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Determination of Photopeak Efficiencies of Voluminal Samples for the Measurement of Environmental Radioactivities

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The photopeak efficiencies of a high-purity germanium detector for various voluminal sources of drum-type and Marinelli-type shapes have been experimentally determined, making use of the solution of mixed radionuclide gamma-ray reference standard. Limits in determining concentration of radioactivities in environmental samples with the photopeak efficiencies are also discussed.

KEYWORDS : Gamma-ray spectrometer/Photopeak efficiency/Environmental radioactivity/Marinelli beaker/

1. INTRODUCTION

Because of their excellent energy resolution and large detection efficiency, high-purity germanium detectors are usually employed to measure gamma activities in environmental samples. Even with such detectors of high performance, it is difficult to measure some artificial radioactivities in soil, water and aerosol, which were produced by many nuclear tests for the past several decades. One way to measure the very low level radioactivities is to concentrate the activities using chemical techniques. However, the concentration is practically not possible in many cases. The other way is to use as a large amount of samples as possible in the measurement. Therefore, large vessels of drum-type or Marinelli-type are often used in the low level measurements for environmental samples.

To determine precisely the concentration of radioactivities (Bq/kg or Bq/m^3), it is necessary to know the photopeak efficiency (or full-peak efficiency), i.e., a probability with which the whole energy of a gamma ray emitted from sample is absorbed in the detector. As expected, the photopeak efficiency is a complicated function of many parameters related to gamma ray, source and detector, e.g., energy of gamma ray, dimension of detector, dimension of voluminal source, geometrical arrangement of detector and source and density of sample. Moreover, the efficiency gradually changs in a long period, e.g., several years, mainly because of the change in the sensitive volume of high-purity germanium detector. Numerical estimations such as Monte Carlo calculations for the efficiencies of voluminal sources were developed by previous workers[1,2]. With the numerical methods, it is actually difficult to obtain the photopeak efficiency within an

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accuracy better than 5%.

A more reliable method is to determine experimentally the efficiencies with the mixed radionuclide gamma-ray standard solution. This solution, commercially available, is enough precisely calibrated for the intensities of principal gamma rays, which are well spaced in energy from 88 keV to 1836 keV. In the present work, the photopeak efficiencies for various voluminal sources has been determined with the standard solution. These efficiencies will be used to measure the background level of artificial radioactivities on Gununq Muria in the middle of Java Island, where a nuclear power plant will be built at 2005, and Karimunjawa Archipelago in Jawa Sea, where facilities for radioactive waste products are planted to be constructed in near future.

2. GAMMA-RAY SPECTROMETER

The high resolution gamma-ray spectrometer of Radioisotope Research Center of Kyoto University has been used in the present work. In Fig. 1 is shown the gamma-ray spectrometer, which mainly consists of a high-purity germanium detector (GMX-18200-S, EG&G ORTEC), a shield vessel and a tank for liquid nitrogen. The size of the detector is 52-mm diameter × 48-mm height (102 cc). The relative efficiency, i.e., a ratio of the efficiency of this detector for ^{137}Cs 662-keV gamma ray to that of 3″ × 3″ NaI(Tl) crystal, is 22.3%. The entrance window is 0.5-mm thick beryllium, which makes possible to detect low energy photons (>3 keV) with high efficiency. The energy resolution is 540 eV for 5.9-kev X ray from ^{55}Fe and 1.8 keV for 1.33-MeV gamma ray from ^{60}Co. As seen in Fig. 1, the head of the detector and the sample to be measured are mounted in a cylindrical vessel made of 10-cm thick lead; the inner size of the vessel is 52-cm diameter × 69-cm height. This vessel is necessary for shielding the detector head from natural radiations. To deduce gamma rays backscattered from the sample and also to absorb X rays from the lead of vessel, the inner side of vessel is covered with 3-mm thick tin, 1-mm thick copper

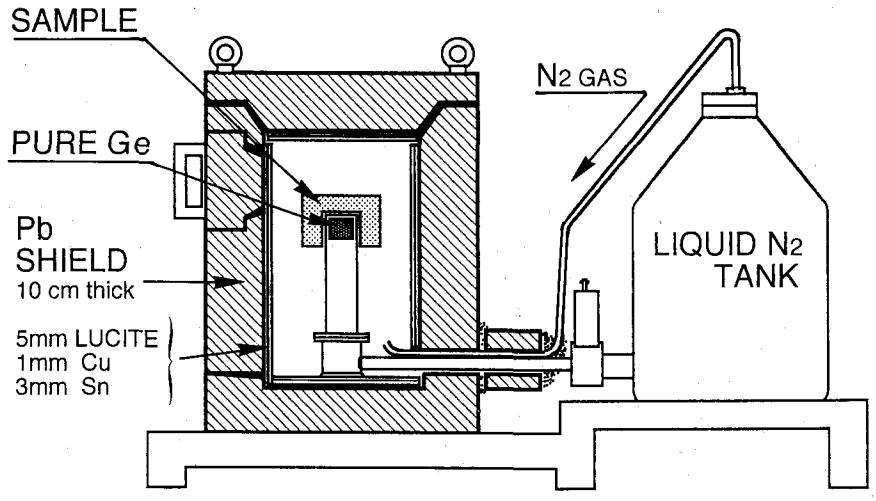

Fig. 1. Gamma-ray spectrometer for the measurement of environmental radioactivities.

and 5-mm thick Lucite in order. Nitrogen gas evaporating from the tank of liquid nitrogen is passed through the inner room of vessel. This is effective to deduce background counts caused by aerosol in air, which are contaminated by radioactive daughter nuclides of ^{222}Rn, such as ^{214}Bi and ^{214}Pb[3].

A typical background spectrum is shown by Fig. 2; net count rates at 2614.5-keV ^{208}Tl, 1460.7-keV ^{40}K, 803.4-keV ^{210}Po, 238.6-keV ^{212}Pb, 186.0-keV ^{226}Ra, 92.4, 92.8-keV ^{234}Th and 46.5-keV ^{210}Pb gamma-ray peaks are 1.7 (77%), 3.9 (77%), 1.8 (31%), 4.4 (5.4%), 7.7 (11%), 14.5 (18%) and 18.4 (25%) counts/ksec, respectively, where values in parentheses are a ratio of net counts to full counts at each peak.

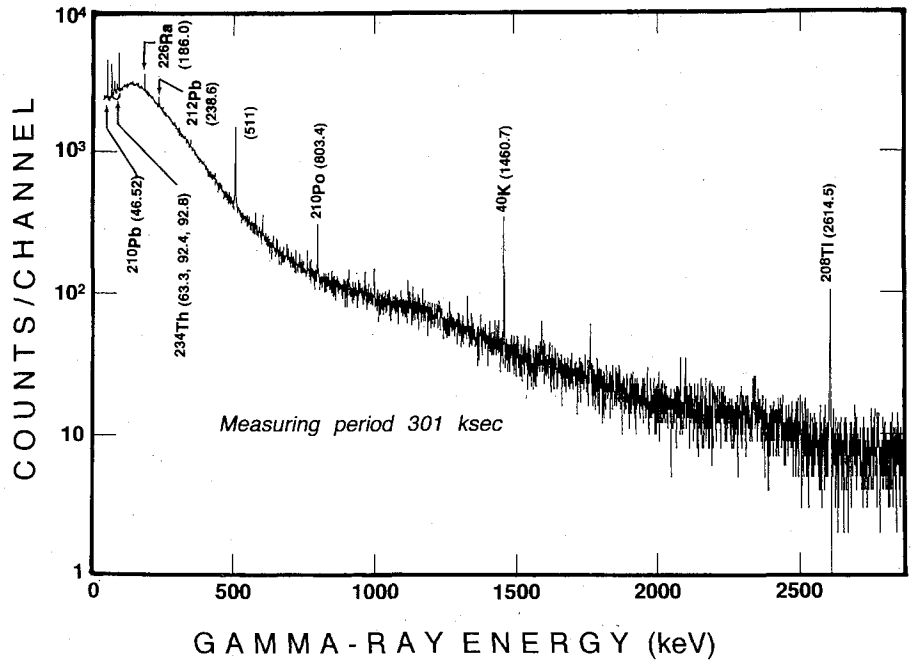

Fig. 2. Background spectrum obtained by the gamma-ray spectrometer.

3. PREPARATION OF VOLUMINAL SOURCES

Voluminal sources with gamma activities have been prepared by mixing the gamma-ray standard solution with a large volume of aluminum silicate power (Al_2O_3 $3SiO_2$). The standard solution is the mixed radionuclide gamma-ray reference standard (QCY.44) obtained from Radiochemical Center Ltd., Amersham. In Table 1 are listed nuclides in the solution, number of gamma rays per gram per second and uncertainties in the number of gamma rays.

The procedure for mixing the solution with the powder is as follows :

1) The whole solution in 4M HCl ($\sim$5 cm^3) is diluted in twice the original volume.

2) The half of the diluted solution, exactly divided, is mixed with the solution of methylene blue ($\sim$5 cm^3).

3) The solution colored in blue ($\sim$10 cm^3) is mixed with aluminum silicate powder of about 200 g. The powder is carefully ground down in a mortar until the color of methylene blue becomes uniform in the whole powder.

4) The colored powder ($\sim$200 g) is mixed with about ten times amount of aluminum silicate in a polyethylene vessel of a large volume (20 liters). The color of methylene blue is again useful to check the uniformity of radioactivities in the powder.

5) The weight of the whole powder is precisely measured to estimate the concentration of the radioactivities.

To examine the distribution of activities in the powder, we measured total count rates per unit weight of gamma rays emitted from the powder. Results for 3 samples are given in Table 2. There appears a deviation larger than the statistical uncertainties between the count rate of sample 2 and those of samples 1 and 3. However, the deviation is below 5%, which has been required in the present work.

Table 1 Absolute intensities and their uncertainties of gamma-rays emitted from the standard solution.

Parent radionuclide	Gamma-ray energy (keV)	Gamma-rays per second per gram of solution	Uncertainty
^{109}Cd	88.03	638	3.3%
^{57}Co	122.1	584	1.8%
^{139}Ce	165.9	798	1.4%
^{203}Hg	279.2	1941	2.1%
^{113}Sn	391.7	2085	4.2%
^{85}Sr	514.0	4011	1.8%
^{137}Cs	661.7	2487	2.2%
^{88}Y	898.0	6270	3.7%
^{60}Co	1173	3489	0.7%
^{60}Co	1333	3492	0.7%
^{88}Y	1836	6644	3.6%

Table 2. Total count rates per gram of 3 samples.

sample	No.1	No.2	No.3
cps/g	6.75$\pm$0.04*	6.64$\pm$0.04*	6.67$\pm$0.04*

*) Error estimated from statistical uncertainties of total counts.

4. DETECTION EFFICIENCY

In Fig. 3 are shown the shape of vessels used in the present work, i.e., drum-type and Marinelli-type. Geometrical dimensions of drum-type vessels and Marinelli-types are listed in Tables 3 and 4, respectively.

The photopeak efficiency of each vessel for gamma ray with an energy E was estimated from

$$D(E)=n(E)/[S\cdot w\cdot t\cdot N(E)] \quad (1/2)^{T/D}, \tag{1}$$

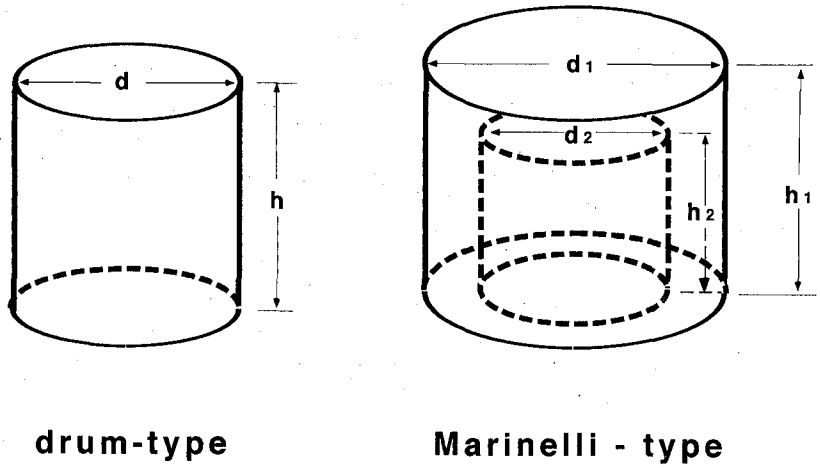

Fig. 3 Drum-type and Marinelli-type vessels for environmental samples.

Table 3. Dimensions of drum-type vessels.

Vessel No.	D1	D2
d(mm)	49	59
h(mm)	8	42
Volume(cm^3)	15.1	115

Table 4. Dimensions of Marinelli-type vessels.

Vessel	M1	M2	M3	M4	M5	M6
d1(mm)	116	94	88	106	98	114
d2(mm)	76	76	76	76	76	76
h1(mm)	98	50	105	115	110	120
h2(mm)	77	35	100	100	100	100
Volume(cm^3)	689	188	185	561	376	772

where $n(E)$ is total photopeak counts in gamma-ray spectrum, S is weight of the original standard solution (g), w is ratio of the weight of powder in each vessel to that of the whole powder, t is measuring period for gamma-ray spectrum (sec), $\mathcal{N}(E)$ is gamma rays per second per gram of the original solution, T is the period from reference time of the original solution to the day of gamma-ray measurement (day), D is half life of radioactive nuclide (day).

Typical results for the vessel D2 are shown in Fig. 4, where the photopeak efficiency is given as a function of gamma-ray energy. The efficiency at 279 keV (gamma ray from ^{203}Hg) is certainly lower than the value of the solid curve at the same energy; the curve is obtained by smoothing measured points except the point at 279 keV. The lack of counts of 279-keV gamma rays is probably caused by the adsorption of mercury on the inner side of the polyethylene vessel in mixing radioactive powder with non-radioactive powder, i.e., the procedure 3) described in the previous section; after the sample preparation, we found a very low, but not negligible, activities on the polyethylene vessel.

The efficiency at 279 keV for each vessel is estimated from the smoothing of other efficiencies measured for different energies. Final results of the efficiency for drum-type and Marinelli-type vessels are listed in Tables 5 and 6, respectively. Overall errors of the efficiencies estimated from uncertainties in solutions (forth column in Table 1) and statistical uncertainties in counting gamma-rays are also list in the tables.

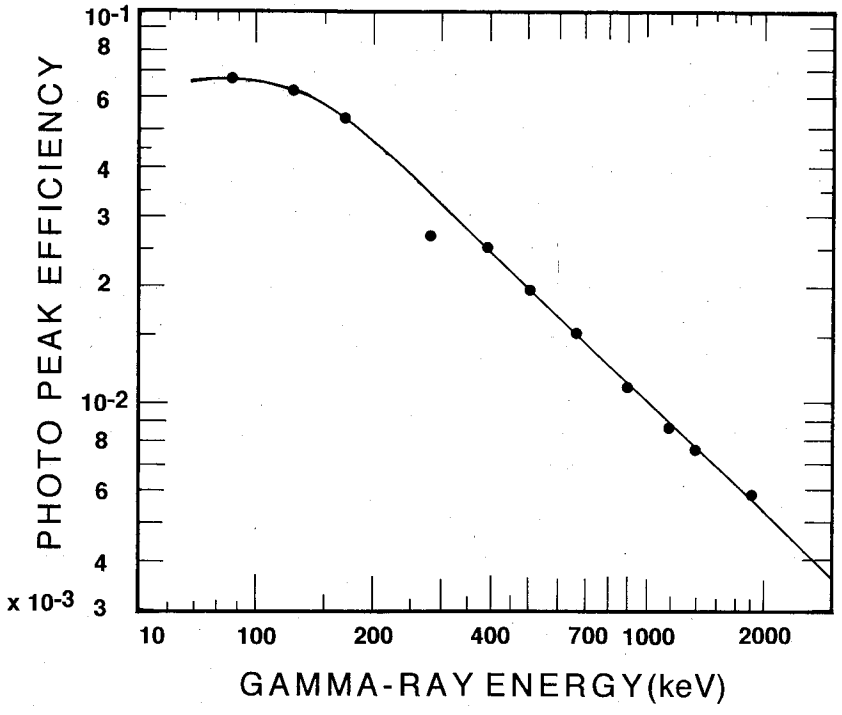

Fig. 4. Photopeak efficiency determined for the drum-type vessel D2 as a function of gamma-ray energy.

5. DISCUSSION

Total photopeak counts from a voluminal source is expressed by

$$N=t\cdot a\int_V C(\vec{r})\cdot\varepsilon(\vec{r})\mathrm{d}\vec{r} \quad , \tag{2}$$

where t is the measuring period, a is the emission rate per disintegration (/Bq) of gamma ray to be detected, V is the volume of the source (cm^3), $C(\vec{r})$ is the concentration of gamma activity (Bq/cm^3) and $\varepsilon(\vec{r})$ is the photopeak efficiency for unit volume of the source at the position $\vec{r}$. Assuming that $C(\vec{r})$ is constant in the source, we obtain

$$N=t\cdot a\cdot C\cdot V\cdot D \quad . \tag{3}$$

It is noted that D in Eq. (3), i.e.,

$$D=\int_V \varepsilon(\vec{r})\mathrm{d}\vec{r}/V \quad , \tag{4}$$

Table 5. Photopeak efficiencies of drum-type sources as a function of gamma-ray energy.

Energy (keV)	Vessel No D1	D2
88	0.1473	0.0655
	(3.3%)	(3.3%)
122	0.1384	0.0623
	(1.9%)	(1.8%)
166	0.1040	0.0502
	(1.5%)	(1.4%)
279	0.0636	0.0320
	(2.2%)	(2.1%)
392	0.0486	0.0238
	(4.2%)	(4.2%)
514	0.0367	0.0180
	(1.9%)	(1.8%)
662	0.0282	0.0141
	(2.3%)	(2.2%)
898	0.0194	0.0101
	(3.7%)	(3.7%)
1173	0.0151	0.0078
	(0.9%)	(0.7%)
1333	0.0132	0.0068
	(0.9%)	(0.7%)
1836	0.0098	0.0052
	(3.7%)	(3.6%)

Table 6. Photopeak efficiencies of Marinelli-type sources as a function of gamma-ray energy.

Energy (keV)	Vessel No. M1	M2	M3	M4	M5	M6
88	0.0548	0.0809	0.0738	0.0545	0.0640	0.0504
	(3.3%)	(3.3%)	(3.3%)	(3.3%)	(3.3%)	(3.3%)
122	0.0546	0.0788	0.0738	0.0547	0.0639	0.0507
	(1.8%)	(1.8%)	(1.8%)	(1.8%)	(1.8%)	(1.8%)
166	0.0465	0.0649	0.0614	0.0464	0.0537	0.0433
	(1.4%)	(1.4%)	(1.4%)	(1.4%)	(1.4%)	(1.4%)
279	0.0288	0.0415	0.0388	0.0291	0.0347	0.0271
	(2.1%)	(2.1%)	(2.1%)	(2.1%)	(2.1%)	(2.1%)
392	0.0219	0.0304	0.0294	0.0222	0.0252	0.0206
	(4.2%)	(4.2%)	(4.2%)	(4.2%)	(4.2%)	(4.2%)
514	0.0168	0.0234	0.0225	0.0170	0.0197	0.0159
	(1.8%)	(1.8%)	(1.8%)	(1.8%)	(1.8%)	(1.8%)
662	0.0133	0.0183	0.0176	0.0142	0.0153	0.0125
	(2.2%)	(2.2%)	(2.2%)	(2.2%)	(2.2%)	(2.2%)
898	0.0097	0.0132	0.0126	0.0097	0.0111	0.0091
	(3.7%)	(3.7%)	(3.7%)	(3.7%)	(3.7%)	(3.7%)
1173	0.0075	0.0102	0.0096	0.0075	0.0086	0.0070
	(0.7%)	(0.8%)	(0.7%)	(0.7%)	(0.7%)	(0.7%)
1333	0.0067	0.0090	0.0086	0.0066	0.0076	0.0062
	(0.7%)	(0.8%)	(0.7%)	(0.7%)	(0.7%)	(0.7%)
1836	0.0051	0.0068	0.0065	0.0051	0.0058	0.0048
	(3.6%)	(3.6%)	(3.6%)	(3.6%)	(3.6%)	(3.6%)

is equal to the photopeak efficiency defined by Eq. (1).

The background count under the photopeak in gamma-ray spectra is given by

$$N_B = 2E \cdot R \cdot n_B \cdot t \quad , \tag{5}$$

where E is gamma-ray energy, n_B is background counts per unit energy at E. R is energy resolution of high-purity germanium detector for gamma rays with the energy E, which is defined by a ratio of the half width of gamma-ray peak dE to the energy E:

$$R = dE/E \quad . \tag{6}$$

In Eq. (5), $2E \cdot R$ equals to twice the half width dE. When the intensity of gamma-rays is very weak, the photopeak is buried in the statistical fluctuation of the background count N_B. Therefore, to separate the photopeak from the background in the spectra, N is required to be larger than $N_B^{1/2}$, i.e., the statistical error of N_B:

$$N > N_B^{1/2} \quad . \tag{7}$$

Substituting Eqs. (3) and (5) in the above relation, we obtain

$$C > (2\ E \cdot R \cdot n_B)^{1/2}/(a \cdot V \cdot D) \quad , \tag{8}$$

which clearly shows a limit in determining the concentration of activities C. A possible way to improve the sensitivity for C is to increase $V \cdot D$ by using vessels of larger volume such as drum type and Marinelli-type; Marinelli beakers are more effective than drum-type vessels.

To estimate the limit, it is necessary to know other factors in Eq. (8). The resolution of high-purity germanium detector $dE(=E \cdot R)$ depends on E, e.g., 1.8–2.0 keV for 1.33-MeV ^{60}Co gamma ray; with proper standard gamma-ray sources, the resolution can be experimentally determined as a function of E. The measuring period t is practically the order of one day (8.64×10^4 sec) in the measurement for environmental samples. Since n_B strongly depends on E as well as kinds of samples, i.e., soil, water, dust and so on, it is not possible to estimate n_B theoretically. To obtain n_B, we are now examining background spectra of soil samples from various places including Indonesia. Results for the limit obtained with the vessels D1, D2 and M1-M6 will be given elsewhere.

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