$ : average background counting-rate of the region of interest (ROI) in pulse height distribution [counts/kev].

the  $n_1, n_2$ : additional counting rate averaged over ROI, which is accompanied with the



evaluation of background in ROI [counts/kev].  
 the  $h$ : the factor which when multiplied by the energy resolution  $R(i,j)$  FWHM I, gives the region (width) of the ROI

The minimum acceptable radioactivity of  $^{137}\text{Cs}$ ,  $M$  has been done by the above relationship as a function of relative error,  $\epsilon$  and measurement time,  $t$  taken background level,  $B$  into account.

In the evaluation the following instrumental and additional parameters have been used.

FWHM (MeV keV): 1.51 keV

Full-energy peak efficiency: 0.0208  
 (volume source: 75 mm diam. x 74 mm source-distance: 1 mm)

Gamma-ray emission probability of the 661 keV gamma-rays from  $^{137}\text{Cs}$ : (0.85)

Background counting-rate of the ROI:  $7.88 \times 10^{-1}$  (counts/kev) in single mode.

$1.21 \times 10^{-4}$  (counts/kev) in Compton suppression mode

the constant to define the width of the ROI,  $h$ : 2

In usual low activity measurements, especially in many cases of the measurement of environmental sample, additional natural radioactivity associated with the samples is, more or less, introduced inside the detector shield, so that the sensitivity is decreased.

Table 7 presents background counting-rates at the energy region of the 661 keV from  $^{137}\text{Cs}$  with typical environmental samples, together with the result of Fig. 14 without any sample. The result of Table 7 is obtained with measurement time of 200 s to 800 seconds, depending on the sample.

Table 7 shows that the background level of the non-sample and the sea plant is comparable, and also the soil sample and the lake sediment is comparable. Therefore, the calculations of the minimum acceptable radioactivity of  $^{137}\text{Cs}$  based on the background levels of non-sample and the soil sample have been made as a function of the coefficient of variation. In the calculations, measurement time of  $X$  hours and  $ISX$ , of background counting-rate as the  $B$ , have

been used.

The results of the above calculations are shown in Fig. 10(i).

When the maximum acceptable error is assumed to be 10%, the minimum acceptable activity of the Ge-8GO Compton suppression gamma-ray spectrometer for  $^{137}\text{Cs}$  are resulted in the activity listed in Table 8.

Regarding the result of Table 8, it is preferable to take the acceptable coefficient of variation (relative error) less than 0.3 (30%). It is widely recognized that the existence of the peak having an error more than 30% in peak area is uncertain (6, 7).

The minimum acceptable activity of  $^{137}\text{Cs}$  has been calculated as a function of measurement time under the same conditions as Fig. 10. The results calculated with the coefficient of variation of 0.1 and 0.3 are shown in Fig. 10(i) and Fig. 17, respectively.

The result of Table 8 clarify that the Ge-8GO Compton suppression gamma-ray spectrometer improves the sensitivity of detection of  $^{137}\text{Cs}$  activity about twice compared to the single spectrometer. The additional background introduced by the soil sample makes poor the sensitivity about 1.5 times compared to the sensitivity without the soil sample.

The minimum acceptable activities shown in Figs. 17 and 18 are summarized in Table 9, in which measurement times of 8 hours, one day and 2 days are combined with each coefficient of variation, 0.1 and 0.3.

#### Conclusion

A HP Ge Compton suppression gamma-ray spectrometer has been constructed with a BGO scintillation detector system as a Compton shield detector.

Energy resolution (FWHM) of the HP Ge and 25 eV relative full-energy peak efficiency is 1.97 keV and 40% for the 1332 keV gamma-rays from  $^{60}\text{Co}$ , respectively.

The BGO detector system consists of three detectors, main (annulus type, 183.2 mm diam. x 200 mm annulus diameter: 83.2

nun). plug (cylindrical) t~pc. 7(2) IIUI diam ~7(0.2 mill and back-catcher (annulus type, X(0)-1 nun diam. x50 nun. annulus diameter: 2X.-lmJ11).

Energy resolution (FWHM) and peak-to-total ratio of the BGO detector system is 19%, and OX5%, for the 00 I keV gamma-rays from IrCs.

Total detection efficiency of the BGO detector system for the 1173 keV gamma-rays from <sup>137</sup>Cs and scattered gamma-rays of the Lr12 keV in Ge crystal is 8X% in total.

Peak-to-Compton ratio of the Ge-BGO Compton suppression gamma-ray spectrometer has been improved from 12R to 7X(0) with the Compton suppression spectroscopy in the case of the 061 keV gamma-rays.

Natural background level at the full-energy peak region of the 00 I keV gamma-rays has been lowered from 7.9 x 10<sup>4</sup> cps/keV to 1.2x(rl<sup>4</sup>) cps/keV with the Compton suppression spectroscopy.

With the Compton suppression spectroscopy the minimum acceptable radioactivity of <sup>137</sup>Cs radioactivity has been improved from 0.091 Bq to 0.044 Bq under the conditions of measurement time of 8 hours and the coefficient of variation of 3(0)%.

Under the additional natural background introduced by IX-1 g of soil (2x 10<sup>3</sup> cps/keV in the single spectroscopy), the minimum acceptable radioactivity of IrCs has been improved from 0.1-1 Bq to 0.066 Bq under the same parameters as the above.

The ultimate minimum acceptable radioactivity of <sup>137</sup>Cs can be actually concluded as 0.015 Bq that is obtained under the conditions of 3(0)X, or the coefficient of variation, measurement time of 2 days and non-additional background.

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Table 1 The result of the measurements of characteristics of the BGO detectors and the BGO detector system

	FWHM (661 keV: J:17Cs)	Peak-to-total Ratio (661 keV: 137CS)	Source position
Main BGO	18.3"	0.811±0.1670	Center of annulus
PlugBGO	12.7"	0.647±0.08%#	15 em from BGO
Back-catcher BGO	25.0"	0.461±0.100"	5 ern above BGO
BGO detector system	14.9"	0.830±0.10"	Center of annulus
BGO detector system	19.1"	0.848±0.07"	3 em from Ge, 2 em from plug BGO
BGO detector system	10.6" (2505 keV: sum of =C <sub>K</sub> )	0.279±0.09)" (2505 keV: sum of <sup>60</sup> Co)	Center of annulus
BGO detector system	12.1" (2505 keV: sum of <sup>60</sup> Co)	0.273±0.16" (2505 keV: sum oPOCo)	3 ern from Ge, 2 cm from plug BGO

# 0.534 for 3 inch diam. x3 inch NaI(Tl) crystal (3)

Table 2 Energy resolution of the HP Ge gamma-ray spectrometer

	FWHM (keV)	FWTM (keV)	FWTM/FWHM*
1332 keV (60CO)	1,97	3,65	1,86
661 keV (1:17CS)	1,51	2,79	1,85

\*: the theoretical value: 1.83

Table 3 Full-energy peak efficiency of the HP Ge detector for volume source (Sample:75 mm diam.x34±1 mm, SD: 3 mm)

Nuclide	Energy (keV)	Efficiency in Single Mode, ES	Efficiency in Compt. Sup. Mode, eo	eslec.	Density (g/cm <sup>3</sup> )
1:17CS	661	0.0208±2.5%	0.0208±2.5%	1,00	0.674
1:1~CS	795	0.0158±3.8%	0.00073±18%	21,7	0.674
ti <sup>o</sup> Co	1332	0.0129±5.4%	0.00161±5.4%	8.01	0.347
-oK	1461	0.0113±4.2'10	0.0113±4.2%	1,00	0.674
2 <sup>o</sup> ~TI	2614	0.00614±1.0'0%	0.000326±3.6%	18.8	1,50



Table 1 Energy and abundances of the gamma-rays concerned and the associated gamma-rays

Nuclide	Gamma-ray concerned		Gamma-rays associated	
	Energy (keV)	Abundance (%)	Energy (keV)	Abundance (%)
137Cs	661.5	84.6	647.1	1.4
			604.0	17.0
208Tl	2614.5	100	1120.3	88.4
			277.5	6.8
			511.0	21.6
			583.0	86.0
			860.0	12.0

Table 5 Peak-to-Compton ratio for 661 keV (137Cs) and 1120 keV (208Tl) Gamma-rays

Energy (keV)	Nuclide	Mode	Peak-to-Compton Ratio	Channel Width (keV)
661.5	137Cs	Single	128	0.20G
		Compt. Sup.	78	
1120.3	208Tl	Single	5D	0.11H
		Compt. Sup.	11(-)	

Table 6 Peak area of main peaks in natural background spectrum measured with single and Compton suppression mode

Energy (keV)	Nuclide	Peak Area (c/s)		Ratio of Single/Compt. Sup. *
		Single	Compton Sup.	
186	214Pb	0.007BR ± 6.2%	0.00805 ± 4.0%	0.9 ± 7.4%
214	214Pb	0.0122 ± 4.1%	0.0119 ± 3.0%	1.0 ± 5.2%
511	Annihilation	0.00477 ± 8.1%	0.00441 ± 5.1%	1.06 ± 9.9%
511	Annihilation	0.0164 ± 1.1%	0.0019G ± 1.0%	7.86 ± 0.5%
214	214Pb	0.0101 ± 2.1%	0.0094D ± 0.6%	8.0 ± 0.0%
214	214Pb	0.0047 ± 1.9%	0.0047 ± 5.0%	2.00 ± 8.3%
214	214Pb	0.0048 ± 1.1%	0.0041 ± 4.8%	1.1 ± 7.1%
220	220Ac	0.00127 ± 7.7%	0.0010 ± 1.1%	1.07 ± 0.2%
214	214Pb	0.011 ± 1.0%	0.0006 ± 1.8%	1.8 ± 1.1%
214	214Pb	0.001 ± 18.0%	0.001 ± 8.1%	0.1 ± 0.3%
214	214Pb	0.0484 ± 1.1%	0.041 ± 1.0%	1.02 ± 1.1%
214	214Pb	0.001 ± 4.1%	0.001 ± 5.2%	1.77 ± 0.7%

\* = Ratio of peak area of single mode to Compton suppression mode  
 ± = Doublet (or; interfered by the neighboring peak)

Table 7 Background counting-rate at the full-energy peak region of the 06 I keV gamma-rays from  $^{137}\text{Cs}$  measured with the Ge-BGO gamma-ray spectrometer

Sample	Background Counting-rate (cps/keV)		Remarks
	Single Mode	Compt. Sup. Mode	
None	$7.58 \times 10^{-4} \pm 8.0\%$	$1.23 \times 10^{-4} \pm 2.0\%$	
Soil	$1.99 \times 10^{-4} \pm 5.0\%$	$3.0 \times 10^{-4} \pm 12\%$	75 diam, $\times 33$ mm 184.2 g, 1.26 g/cm <sup>3</sup>
Seaplant	$8.0 \times 10^{-4} \pm 100\%$	$1.48 \times 10^{-4} \pm 29\%$	75 diam, $\times 32$ mm 91.34 g, 0.646 g/cm <sup>3</sup>
Lake Sediment	$1.1 \times 10^{-4} \pm 9.3\%$	$2.19 \times 10^{-4} \pm 24\%$	75 diam, $\times 35$ mm 275.4 g, 1.78 g/cm <sup>3</sup>

Table 8 Minimum acceptable radioactivity for  $^{137}\text{Cs}$  of the Ge-BGO Compton suppression gamma-ray spectrometer (Relative error: 30%. Measurement Time: 8 hours)

Sample	Single Mode	Compton Suppression Mode
None	0.091 Bq	0.044 Bq
Soil	0.14 Bq	0.006 Bq

Table 9 Minimum acceptable radioactivity for  $^{137}\text{Cs}$  of the Ge-BGO Compton suppression gamma-ray spectrometer with measurement time and the coefficient of variation

Sample	Measurement Time	Single Mode		Compton Suppression Mode	
		Coefficient of Variation		Coefficient of Variation	
		0.1	0.3	0.1	0.3
None	8 hours	0.35	0.091	0.23	(1,044)
	1 day	0.17	0.049	0.016	0.022
	2 days	0.11	(1,021)	(1,021)	(1,015)
Soil	8 hours	0.09	0.14	0.29	0.066
	1 day	0.25	0.077	0.11	0.035
	2 days	0.17	0.053	0.085	0.024

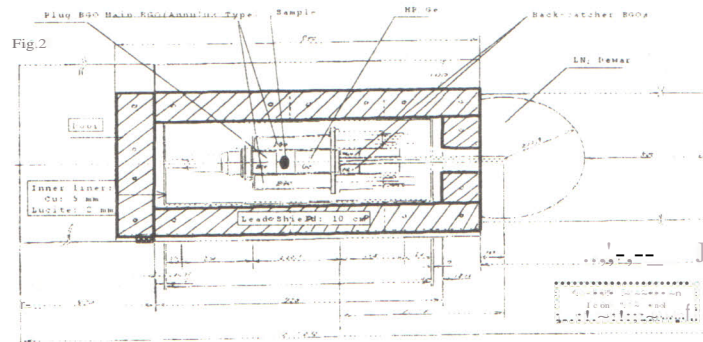


Fig. 1. Assembly of the HP Ge detector with the BGO shield located between the HP Ge and the LN Dewar. The HP Ge detector is a 10 cm long, 10 cm diameter, 10 cm diameter.

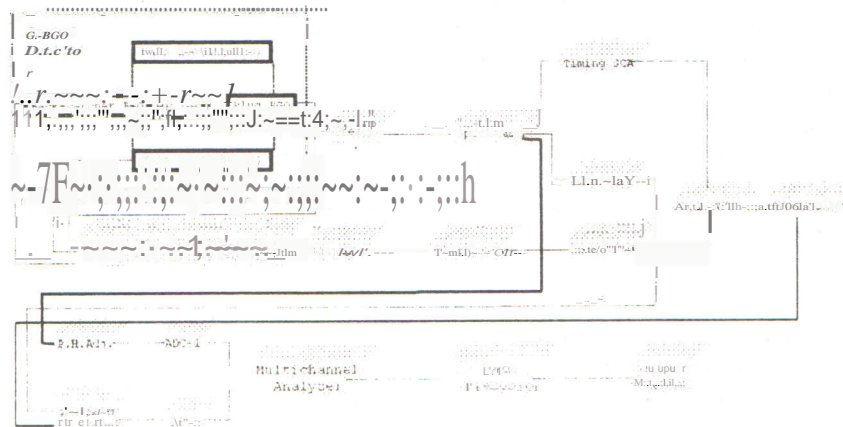


Fig. 2. Block diagram of the data acquisition system of the G-BGO detector. The HP Ge detector is a 10 cm long, 10 cm diameter, 10 cm diameter.

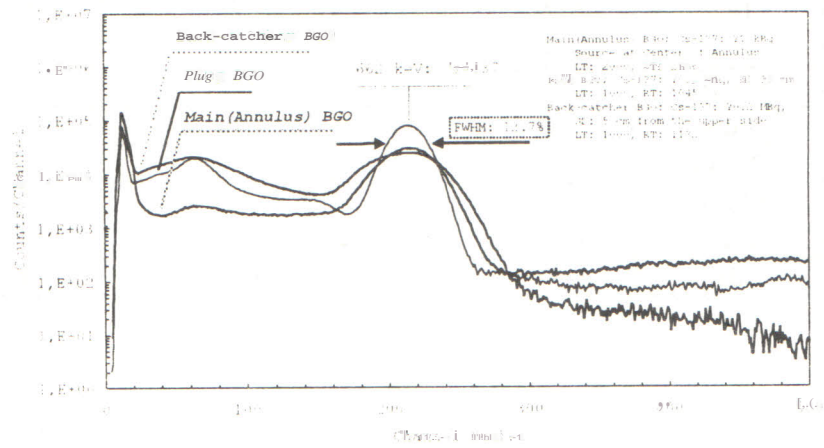


Fig. 3. Pulse-height distributions of Cs-137 by the Main, Plug and back-catcher BGO Compton shield Detector.



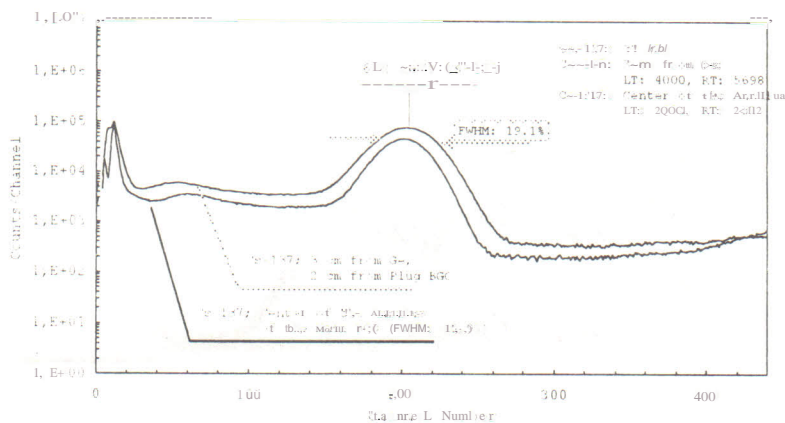


Fig. 1. Spectrum of the distribution of Cs-137 with the Cs-137 Detector. The spectrum is the result of the Main, Fluor, and Background (BGR) at two different source positions.

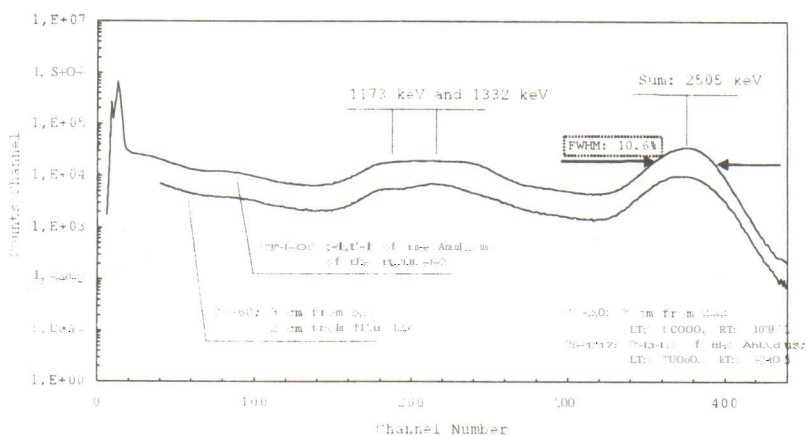


Fig. 2. Spectrum of the distribution of Cs-137 with the Cs-137 Detector. The spectrum is the result of the Main, Fluor, and Background (BGR) at two different source positions.

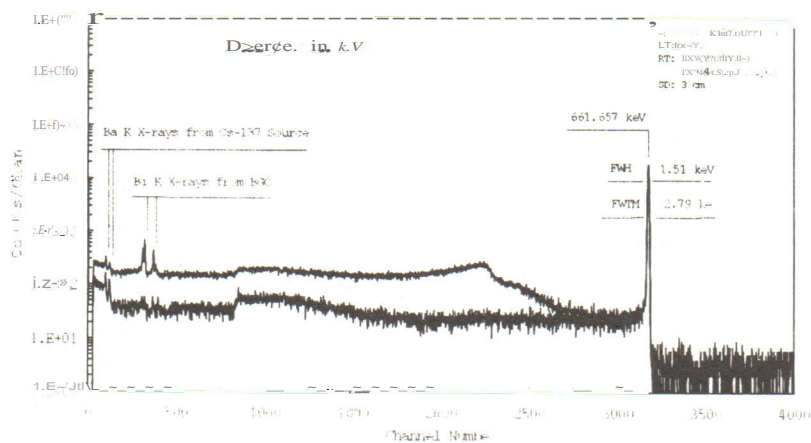


Fig. 3. Spectrum of the distribution of Cs-137 with the Cs-137 Detector. The spectrum is the result of the Main, Fluor, and Background (BGR) at two different source positions.

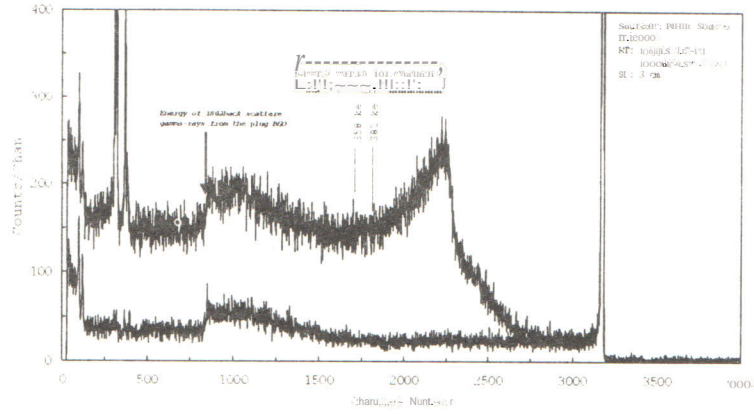


FIG. 7. Gamma-ray spectrum of Co-60 with and without Compton suppression. Apparatus: (1) Genwin-rev, (2) JEOL GEM-400 Compton, (3) JEOL GEM-400 Compton, (4) JEOL GEM-400 Compton.

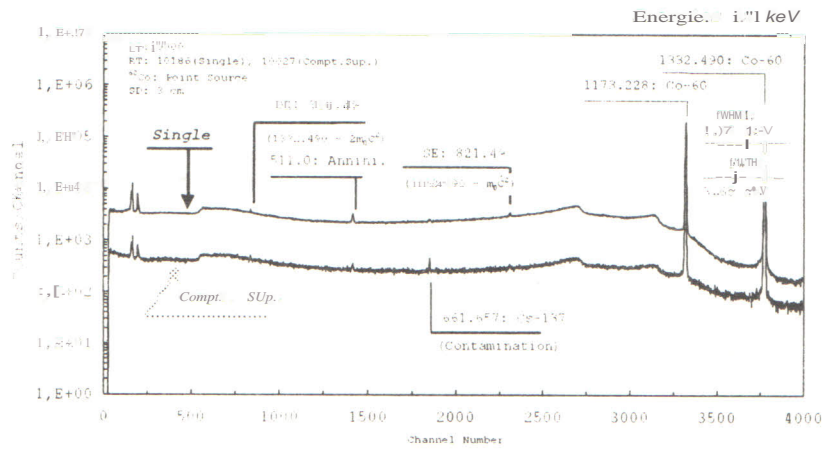


FIG. 7A. Pulse Height Distribution of Co-60 gamma-rays with and without Compton suppression. Apparatus: (1) Genwin-rev, (2) JEOL GEM-400 Compton, (3) JEOL GEM-400 Compton, (4) JEOL GEM-400 Compton.

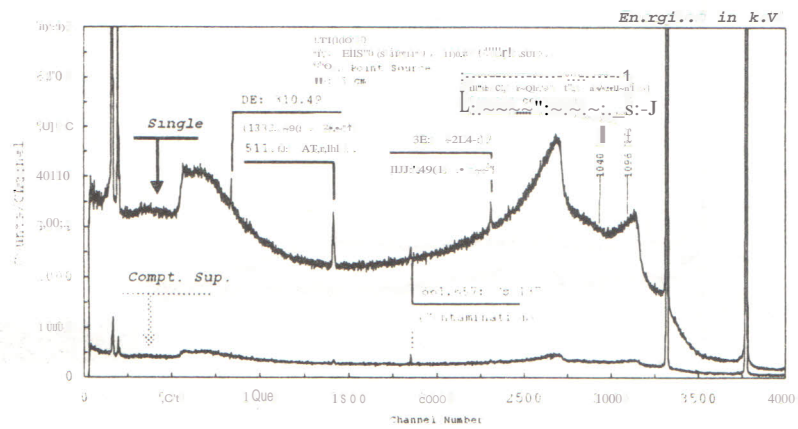
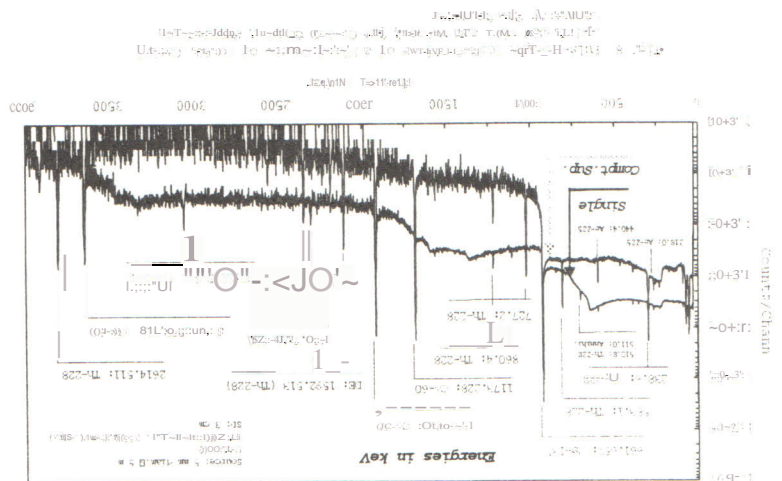
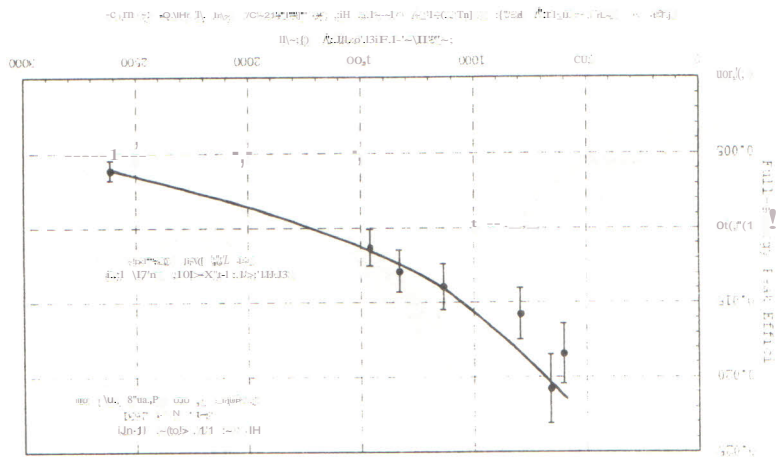
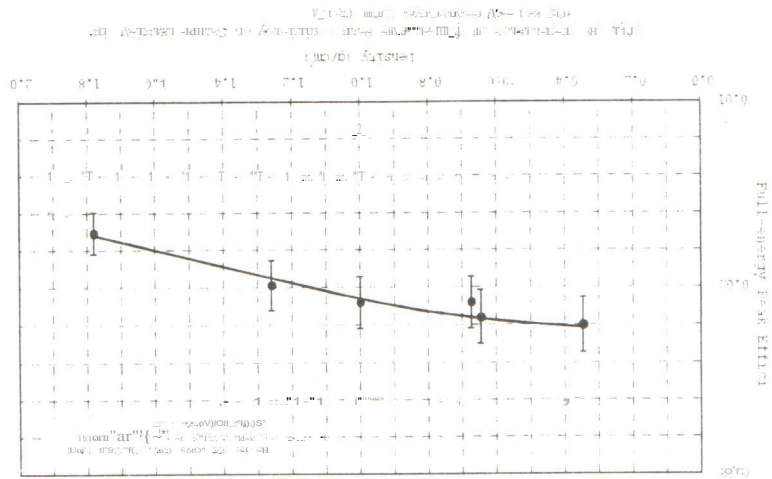


FIG. 1E. Compton Spectrum of Co-60 gamma-rays with and without Compton suppression. Apparatus: (1) Genwin-rev, (2) JEOL GEM-400 Compton, (3) JEOL GEM-400 Compton, (4) JEOL GEM-400 Compton.





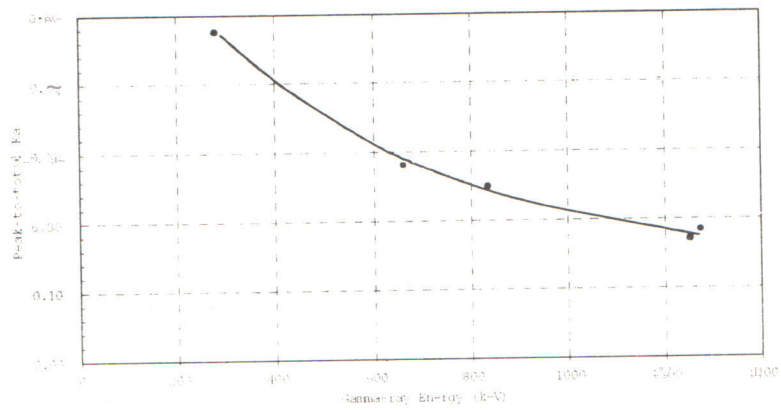


Fig. 11 Scatter count ratio of Ba HPGe detector

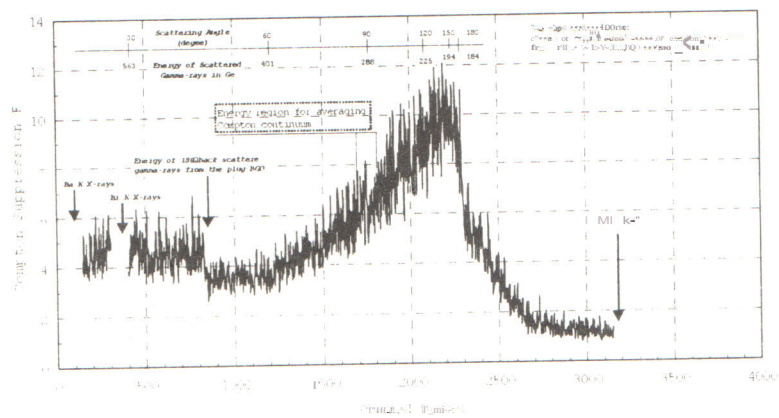


Fig. 12 Total Ba suppression factor of the BaKAl detector against angles for the scattered gamma rays from  $^{137}\text{Cs}$

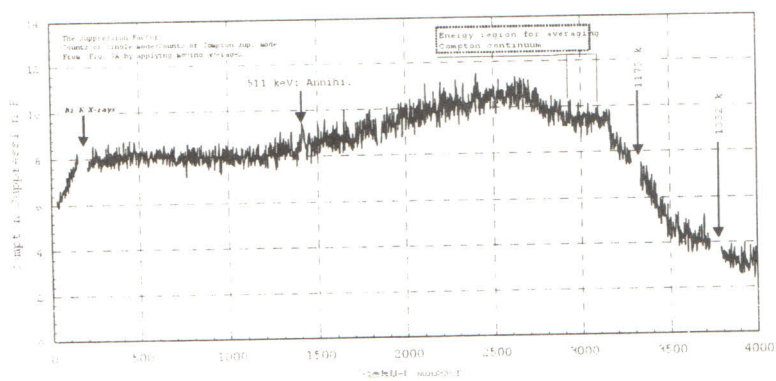


Fig. 13 Total Ba suppression factor of the BaKAl detector against angles for the scattered gamma rays from  $^{137}\text{Cs}$

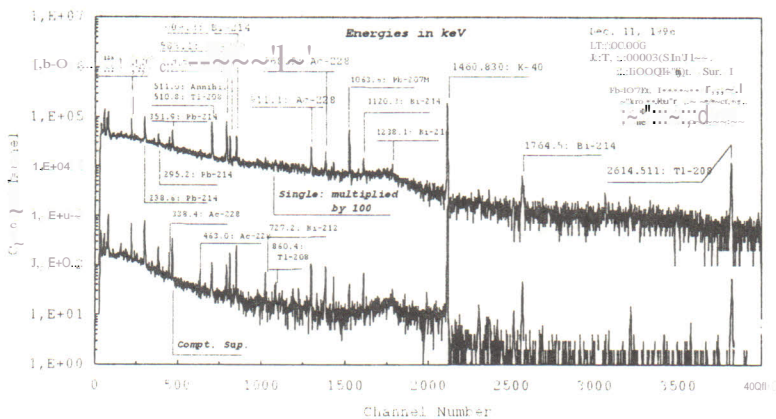


Fig. 14. Pulse Height Distributions of Natural Background with and with out the Compton-suppression Gamma-ray spectrometer

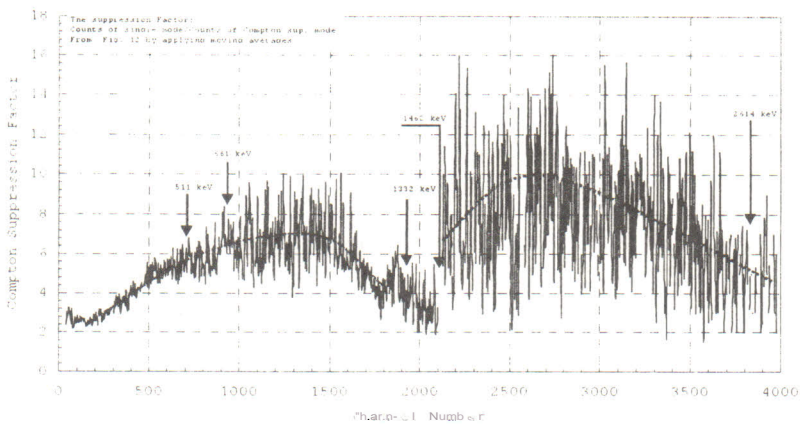


Fig. 15. Compton Suppression Factor versus Channel Number for Natural Background

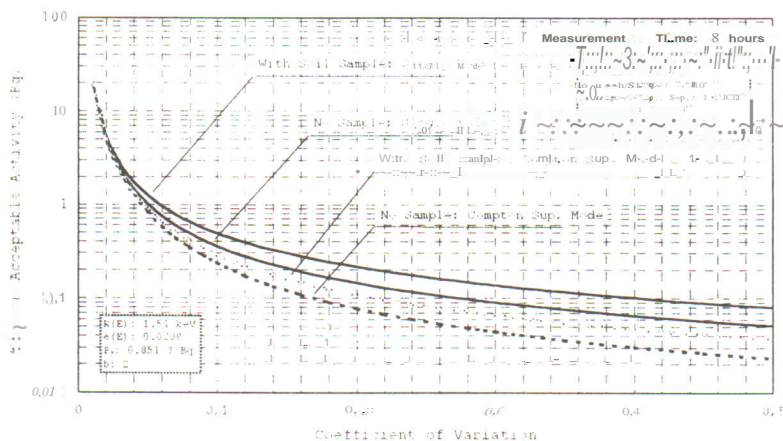


Fig. 16. Maximum Acceptable Coefficient of Variation of 80-90% as a Function of the Coefficient of Variation (Measurement Time = 8 hours)

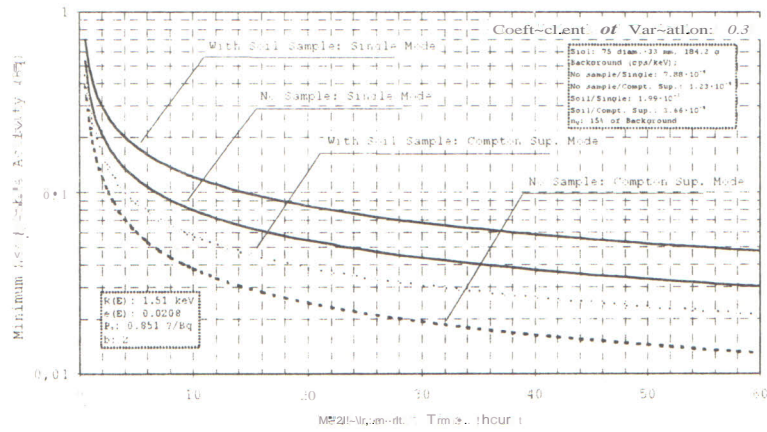


Fig. 15 Manganese-54 Minimum detectable activity (Bq) vs a function of Measurement Time (Base coefficient of Variation: 0.3)

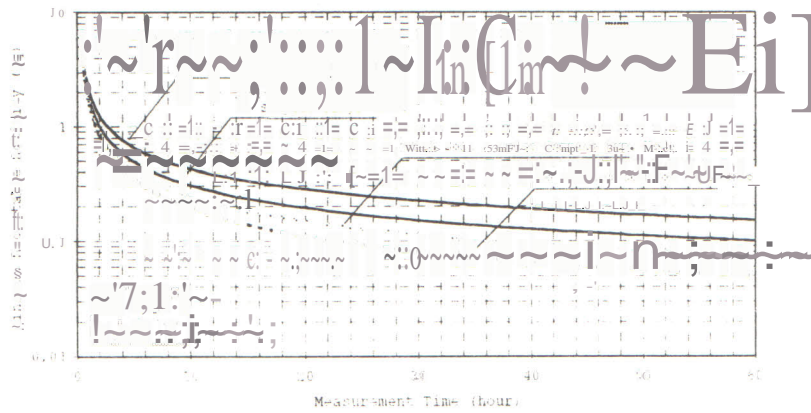


Fig. 16 Manganese-54 Minimum detectable activity (Bq) vs a function of Measurement Time (Base coefficient of variation: 0.1)