The Characteristics of the Ge(Li) detector Installed Kanazawa University and Its Applicatione to Environmental problems

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The Characteristics of the Ge (Li) Detector Installed in Kanazawa University and Its Applications to Environmental Problems

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Abstract The characteristics of the Ge (Li) detector installed in Kanazawa University was examined in regard to its energy resolution, the absolute counting efficiency at various gamma-ray energy and the back ground by shielding with lead. It was found that this detector was more useful than the conventional NaI(Tl) detector for the environmental low level radioactivity measurements.

Some applications to radioactivity pollution problems were made, namely, the quantitative analysis (nondestructive) of several radionuclides introduced into environments owing to either the recent operation of nuclear power plant at Tsuruga or the atomic bomb explosion in Nagasaki in 1945.

1. Introduction

Recently, gamma-ray spectroscopy has become to be more and more useful for the determination of radioisotopes by the development of high resolution lithium-drifted germanium detector, usually expressed as Ge(Li). Though the most Ge(Li) detectors now used in Japan have been imported from U.S.A., the Ge(Li) detector installed in the central radioisotope laboratory of Kanazawa University was fabricated in 1970 by Dr. Y. Ishizuka in Yokohama National University on our request. This detector is a p-i-n junction coaxial type and mounted in a dewar vessel of gravity feed type cryostat made by Tōrisha Co. Ltd. This detector system is schematically shown in Fig. 1. Since both the method of fabrication of Ge (Li) detector and the theory of its gamma-ray detection have been described in other papers[1)-3)], these are omitted in this paper. In this work, the study of the performance of this detector was carried out as to the various characteristics, such as energy resolution (FWHM)* and absolute peak efficiency, for the purpose of the effective utilization in various fields. And some examples of the applications to

* Full Width at Half Maximum (Peak hight)

the quantitative analysis of environmental radionuclides are reported in this paper.

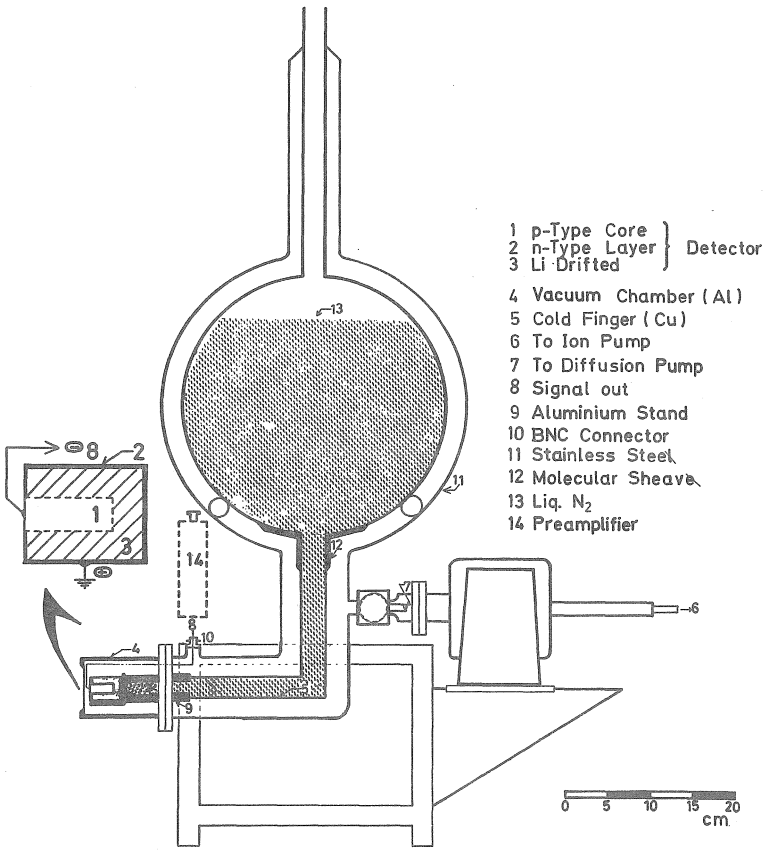


Fig. 1 Schematic illustration of the Ge (Li) detector and its cryostat

2. Instrumentation

The outer size of germanium crystal used in this detector had the diameter of 28 mm. The diameter of P core was 10 mm, the thickness of Li^+ layer (n side) being 1mm. The active volume was estimated to be about 18~20 cm^3, the capacitance being calcuated to be about 40 pF[4]. The cooling of this detector was made by liquid nitrogen in the gravity feed type cryostat as shown in Fig. 1. The electronic pulse from the detector was led to the preamplifier whose type was either an ORTEC 109A or an ORTEC 120-3F. The latter preamplifier was more suitable to this detector than the former, as seen from the followiog results in regard to the resolving power. The detector bias was supplied from Ōsaka Denpa MPS-1282 unit. The pulse was amplified by the linear amplifier of Ōsaka Denpa MPS-1203 unit and then the pulse height analysis was made by Tōshiba model EDS-34208A 200 channel pulse height analyser. The block diagrams of such electronics are shown in Fig. 2.

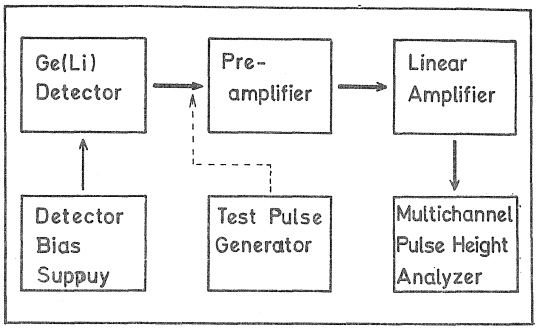


Fig. 2 Block diagram of the electronic components

3. Energy resolution

One of the basic characteristics of the Ge(Li) detector is its energy resolution, usually expressed by FWHM (full width at half maximum of peak height). The interactions of gamma-ray quantum with depletion layer of Ge(Li) detector result in the production of a number of electron-hole pairs. The charge collection depends on the electric field strength in depletion layer. If the applied bias voltage is low, the electron-hole pairs will not be thoroughly collected on both electrodes and the tailing of spectrum to lower energy side occurs. However, such tailing effect decreases according to the increase of the bias voltage as shown in Fig. 3, ORTEC 120-3F preamplifier being used in this case.

The dependence of FWHM upon detector bias voltage was studied both by ORTEC 109A and by 120-3F preamplifier. As seen in Fig. 4 (up to the 1000 volt bias voltage), the best resolution (FWHM) at 661.6 KeV of ^{137}Cs line was about 10 KeV by 109A and 5 KeV by 120-3F, respectively. At 1173 KeV and 1332 KeV of ^{60}Co gamma-ray, the minimum FWHM was about 12 KeV by 109A for both ^{60}Co lines and was about 6.5 KeV and 7.2 KeV by 120-3F for two above-mentioned ^{60}Co lines, respectively. It is evident from these results that the preamplifier used in the first amplifying stage is one of the critical factors for the resolution of gamma-ray energy.

4. Counting efficiency

For quantitative analysis by Ge(Li) gamma-ray spectrometry, it is essential to know the absolute efficiency of each full energy peak, namely the ratio of the peak counting rate to the emission rate of a definite energy gamma-ray. In order to know the peak efficiencies at various gamma-ray energy, the two methods, the relative and absolute calibration techniques, were applied and the disintegration rate of a radioactive nuclide in complex mixture was calculated from the measured peak intensity by using this absolute efficiency.

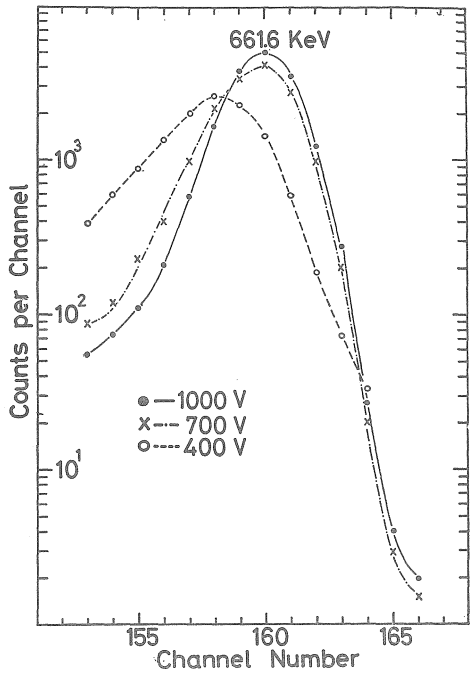


Fig. 3 The tailing effect of the Cs-137 peak as a function of applied bias voltage

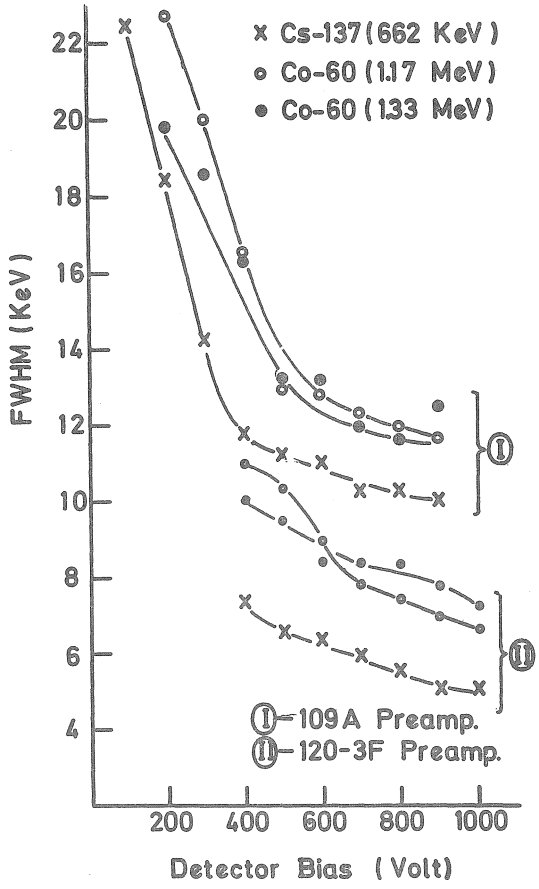


Fig. 4 The change of energy resolution (FWHM) with applied bias voltage by two different preamplifier

For relative calibration of peak efficiency, both a point source of Eu-152 and a check piece of Ra-226 with its daughter nuclides were used to estimate the relative change of counting efficiency with γ ray energy. The γ ray spectra of these source are shown in Fig. 5(a), (b) and the relative efficiency of each peak was calculated from its intensity in referring to the decay characteristics of these nuclids.

In the absolute calibration method, by using a standard source containing a known amount of a radioactive nuclide, the counting rate of a peak was compared with the disintegration rate of this nuclide. The accuracy of the absolute efficiency depends on the reproducibility of the source-detector geometry, the peak area chosen for measurement and the accuracy of the content of the standard radioactive nuclide. The full energy peak area was estimated by considering the background interpolated linearly from both sides of a peak.

In this study, the standard samples issued from IAEA (International Atomic Energy Agency) were used for the absolute calibration. These standards were the powdered sample of uranium ore, namely, Torbernite S2 (U_3O_8=0.313%), Carnotite S3 (U_3O_8= 0.418%) and Uraninite S4 (U_3O_8=0.375%). The following sample containers were used for

measurement, a commercially available weighing glass bottle of 5 cc, a plastic container of 4 cm diameter and 1.5 cm height and another large plastic container of 7 cm diameter and 2.5 cm height. In the glass weighing bottle, the standards samples of various weights ((a) 1.6 gr, (b) 4 gr, (d) 16 gr, (e) 17 gr) were prepared for measurement. The weights of the sample packed in the above-mentioned plastic containers were about 15 gr (c) for the small container and about 100 gr (f) for the large container, respectively.

On account of the bottom surface area of each container, the samples in plastic containers ((c) and (f)) were measured at the distance of about 0.7 cm from the aluminium wall of the detector head, though the samples in the glass bottles ((a), (b), (d), (e)) could be measured in close contact with this wall.

The results in regard to the relative and absolute peak efficiency are summarized in Fig. 6 which shows the decrease of the absolute efficiency with the increase of γ ray energy. In Fig. 6, the change of the counting efficiency with the distance from the center of the detector head in transverse or vertical direction surface are also shown according to the results obtained by the radioactive point source of Eu-152. The change was larger for low energy γ ray than for higher energy γ ray, on account of the absorption of γ ray.

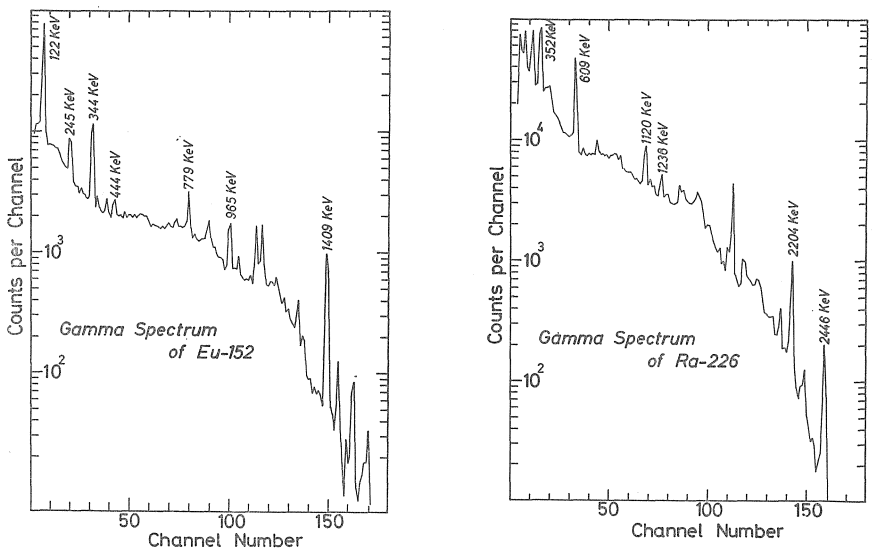


Fig. 5 The gamma-ray spectra of the gamma-ray sources, Eu-152(a) and Ra-226 with its daughter nuclides in radioactive equilibrium (b), used for relative efficiency calibration

5. Contributions of natural back grounds

Since the radioactivity levels of some environmental samples were of the same order with that of natural background without shield, the lead shield was formed around the detector head to minimize the natural background by piling up several lead blocks of 5 cm thickness as shown in the photograph of Fig. 7. The two background spectra

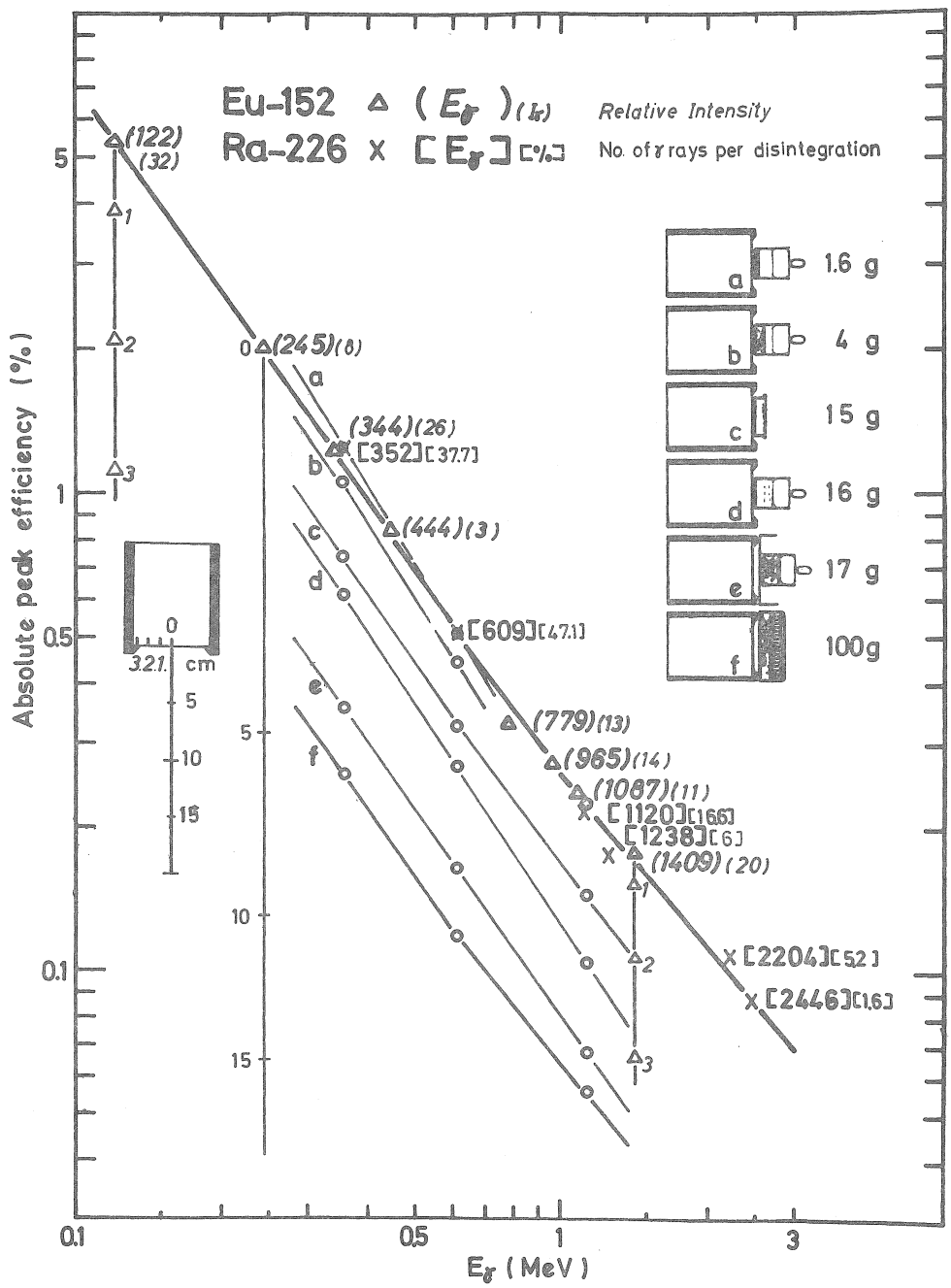
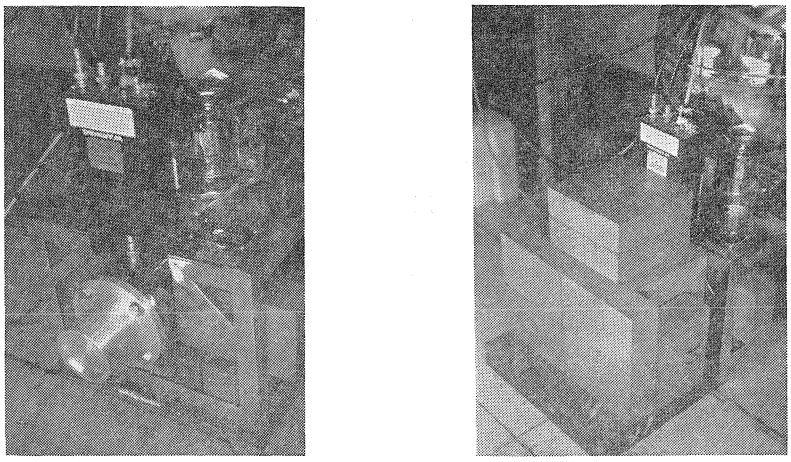


Fig. 6 The absolute peak efficiency versus both gamma-ray energy and the distance from the center of the detector head surface



A B

Fig. 7 The photographs of the detector head with (B) and without (A) lead shield

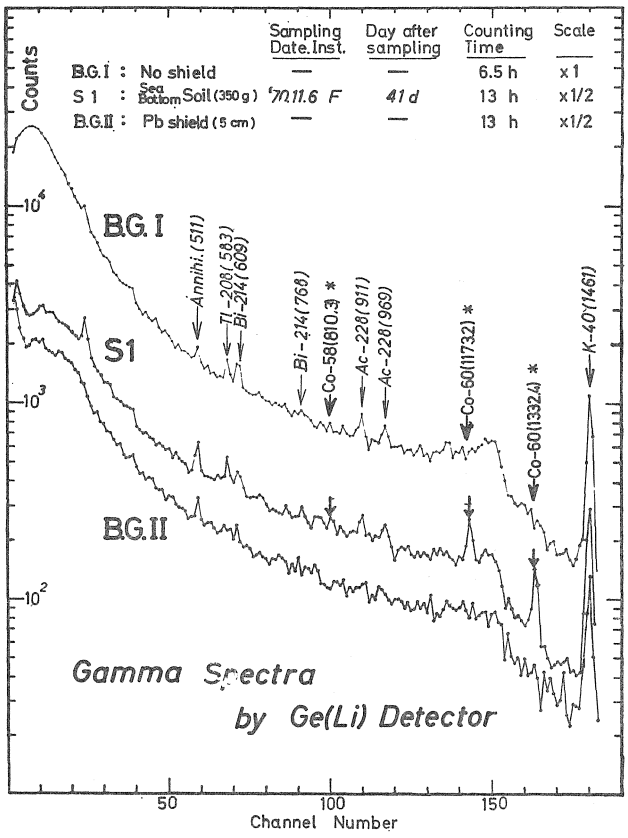


Fig. 8 The two typical background spectra with (BG II) and without (BG I) lead shield and the gamma-ray spectrum of the sea bottom soil (S-1) in Urazoko bay with lead shield

in Fig. 8 with and without lead shield show the decrease of background by shielding and the usefulness of the shielded detection system for low level γ ray spectrometry. The photopeaks obseved in the spectrums of the natural background are due to the γ rays from K-40 and the daughter nuclides of both uranium and thorium radioactive decay series. A relative estimate for the effect of shielding may be made by considering the decrease of the peak (1461 KeV) due to K-40 and its compton continuum observed even after shielding. And as the γ rays of K-40 and other nuclides are considered to come mainly from the surrounding concrete, the background will largely be depressed by setting the whole detector system in a box having thick iron walls. And the further improvements may be made by using either the Compton suppression anticoincidence technique[7] or other coincidence technique. And such improvements will open the way to the more extensive application of this detector to very low level radioactive samples in various environmental problems.

6. Applications to environmental problems

Several artificial radioactive nuclides are introduced into our environments by nuclear power industry and atomic bomb test explosions. The detection of these radioactive nuclides by γ spectrometry has been made in Japan by using the conventional NaI(Tl) detector. However, because of its poor energy resolution, this detector is not suitable to analyse minor radioactive components nondestructively and the chemical separation procedures to concentrate these radioactive nuclides are time-consuming and introduce errors in regard to over-all chemical yield. In place of the NaI(Tl) detector, we have applied above mentioned Ge(Li) detector to the nondestructive quantitative analysis of several environmental radionuclides with regard to the following two cases.

6-1. Environmental radioactivities in the samples around an atomic power plant.

The first case of our applications was related to the environmental contamination due to the out flow of the radioactive waste water by the operation of an atomic power plant at Tsuruga, Fukui Prefecture in Japan.

Figure 9 shows the monitoring stations around this atomic power plant and several sampling positions for various samples measured in this study, together with the records of operation of this atomic power plant. Their sampling dates by either Tokai-ku Research Institute of Fishery (reprensented by T) or Fukui Prefectural Institute of Public Health (represented by F) are shown in Fig. 10. All samples collected were dried and the biological materials were ashed. The gamma-ray spectrometries of these samples were performed by applying the same kind of glass bottle or plastic containers as those used for the determination of the absolute efficiency mentioned above.

Three different samples ($H_1 \sim H_3$) of sargassum gave the gamma spectrums shown in Fig. 11, H_2 and H_3 being taken from Urazoko bay near Tsuruga atomic power plant and H_1 from Niyū bay situated at the opposite side of Urazoko bay. In these spectrums, the

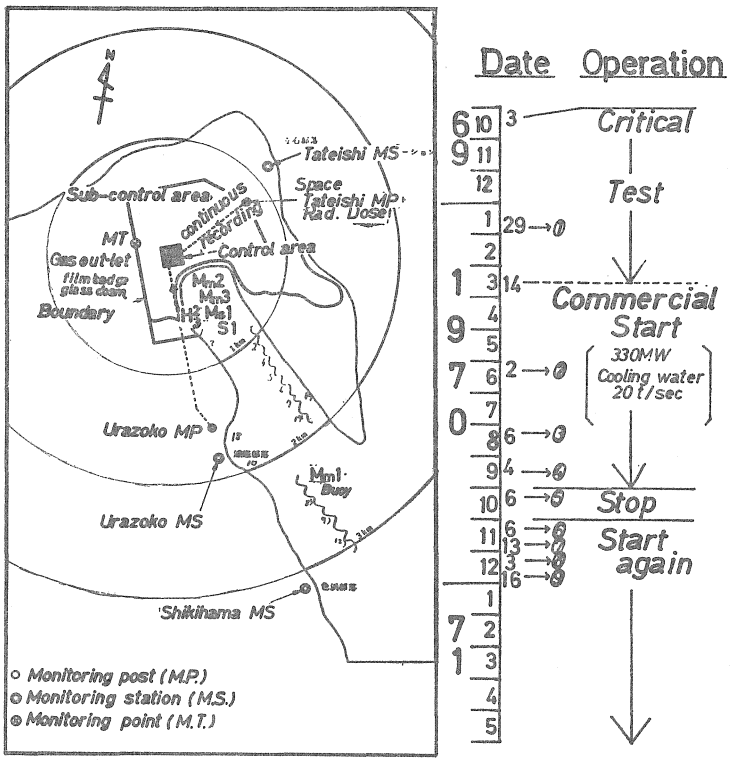


Fig. 9 The monitoring stations, several sampling locations and the records of operation of the atomic power plant at Urazoko, Tsuruga

peaks of ^{95}Zr-^{95}Nb may be due to the world-wide fission product fallout by atomic bomb test explosions. On the other hand, the peaks of Co-58, Mn-54 and Co-60 were obseved only for H_2 and H_3 and not observed for H_1. These radioactive nuclides must be derived from the operation of the Tsuruga atomic power plant because another Mihama atomic power plant on the coast of Niyū bay had not yet been fully operated on the sampling dates of these samples. The gamma spectrum of the soil sample (Sl) taken from the bottom of Urazoko bay is represented in Fig. 8, showing also the existence of these induced radioactive nuclides in this sample. And it was also found that the mussel sample had concentrated these nuclides together with Fe-59 as shown in Fig. 12.

Though the discrimination of γ-ray peaks between Co-58 (810.3KeV) and Mn-54 (834.8 KeV) had been impossible by means of NaI (Tl) detectors as seen in the right part of Fig. 12, these two peaks were clearly discriminated each other by means of our Ge(Li) detector even with the preamplifier of ORTEC 109A. And, the peak of 1098.6 KeV due to Fe-59 could also be easily discrimated from the peak of 1173.2 KeV due to Co-60 in the spectrums of the mussel samples. From these spectrums, the contents of each radionuclide were evaluated by using the absolute peak efficiency known from Fig. 6 for respective gamma energy. And the results are listed in Table I.

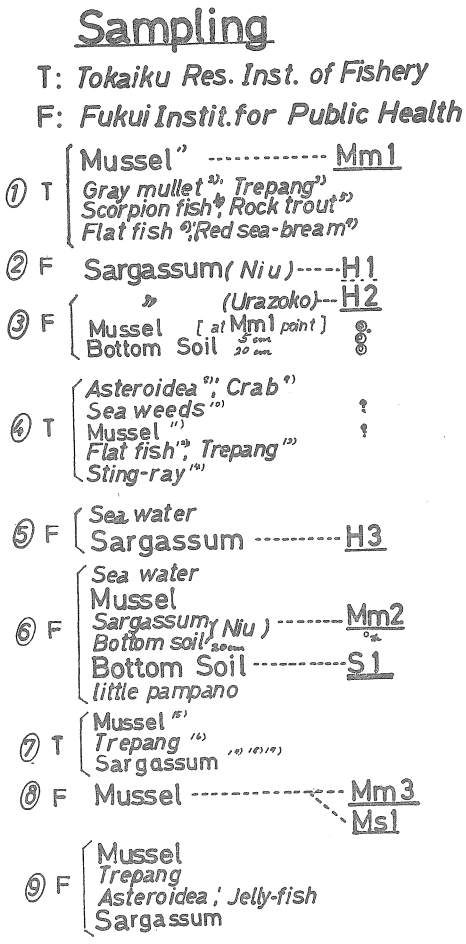


Fig. 10 Samples collected by two Institutes (T and F) and their sampling dates

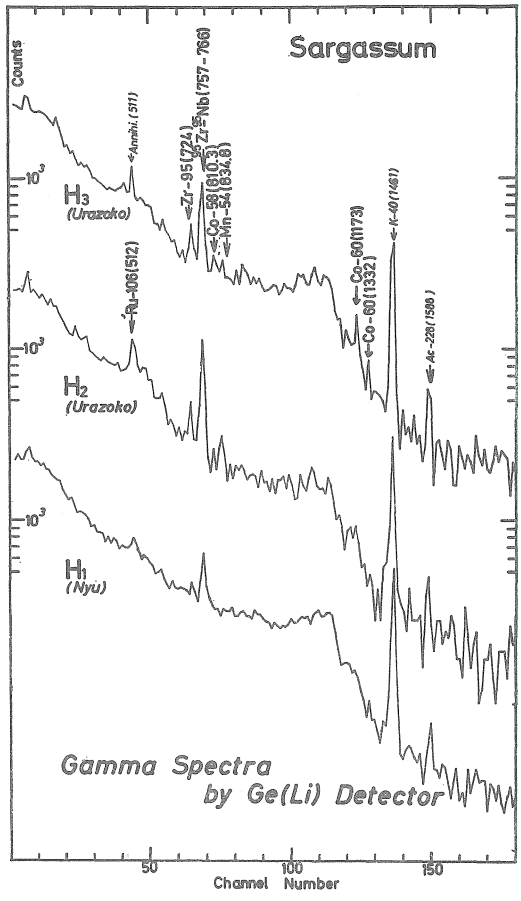


Fig. 11 The gamma-ray spectra of the sargassum samples

Table I The contents of several induced radioactive nuclides in the samples collected near the atomic power plant.

			Mussel			Sea bottom Soil
Nucl. Reac.	Nuclide	E_γ(KeV)	Mm-1	Mm-2	Mm-3	S-1
^{59}Co(n,γ)	Co-60 (5.24 yrs)	1332 1173	36 pCi/g.ash 33 〃	28 pCi/g.ash 44 〃	4.8 pCi/g.ash 6.3 〃	1.9 pCi/g.dry 1.8 〃
	Other data		(T) (850 pCi/kg) = 34 pCi/g.ash*	(F) 26 〃	(F) 1.1 〃	(F) 1.67 〃
^{54}Fe(n,p)	Mn-54 (291 d)	835	19 pCi/g.ash	8.6 〃	2.9 〃	0.2_8 〃
^{58}Ni(n,p)	Co-58 (71.3 d)	810	3.6 〃	——	——	0.5_5 〃

* if 2.5% ash

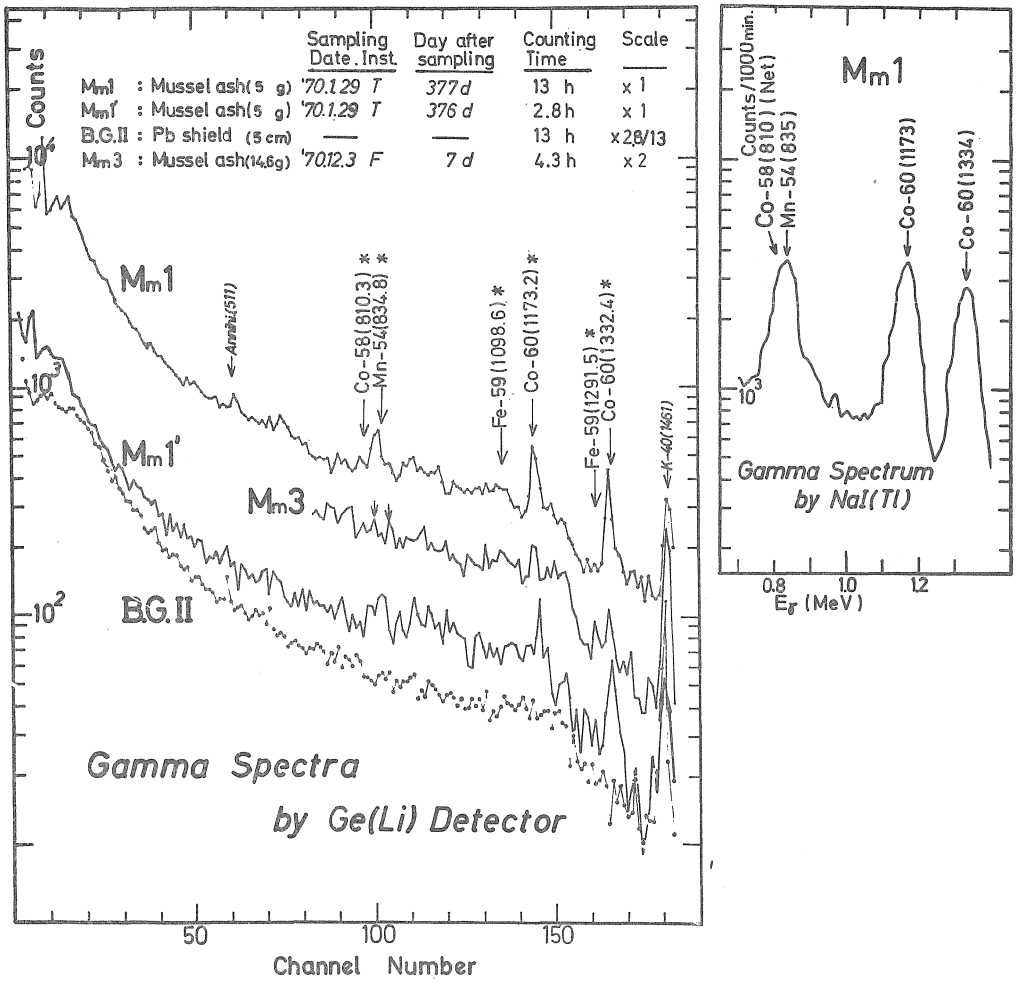
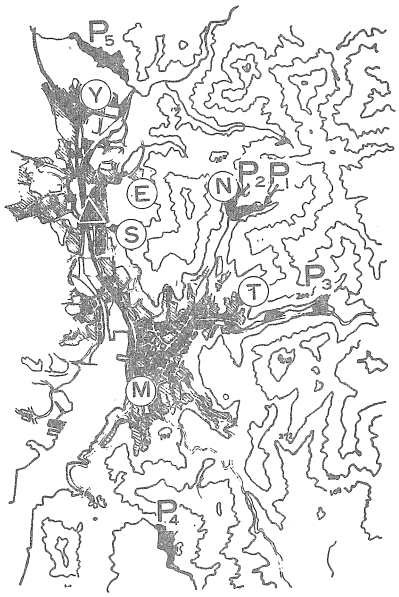


Fig. 12 The gamma-ray spectra of the mussel samples

6-2. Cs-137 remaining in soil samples from Nagasaki

The second case of our applications concerned with the study of the soil samples from Nagasaki subjected to a plutonium atomic bomb in 1945. In our laboratory, a fairly large amount of plutonium-239 were found in some soil samples taken in 1969 from several locations (Fig. 13) in Nagasaki city by α spectrometry after the radiochemical separation of plutonium.

The γ spectrometric measurements for these samples were also carried out nondestructively with our Ge(Li) detector by filling the plastic container (c) (Fig. 6) with each sample. The examples of these γ spectrums are shown in Fig. 14(a), the limitted part near Cs-137 peak being expanded in Fig. 14(b). The contents of Cs-137 in several samples were estimated from its 661 KeV γ-ray peak by referring to the absolute efficiency at this γ ray energy, ORTEC 120-3F preamplifier being used in these experi-



▲ A centre of atomic bomb explosion

Fig. 13 The sampling locations of some soil samples in Nagasaki city

Table II The contents of Cs-137 in several soil samples from Nagasaki

Sampling Location (Depth 0–10 cm.)	Sample Weight(g)	Counting time(hr)	Cs-137 nCi/kg
P_1 (Nishiyama A)	16	26	2.03
P_2 (Nishiyama B)	16	20	2.49
P_3 (Hongochi)	16	21	2.02
P_4 (Ogakura)	16	26	2.32
P_5 (Urakami)	16	24	1.91

Nishiyama B 0–25 cm.

Depth (cm)	Sample Weight(g)	Counting time(hr)	Cs-137 nCi/kg
1 – 2.5	15	24.0	2.08
2.5 – 5.0	15	24.1	2.15
5.0 – 7.5	9	38.6	3.32
7.5 –10.0	15	24.0	3.19
10.0 –12.5	15	36.0	2.92
12.5 –15.0	15	36.0	2.83
15.0 –17.5	15	24.0	2.19
17.5 –20.0	15	24.0	2.14
20.0–22.5	15	24.0	2.07
22.5–25.0	15	25.0	1.50

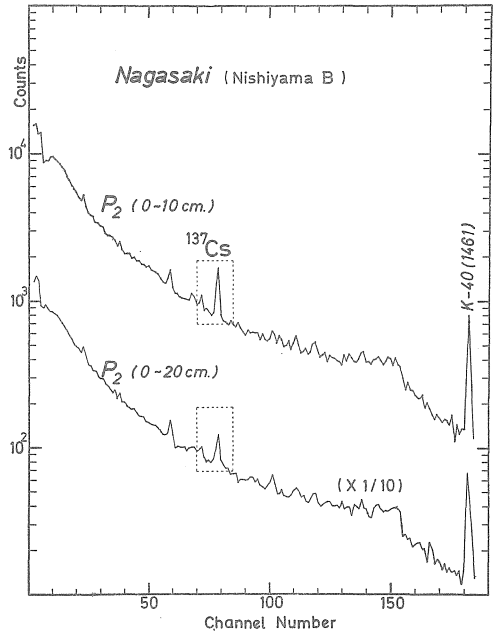


Fig. 14 (a) The examples of the gamma-ray spectra of soil samples from Nagasaki

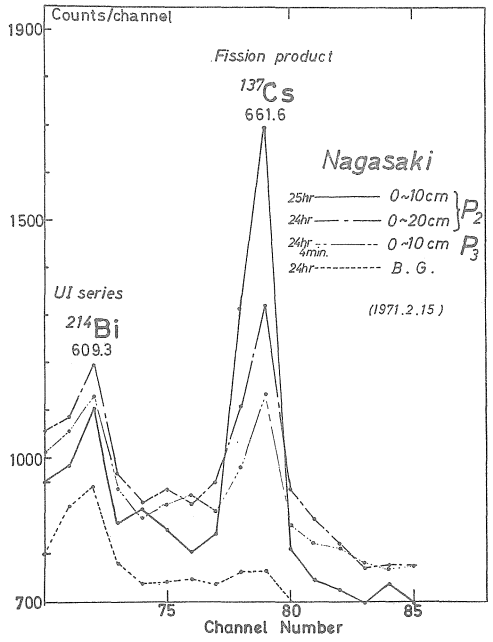


Fig. 14 (b) The expanded part near Cs-137 peak

ments. From these results listed in Table II, it was found that some soil samples in Nagasaki were still now more largely contaminated with Cs-137, a long half-life fission product, owing to the disastrous atomic bomb explosin in 1945 over this city than the world wide surface contamination with this nuclide due to various big atomic bomb test explosion in stratosphere. Furthermore, the distribution of this nuclide along the depth of a soil column through 24 years is very interesting, as compared with the distribution of plutonium.

In conclusion, as well as for various radio-chemical and radio-analytical studies, the Ge(Li) gamma-ray detector system installed in our university was found to be more useful even for the environmental low level radioactivity studies, because of its high energy resolution than the conventional NaI (Tl) detector, though the counting efficiency of Ge (Li) detector was lower than that of NaI(Tl). Based on the results reported in this paper, this detector will be available very effectively for the nondestructive quantitative analysis to obtain the accurate data required for the problems of radioactivity pollution.

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