退官記念論文

Fabrication of Mercuric Iodide Nuclear Radiation Detector and Its Application

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ABSTRACT

The performance of a nuclear radiation detector constructed with a mercuric iodide (HgI_2) crystal grown by the vapor transport method of the conventional and simple static sublimation were measured.

It was shown that the HgI_2 detector operated at room temperature and that the initial detection characteristics were maintained for a long period.

The authors applied the detector to the measurements of fission products in JRR-3 cover-gas of JAERI and demonstrated that quantitative determination of 80keV γ-ray released from ^{133}Xe could be performed by the detector in spite of the extremely small size of its element ($1.95mm^2 \times 0.7mmt$).

1. Introduction

The Ge semiconductor detector, which is widely used in the field of spectral measurement of radiation, especially of γ-ray, has an excellent energy resolution, but requires liquid nitrogen cooling. Therefore, the tedious supply and the big container of liquid nitrogen required are undesirable problems for its use. As a γ-ray detector which can be used at room temperature, the NaI(Tl) scintillation counter, which is often used for spectral measurement, is not so good in energy resolution, and cannot be used for complicated γ-ray spectral measurements.

Therefore, the development of a small semiconductor detector which can be operated at room temperature without liquid nitrogen cooling and shows a better energy resolution than NaI (Tl) scintillation counter has been requested.

As semiconductor detectors which can be operated at room temperature, Si, GaAs, CdTe, HgI_2 etc. are considered. If one compares atomic numbers, forbidden band gaps, mobilities of

electron and hole, mean free times etc. of these semiconductor materials[1], the Si detector does not fit the γ-ray spectral measurement because of its small atomic number. Although the GaAs detector was the first to have shown a good energy resolution at room temperature among the compound semiconductors, the atomic number is almost the same as the Ge detector and only small single crystals can be obtained, and therefore its practical application is difficult. Although the CdTe detector has a larger atomic number than the Ge detector, there are problems such that the growth of big crystals is not obtained and the pulse height fluctuates during the time course.

Compared with these detectors, the mercuric iodide detector has superior characteristics as a detector of γ-ray spectral measurement since it has

(1) a broad band gap which is necessary for performance at room temperature and
(2) a large atomic number which is required for the efficient detection of photo electron peaks even by small crystals.

Noticing the above points, the authors tried to fabricate the HgI_2 detector and to apply it to the field of γ-ray measurement for a nuclear reactor. The detector was applied to the quantitative measurement of radioactivity in the cover-gas of a nuclear reactor and was ascertained to be used practically as a monitor detector.

2. Fabrication of γ-Ray Detector and Its Detection Characteristics

2.1 Growth of HgI_2 Crystal and Fabrication of Detector

There are several methods to grow HgI_2 crystals such as the solvent evaporation[2,3], and the vapor transport[4-7] which includes the static sublimation, the dynamic sublimation and the temperature fluctuation methods. The authors have grown HgI_2 crystals by these methods and fabricated a HgI_2 detector by mounting the crystals on a detector holder[8,9]. Through the measurement of detecting characteristics, it was found extremely difficult to fabricate a detector which has a high detecting efficiency and a good energy resolution in addition to the ability to maintain the initial detecting characteristics for a long period.

Among the detectors fabricated, an HgI_2 detector which employed a crystal grown by the vapor transport method, utilizing simple static sublimation, exhibited a relatively good energy resolution and stable detecting characteristics. Therefore, it was selected as a detector to be studied in detail. The vapor transport method by the static sublimation is a crystal growth method where the starting material of HgI_2 is sealed into one end of a glass tube under vacuum and placed in the high temperature part of an electric furnace having two temperature regions. Many small HgI_2 crystals are grown in the low temperature part. Figure 1 shows the temperature distribution on the wall of the glass tube used for crystal growth. In #1 and #2, commercially available HgI_2 powder was employed as a starting material and in ##2, the crystal group grown in #2 was used as a starting material and sublimated again.

After crystal growth for 10 days for #1, #2 and 5 days for ##2, many crystals with the size of a few mm square and 1 mm thick were obtained at the region where the tube wall temperature was about 110°C.

Among these crystals, highly transparent ones were chosen and cracked at easily cleavable

planes with a thin blade, etched by dipping into a 20-25% KI solution and washed. They were coated with Aquadag (glue like graphite) as the electrode and mounted on a detector holder with a SHV connector as shown in Fig.2. The points paid attention to during the fabricating process are:

1. In order to avoid an active amalgum reaction between mercury in the crystals and metal material, cover glass was intervened on the brass pedestal in the detector holder.

2. The gold lead wire was coated with Aquadag. As a finishing treatment, HumiSeal (transparent resin of acrylate) was coated on the whole surface of the crystals in order to maintain the stable detection characteristics of the detector for a long period.

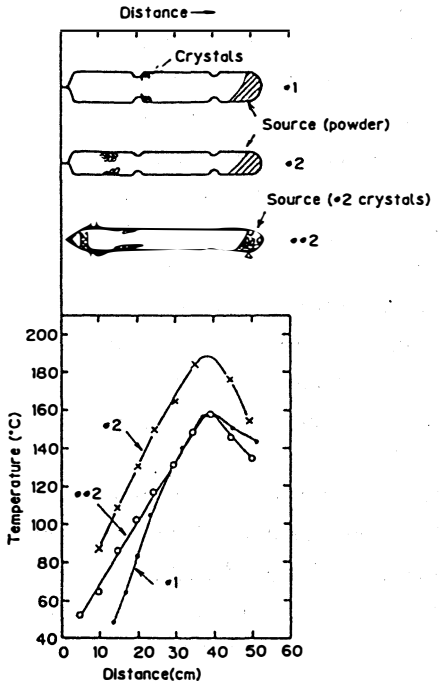

Fig. 1 Positions of crystals grown and corresponding temperature distributions

2.2 Detection Characteristics of α-Ray

Figure 3 is a block diagram of the measurement circuit of α- and γ-rays. From the observation of the output wave of the preamplifier in the detection of γ-ray from ^{241}Am, the initial rise time of the wave was obtained and the mobility of carrier, μ, was calculated.

By observing the voltage dependence of the pulse height, (mobility of carrier μ) $\times$ (mean free time τ) $= (\mu\tau)$ product was obtained. The $(\mu\tau)$ products of electron and hole, $\mu_e\tau_e$ and $\mu_h\tau_h$ were $10^{-4}\sim10^{-5}$ and $2\times10^{-7}cm^2/V$, respectively. The product of electron showed almost the same value as the theoretical value, but that of hole was found to be worse by 2-3 magnitudes.

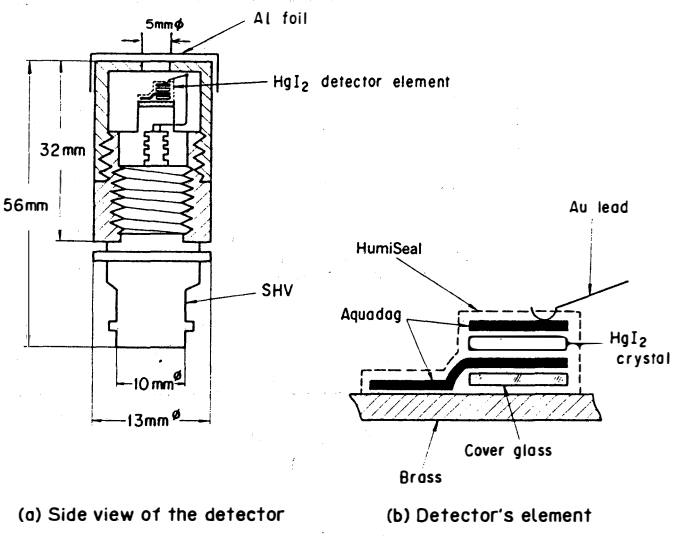

Fig. 2 Structure of detector and its element

2.3 Detection Characteristics of γ-Ray

The distribution of pulse

height for a ^{241}Am γ-ray source observed when $-1{,}000V$ (on electron traversing) and $+1{,}000V$ (on hole traversing) were applied to the electrode at the radiation injection site of the ##2-2 HgI_2 detector (HgI_2 single crystals of 1.95mm^2 $\times$0.7mm of thickness) is shown in Figs.4(a) and 4(b). A better energy resolution was observed for a negative bias than for a positive bias. This behavior comes from the fact that the charge collection characteristics of electrons are superior than those of the hole. When a negative bias was applied, the half width energy resolution was 4.9 keV for γ-ray of 59.5keV.

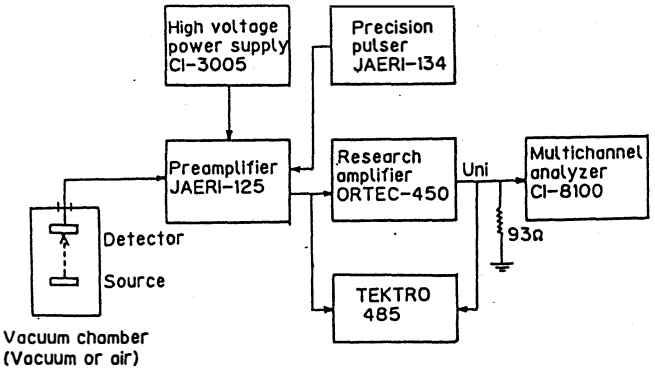

Fig. 3 Schematic diagram of electronics

The pulse height distribution of the #1-3 HgI_2 detector (HgI_2 single crystals of $1\times1.6\times0.6$mm^3), which showed the best half width energy resolution among HgI_2 detectors fabricated by the authors, is shown in Fig.5. The half width energy resolution of 3.5keV was obtained for the γ-ray of 59.5keV.

Furthermore, for the #1-3 HgI_2 detector, a stability test of long term detection characteristics was carried out by using a ^{241}Am ray source. As illustrated in Fig.6, it was ascertained that the spectral characteristics did not change even after 5 years and the detector could be used as a long-term γ-ray monitor etc..

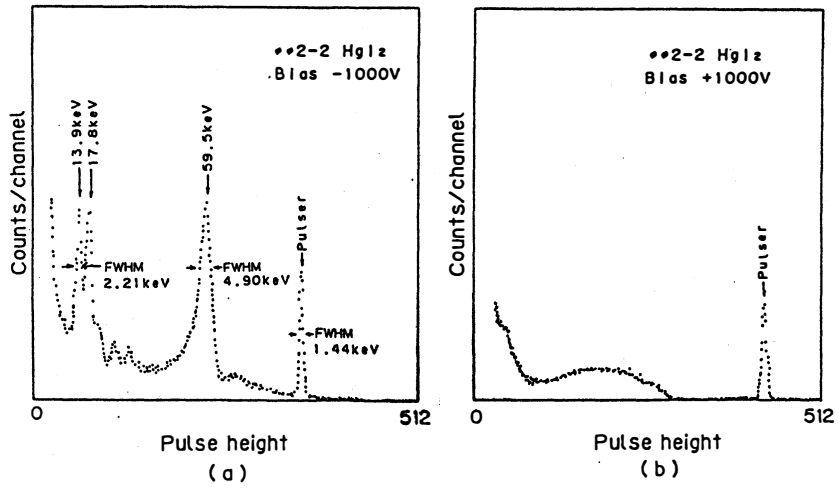

Fig. 4 ^{241}Am γ-ray spectra obtained from HgI_2 detector (1.95mm$^2\times$0.7mm) with (a) $-1{,}000$ V bias and (b) $+1{,}000$ V bias at room temperature

3. Application

At present, there is no report that HgI_2 detector was tried to be used for the γ-ray measurement related to nuclear reactor. The authors measured the γ-ray spectrum from the radiation nucleus in the cover-gas of JRR-3 using the HgI_2 detector fabricated.

Figure 7 shows a system diagram for detecting a failed fuel of JRR-3. The cooling material (heavy water) sampled each from 243 fuel-vessels is divided into two pathways and led to a gas stripper and separated into gas and water by the cover-gas (He gas) and the radiation activity in this gas was measured to detect the failed fuel [10]. The positions where the authors carried out γ-ray measurements using the HgI_2 detector are indicated by asterisks.

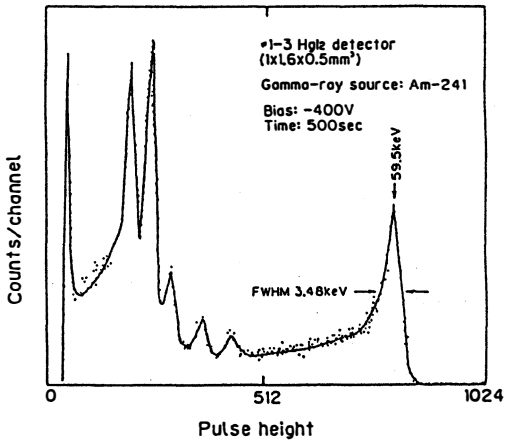

Fig. 5 ^{241}Am γ-ray spectrum obtained from HgI_2 detector ($1\times1.6\times0.5mm^3$) with -400 V bias at room temperature

In the following, the result of measurement will be shown in comparison with the example of measurement by Ge(Li) detector.

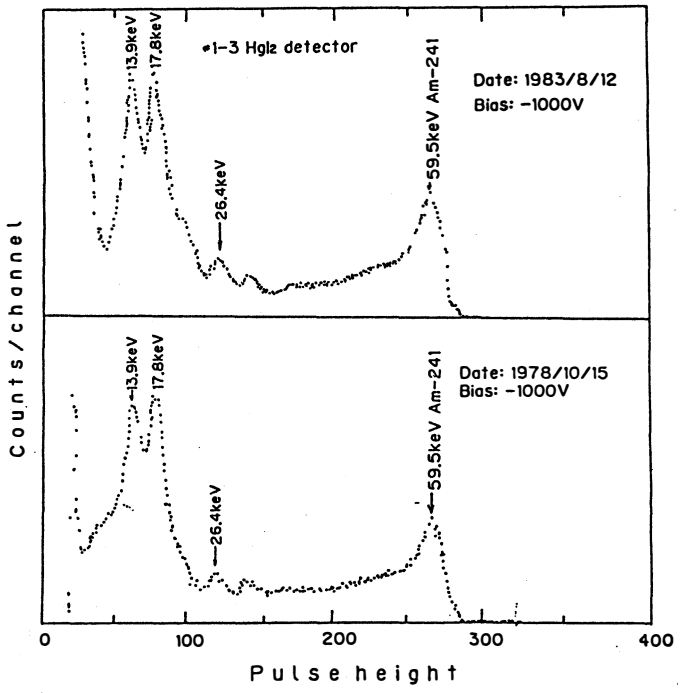

Fig. 6 Comparison of γ-ray spectra obtained from HgI_2 detector ($1\times1.6\times0.5mm^3$), (upper) on Aug. 12, 1983 and (lower) on Oct. 15, 1978

3.1 Measurement of Radionuclides in the Cover-Gas Sample

The radiation activity in the cover-gas introduced into a sampling cell of JRR-3 was measured by ##2-2 HgI_2 detector and Ge(Li) detector. The sampling cell made of aluminum of the thickness of 2mm having an inner volume of 113cm^3, was used. The γ-ray spectral measurements were performed in the arrangement shown in Fig.8 for HgI_2 and Ge(Li) detectors.

The pulse height distributions for the cover-gas measured 11 days after the sampling are shown in Fig.9 and 10 for HgI_2 and Ge(Li) detectors, respectively. In both cases, peaks of γ-ray of 80.99 keV from ^{133}Xe and of Kα and Kβ X-rays of 30.63 and 35.0 keV, emitted on the collapse of Xe to Cs, were observed. The results comparing the characteristics of the two detectors are shown in Table 1. Although the HgI_2 detector is of small size and usable at room temperaure, it is inferior in energy resolution and in detecting efficiency to the Ge(Li) detector. However, the detecting charateristics are good enough for the HgI_2 detector to be available in the measurement of low energy γ-ray emitted from cover-gas.

We tried to apply the HgI_2 detector to a quantitative analysis of the radioactive concentration of ^{133}Xe.

Firstly, we obtained the intrinsic detection efficiency of ##2-2 HgI_2 detector using ^{241}Am and ^{57}Co γ-ray sources. The results are shown in Fig.11. The interpolation of these results gave the intrinsic detection efficiency of γ-ray of 80.99 keV, ε_{int} as 0.25.

Then, using the geometry whose cross section is shown in Fig.12, the absolute detection efficiency for the cover-gas in the sampling cell of the HgI_2 detector was calculated. On obtaining the absolute detection efficiency, ε, the following approximations were employed:

(1) The radiation source distributes evenly in the cell.

(2) Only the radiation passing through the Cu collimator reaches the detector.

(3) Aluminum wall does not depend on the

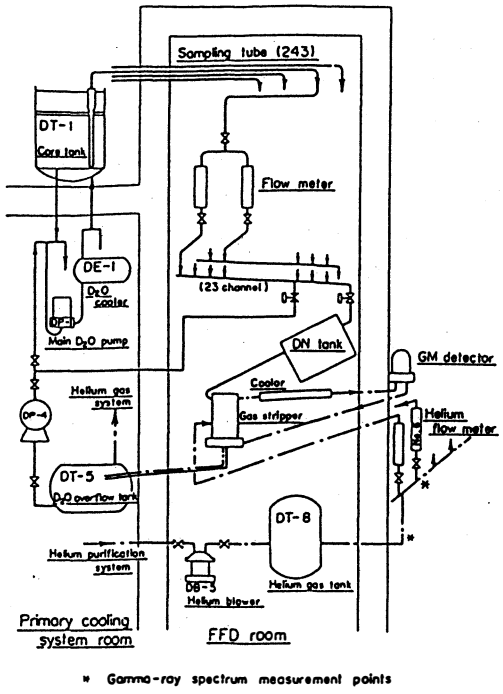

Fig. 7 γ-ray spectrum measurement points in JRR-3 fuel failure detection system

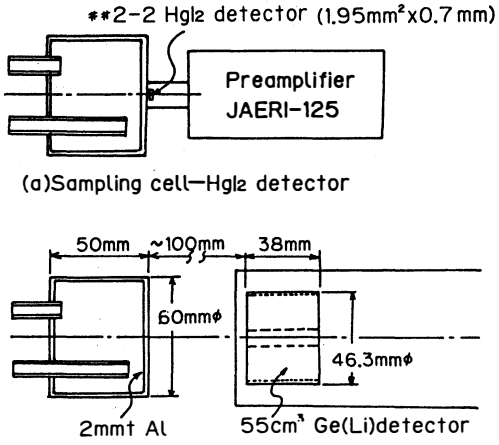

Fig. 8 Geometry between sampling cell and detector for cover-gas γ spectrometry using HgI_2 and Ge(Li) detectors

incidence direction and a constant decay is obtained.

(4) Only the incoming γ-ray on the surface facing the detector cell contributes to the counting rate. The probability that the incidence contributes to the peak counting rate does not depend on the incident angle, but is given by the intrinsic detection efficiency.

Using these approximations, the absolute detection efficiency is given by the following equation :

$$\varepsilon = (\varepsilon_{\mathrm{int}}\ \Delta S/2\pi\ R_1{}^2 T)\exp(-AH) \cdot [R_1(\sqrt{1-\mu_2{}^2}-\sqrt{1-\mu_1{}^2}) - L_1(\mu_1-\mu_2)+T(1-\mu_1)] \quad (1)$$

Here, intrinsic detection efficiency of the HgI_2 detector $\varepsilon_{\mathrm{int}}$: 0.25
Radius of the sampling cell R_1 : 2.8cm
Length of the sampling cell T : 4.6cm
Thickness of the sampling cell H : 0.2cm
Distance from the sampling cell to the surface of HgI_2 crystals L_1 : 0.63cm
Radius of the hole of Cu collimator R_2 : 0.25cm
Distance from Cu collimator to the surface of HgI_2 crystals L_2 : 0.28cm
Absorption coefficient of Al A : $0.51\mathrm{cm}^{-1}$
Sensitive area of HgI_2 detector ΔS : $0.017\mathrm{cm}^2$

$$\mu_1 = \cos\theta_1 = (T_1+L)/\sqrt{(T+L_1)^2+R_1{}^2}$$
$$\mu_2 = \cos\theta_2 = L_2/\sqrt{L_2{}^2+R_2{}^2}$$

Introducing these values into equation(1),

$$\varepsilon = 1.70\times10^{-5}$$

was obtained.

The radioactivity N (μCi) in the cell is calculated by the following equation using the absolute detection efficiency ε, γ-ray branching ratio B and peak counting rate P (cps)

$$N = P/(3.7\times10^4\times B\times\varepsilon) \quad (2)$$

Therefore, noticing the γ-ray peak of 80 keV

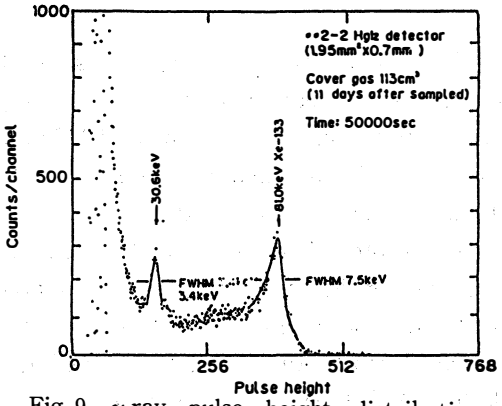

Fig. 9 γ-ray pulse height distribution obtained from HgI_2 detector (1.95mm²×0.7mm) with −1,000 V bias at room temperature

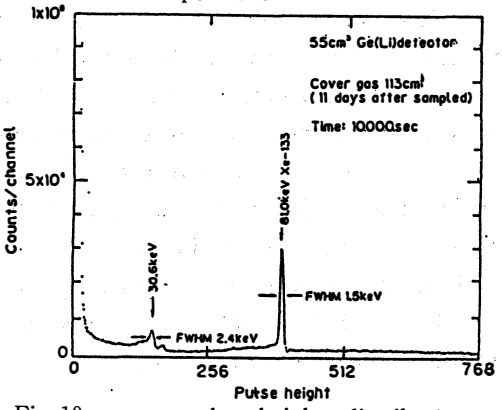

Fig. 10 γ-ray pulse height distribution from Ge(Li) detector

Table 1 Comparison of size and performance of HgI_2 and Ge(Li) detectors

Performance	HgI_2 detector	Ge(Li) detector
Detector size	1.95mm²x0.7mm	46.3mmφ x38mm
Detector volume	1.4 mm³	55 cm³
Detector temperature	Room temperature	77 k
Detector bias	-1000 V	+2400 V
Shaping time	3 μ sec	2 μ sec
Cover-gas pulse height Distribution	Fig. 9	Fig. 10
Xe-133 80keV γ-ray		
FWHM resolution	7.5 keV	1.5 keV
Peak counting rate	0.208 cps	19.46 cps
Cs 30.6keV X-ray		
FWHM resolution	3.4 keV	2.4 keV
Peak counting rate	0.049 cps	4.04 cps

of ^{133}Xe , introduction of P=0.208 cps, B=0.366 into the above equation (2) gives N as 0.90 μCi. Deviding this value by the volume of 113cm^3, the radioactive concentration of ^{133}Xe in the cell was obtained as 8.0×10^{-3} μCi/cm^3.

On the other hand, in order to estimate the correctness of the above approximated analytical calculation, the radioactive concentration was also calculated by the Monte Carlo method considering the three dimensional form. It was ascertained that the radioactive concentration was almost identical, being 7.9×10^{-3} μCi/cm^3.

On the other hand, the radioactive concentration obtained from the counted value measured by the Ge(Li) detector was 9.4×10^{-3} μCi/cm^3. Comparing the two results, the analytical reasult given by the HgI_2 detector is a smaller value by about 15 %, but it is assumed to be of sufficient accuracy considering that the dimention of the detector is small. Thus, it was ascertained that the HgI_2 detector can be used for the quantitative measurement of γ-ray.

3.2 Measurement of the Radionuclides in the Cover-Gas in the Reactor Room

While JRR-3 is operated under an output of 10 MW, the γ-ray of the radioactivity in the cover-gas flowing into the monitor chamber (MC-2, volume of 100 cm^3) at a rate of 2 ℓ/min was measured. As an HgI_2 detector, #1-3 detector was used. The distribution of pulse height of γ-ray measured is shown in Fig.13. The γ-ray of 80 keV from ^{133}Xe and the γ-ray of 250 keV from ^{135}Xe with a shorter half life, which could not be measured for the sampling gas, were clearly observed.

The result of the measurement for the cover-gas, which was sealed into the monitor chamber when cover-gas did not flow by the stop of JRR-3, are shown in Figs.14(a) and 14(b). For Fig.(a), 47000s (about 13 h) were counted and for Fig.(b), further 212000s (about 2.45 d) after the previous

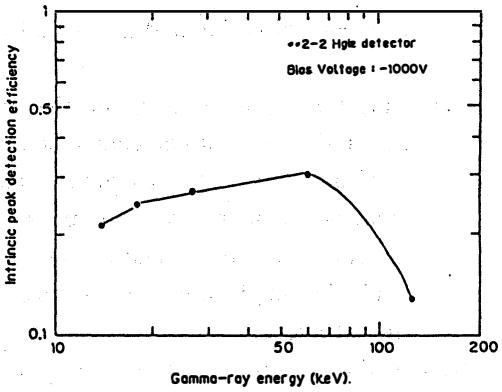

Fig. 11 Intrinsic peak detection efficiency of ##2-2 HgI_2 detector

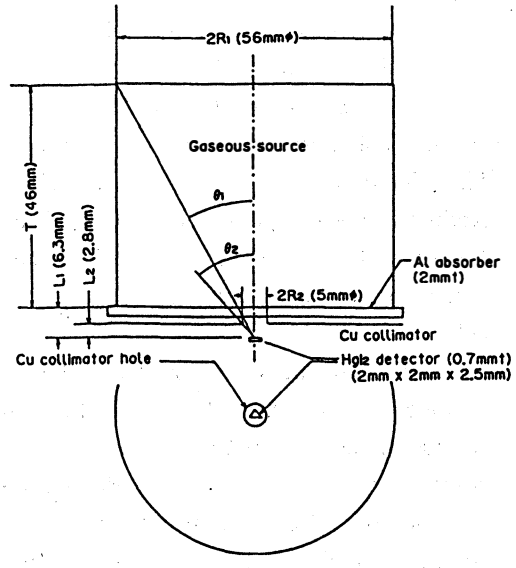

Fig. 12 Calculation model for cover-gas

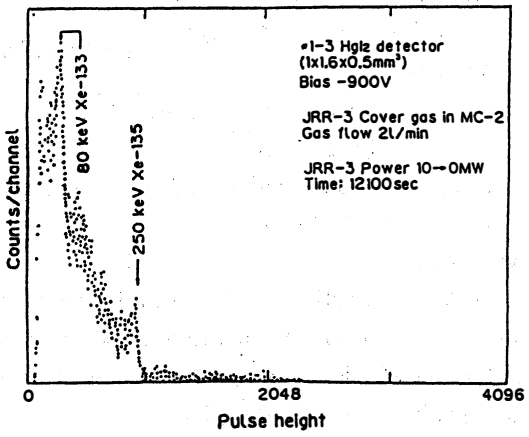

Fig. 13 JRR-3 cover-gas γ-ray spectrum obtained from HgI_2 detector with −900 V bias at room temperature

measurement were counted. In these measurements, the process of decay of radioactive nucleuses, ^{135}Xe (half life of 9.1 h) and ^{133}Xe (half life of 5.25 d) which are the main species in the cover-gas, was measured. Due to this process, the relative ratio of the γ-ray peak of 250 keV of ^{135}Xe with a shorter half life, to the γ-ray peak of 80 keV of ^{133}Xe with a longer half life is changing.

It was ascertained that the changing ratio of both countings shows a good agreement with that calculated using the values of the half life. In this way stable operation of the HgI_2 detector was ascertained.

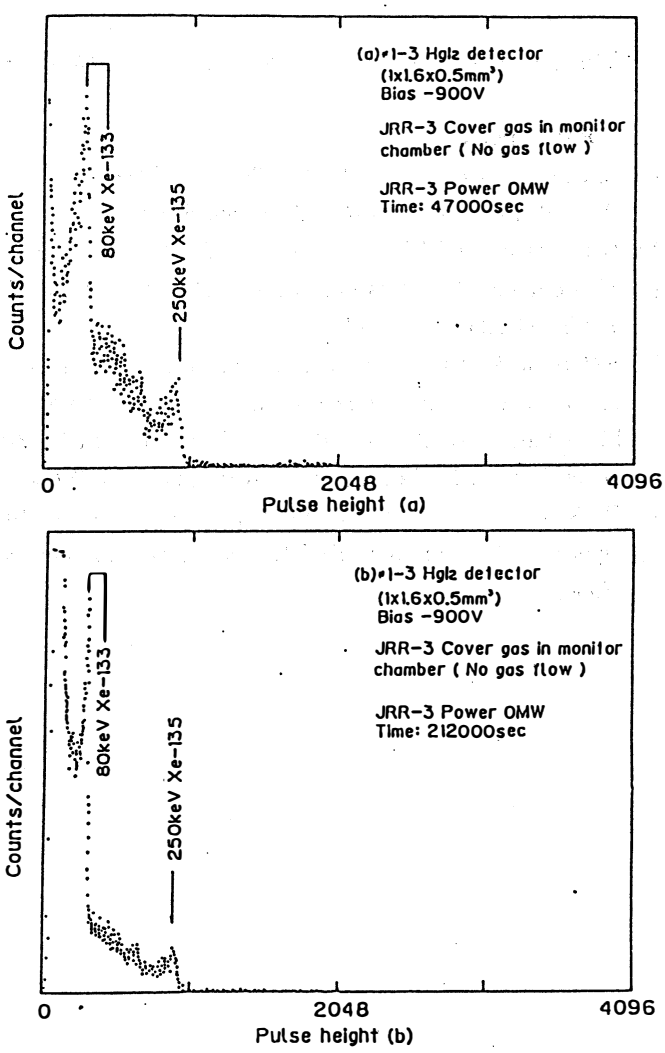

Fig. 14 γ-ray spectrum of JRR-3 cover-gas in monitor chamber obtained from HgI_2 detector after cover-gas flow was shut off

4. Conclusion

As described above, the authors fabricated a small nuclear radiation detector by using single crystals of HgI_2 which were grown by a simple vapor transport method, measured the detection characteristics and applied it to the quantitative determination and the monitoring of radioactivity in the cover-gas of JRR-3 nuclear reactor. As a result, provided that the quantitative calculation is made with due consideration of the geometrical conditions of the detector and the object to be measured, the HgI_2 detector was ascertained to be applicable enough to the quantitative determination in the region of relatively low γ-ray energy such as 50-250 keV, and it was also ascertained that it can be used stably for the γ-ray measurement even at room temperature. The HgI_2 detector will be applied to various fields in the future, due to the merit that it is small and usable at room temperature.

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