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Determination of ^{134}Cs activity by the sum-peak method via a well type Ge detector

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Abstract

The new sum peak method was applied to the samples containing both ^{137}Cs and ^{134}Cs using a well-type HPGe detector. The standard sources containing ^{137}Cs and/or ^{134}Cs were measured via a well-type HPGe detector. The activities estimated by the new sum-peak method agreed with the certified activities of the standard sources. It was proved that the activities of the samples collected around Fukushima Daiichi Nuclear Power Plant can be estimated by the new sum-peak method.

Keywords

sum peak method, radioactivity, 134-Cs, 137-Cs, well type Ge detector

Introduction

Since in the decay of ^{134}Cs more than ten gamma-rays are emitted in several fast cascades, one should thoroughly pay attention to the true coincidence summing (TCS) effect [1,2]. Especially, the effect is severe for well-type detectors [2,8]. The full energy peak (FEP) counting efficiencies for ^{134}Cs may be decreased by more than a factor of two due to the coincidence summing, in comparison with the efficiencies at the same energies in absence of coincidence effects [2,8]. However, there is a kind of absolute measurement technique using the TCS effect, i.e.; the sum-peak method. The formula of the sum-peak method is as;

$$N_0 = \left(\frac{N_1 N_2}{N_{12}} + N_t \right) \overline{w}(0), \tag{1}$$

where N_0 is the activity of the source, N_1 and N_2 are the full energy peak count rates, N_{12} is the sum peak count rate, N_t is the total count rate, $\overline{w}(0)$ is the angular correlation factor, and subscripts 1 and 2 refer to the corresponding gamma rays [1,3-5]. For well-type of detectors, the angular correlation is approximately unity. The formula is rigorously valid for nuclides which decay emitting just two photons, 1 and 2, in a fast cascade, thus the sum-peak method is successfully applied to determine the activity of ^{60}Co, ^{22}Na etc. But since the method requires total count rate, N_t, it cannot be applied to sources containing other radionuclides. To overcome the problem, the authors developed the modified sum-peak method [6,7]. In this method, one should measure a sample with several source-to-detector (SD) distances to obtain an extrapolated value. Therefore, it cannot be applied to well-type detectors because sample should be usually positioned in the well of the detector.

Our purpose is to determine the activity of objects sampled after the accident of Fukushima Daiichi Nuclear Power Plant (FDNPP) [9]. Especially, small amount of objects like pollen, small plant or animal samples, etc. via a well-type detector. The samples mainly contain both ^{137}Cs and ^{134}Cs. Since the total count rate includes the contribution of both nuclides, the total count rate originated solely from the ^{134}Cs cannot be obtained directly from the measurement data.

To solve the problem and to estimate the activity of ^{134}Cs, the total count rate due to ^{134}Cs was estimated as below. The total number of counts originating from ^{137}Cs was guessed from its 662 keV peak counts by measuring a ^{137}Cs mono-nuclide source. By subtracting the contribution of ^{137}Cs from the total counts of the mixed source, the total count rate due to ^{134}Cs can be estimated. Therefore, the sum-peak method can be applied to estimate ^{134}Cs activity. The detail of the calculation is described in the previous article [8]. In this article, we explain the results of application of this method to calculate the activities of real samples collected around the FDNPP.

Theory

The decay scheme of ^{134}Cs is illustrated in Fig. 1, in which the gamma-rays with emission probability below 1% were omitted [10]. We decided to use the main two gamma-rays, 796 keV and 605 keV, for our calculation because of the higher emission probabilities than those of the other gamma-rays. The detailed calculation is explained in the previous paper [8]. Equation (2) is used to estimate the activity of a source containing only ^{134}Cs.

$$N_0 = \frac{N_{(796)} N_{(605)}}{N_{(796,605)}} + N_t \,, \tag{2}$$

where $N_{(796)}$ and $N_{(706)}$ are the peak count rates of 796 keV and 605 keV gamma-rays, respectively. $N_{(796,605)}$ is the sum peak count rate of 796 keV and 605 keV gamma-rays, and N_t is the total count rate. This is not exactly the sum-peak method, so we call this method the sum-peak-like method. On the other hand, for a source containing both ^{134}Cs and ^{137}Cs, the following procedure is used to compute the activity. At first, the total count rate originated from ^{137}Cs, $N'_{t(^{137}\mathrm{Cs})}$, is computed using the peak-to-total (P/T) ratio;

$$N'_{t(^{137}\mathrm{Cs})} = \frac{N_{(662)}}{P/T_{(^{137}\mathrm{Cs})}} \,. \tag{3}$$

Then the total count rate originated from ^{134}Cs, $N'_{t(^{134}\mathrm{Cs})}$, is computed by subtracting the total count rate originated from ^{137}Cs from the total count rate;

$$N'_{t(^{134}\mathrm{Cs})} = N_t - N'_{t(^{137}\mathrm{Cs})} \,. \tag{4}$$

Finally, substituting the total count rate computed by Eq. (4) in Eq. (2), the activity of ^{134}Cs can be computed using the following equation;

$$N_0 = \frac{N_{(706)} N_{(605)}}{N_{(796,605)}} + N'_{t(^{134}\mathrm{Cs})} \, . \tag{5}$$

We call the method the compensated sum-peak-like method.

Experimental

The well-type HPGe detector (GWL-120230, Ortec) has a sensitive volume of 120 cm^3, the well inside diameter and the active depth are 10 mm and 40 mm, respectively. The FWHM was 2 keV at 1333 keV. The gamma ray spectrum ranging from 0 to 2 MeV was converted to 4 kch digital data with an ADC (EASY-MCA-8K, Ortec), which means 0.5 keV/ch. The spectrum was analysed using a software (Spectrum Navigator, SEIKO EG&G).

The sources, ^{134}Cs, ^{137}Cs and their mixture, were prepared from the standard solutions calibrated by Japan Radioisotope Association (JRIA). Several microliters of the standard solutions were put into polyethylene tubes and weighed by a precision balance. The certified activity was estimated from the weight of the solution. Then several sources of various volume were prepared by adding into the tubes quantities in the range from 0.5 to 2.5 mL from a 100 mM CsCl solution. Six sources containing ^{137}Cs, six sources containing ^{134}Cs, and ten sources containing both ^{134}Cs and ^{137}Cs were prepared as listed in Table 1. The schematic diagram of the experiment is illustrated in Fig. 2.

The measurement time was determined such as to collect more than 10,000 counts in the 1401-keV sum peak of ^{134}Cs and the 662-keV peak of ^{137}Cs to reduce the statistical uncertainty below 1%. The apparent FEP efficiencies for the 605 keV and 796 keV peaks of ^{134}Cs and the 662 keV peak of ^{137}Cs were obtained using the prepared sources. The P/T ratios for ^{137}Cs were obtained from the sources containing only ^{137}Cs as a function of the sample height. The activities of the sources containing solely ^{134}Cs were estimated by the sum-peak-like method using Eq. (2). The activities of the sources containing both ^{134}Cs and ^{137}Cs were estimated by the compensated sum-peak-like method using Eq. (3-5). The estimated activities were compared with the certified activities.

Soil sampled on Jun 2016 at 10 km from FDNPP and mouse feces sampled on Oct 2016 at 3 km from FDNPP were measured by the relative method and the new method proposed in this study. The estimated activities were compared.

Results and discussion

The energy spectra of ^{134}Cs are illustrated in Fig. 3. A lot of peaks are observed in Fig. 3 (a), where the scale of the ordinate is logarithmic. The same spectra with linear scale is illustrated in Fig. 3 (b). Three main peaks, 605 keV, 796 keV and their sum peak are obvious, and the other peaks are negligible. The graph shows one of the proofs for understanding the calculation method using Eq. (2), namely the fact that in the case of well-type measurement the sum peak method can be applied with a good approximation to ^{134}Cs.

Table 2 shows the activities of the standard sources containing only ^{134}Cs. The activities estimated by Eq. (2), N_0, were compared with the certified activities, A_0. As seen in the table, the estimated activities almost agreed with the certified activities except for the sources higher than 30 mm.

Table 3 shows the activities of the standard sources containing both ^{137}Cs and ^{134}Cs. The upper six sources have almost the same activity of ^{137}Cs and ^{134}Cs, but different height. The lower four sources have the same sample height but different ratios of the activities of ^{137}Cs to ^{134}Cs. As seen in the table, the estimated activity agreed well with the certified activity.

As seen in Table 4, the activities of the soil and the feces estimated by the compensated sum-peak-like method agreed well with those by the relative method when the activity is higher than 25 Bq. ~~The reasons why the activity of sample with low activity estimated by the compensated sum-peak-like method is somewhat lower than that estimated by the relative method will be the background of natural radionuclides. For example, natural radionuclides ^{214}Bi emits 609 keV gamma-ray, and the HP-Ge detector has such peak in background. The gamma-ray affect the peak counts of 605 keV gamma-ray from ^{134}Cs. To correct enough counts for the sum-peak area, the counting time long; 50,000 s for soil #1,~~

~~524,000 s for feces b. And the peak counts for 605 keV area was not so high for low level samples. So that the peak counts would be underestimated. We considered that the new method should be applied to measure sample with relatively high activity.~~The reason why the activity of samples with low activity estimated by the compensated sum-peak-like method is somewhat lower than that estimated by the relative method, is the background of natural radionuclides. For example, the natural radionuclide ^{214}Bi emits a low intensity gamma-ray (1.3%) of energy 1401.5 keV, which can contribute to the sum peak count rate. Thus the denominator in Eq. (5) is increased due to this contribution and the computed activity is lower. This background contribution to the sum peak is negligible if the ^{134}Cs activity is not very low. But for low activity, when the counting time is very long, 50,000 s for soil #1, 524,000 s for feces b, this background contribution may have an effect. Thus, we consider that the new method should be applied to measure sample with relatively high activity.

Well-type detectors have high efficiency, so that they have a merit to measure small amount of samples. However, the measurements with this detector are considerably affected by the TCS effect [2]. One should carefully correct the TCS effect. The most effective way to avoid the TCS correction is to make measurements with respect to a standard containing the same nuclide, having the same matrix of the sample and the same geometry. But such standards are not always available, or rather expensive. However, if one obtains a sample with relatively high activity, one can estimate the activity using the method proposed in this work. Then one can obtain the apparent relative FEP efficiency using this sample. Samples with relatively low activity can be estimated using the relative measurement with respect to this sample.

The sum-peak method does not require standard sources. It can be applied to assess the activity of ^{134}Cs even if another nuclide, like ^{137}Cs, is also present in the sample, by the appropriate subtraction of total number of counts due to that nuclide. The method requires the knowledge of the P/T ratio of ^{137}Cs. However, most laboratory have ^{137}Cs standard sources for frequent calibration of the detectors and can easily obtain the P/T ratio.

The activities of the ^{134}Cs standard samples agreed well with the certified activities, depending on the sample height. The height limit is 25 mm. The ^{134}Cs activities of the mixture standard sources agreed well with the certified activity. The method is applicable to samples collected around the FDNPP .

Conclusions

The new calculation techniques; (1) the sum-peak-like method and (2) the compensated sum-peak-like method were proposed and were successfully applied to estimate ^{134}Cs activity via a well-type Ge detector. This method is an approximation and has limitations, but is quite simple and practical.

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Table 1 The certified activities of the standard samples

No.	Volume (mL)	Height (mm)	^{137}Cs (Bq)	^{134}Cs A_0 (Bq)	No.	Volume (mL)	Height (mm)	^{137}Cs (Bq)	^{134}Cs A_0 (Bq)
#701	0.005	1	507±5	–	#4701	0.005	1	520±5	487±4
#702	0.5	10	455±4	–	#4702	0.5	10	485±5	491±4
#703	1.0	18	423±4	–	#4703	1.0	18	527±5	478±4
#704	1.5	25	594±5	–	#4704	1.5	25	435±4	481±4
#705	2.0	30	456±4	–	#4705	2.0	30	398±4	482±4
#706	2.5	39	515±5	–	#4706	2.5	39	514±5	466±4
#401	0.005	1	–	490±4	#4712	2.5	39	978±8	490±4
#402	0.5	10	–	486±4	#4714	2.5	39	955±8	232±2
#403	1.0	18	–	496±4	#4716	2.5	39	1,020±8	143±1
#404	1.5	25	–	486±4	#4720	2.5	39	1,061±8	94±1
#405	2.0	30	–	488±4	–	–	–	–	–
#406	2.5	39	–	487±4	–	–	–	–	–

Uncertainty (k=1)

Table 2 The computed activities of the standard sources containing only ^{134}Cs

No.	^{134}Cs Certified activity A_0 (Bq)	^{134}Cs New method N_0 (Bq)	N_0/A_0 (%)
#401	490±4	475±18	97.0±3.7
#402	486±4	476±18	97.8±3.7
#403	496±4	475±18	95.8±3.7
#404	486±4	462±17	95.1±3.6
#405	488±4	454±17	93.1±3.6
#406	487±4	433±16	88.9±3.4

Uncertainty (k=1)

Table 3 The computed activities of the standard samples containing both ^{137}Cs and ^{134}Cs

Sample No.	^{134}Cs Certified activity A_0 (Bq)	^{134}Cs New method N_0 (Bq)	N_0/A_0 (%)
#4701	487±4	476±19	97.9±3.9
#4702	491±4	479±19	97.6±3.9
#4703	478±4	463±18	96.9±3.8
#4704	481±4	454±18	94.3±3.7
#4705	482±4	459±18	95.1±3.8
#4706	466±4	434±17	93.2±3.7
#4712	490±4	455±18	92.8±3.7
#4714	232±2	224±9	96.3±3.8
#4716	143±1	152±6	106±4
#4720	94±1	94±4	100±4

Uncertainty (k=1)

Table 4 The activities of the soil and the feces

Sample	Height (mm)	Weight (mg)	^{137}Cs by relative method	^{134}Cs by relative method A_0 (Bq)	^{134}Cs by new method N_0 (Bq)	N_0/A_0 (%)
Soil #1	3	4.30	77.2±1.4	10.5±0.2	9.8±0.5	93.4±5.4
Soil #2	4	17.26	318±6	43.3±0.8	45±2	103.1±4.8
Soil #3	4	26.79	484±9	65.8±1.3	68±3	103.7±5.0
Soil #4	4	8.98	187±3	25.4±0.5	26±1	102.2±4.7
Feces a	4	1.20	340±6	46.9±0.9	47±2	100.3±4.4
Feces b	4	1.50	10.5±0.2	1.44±0.03	1.21±0.07	84.4±5.0

Uncertainty (k=1)

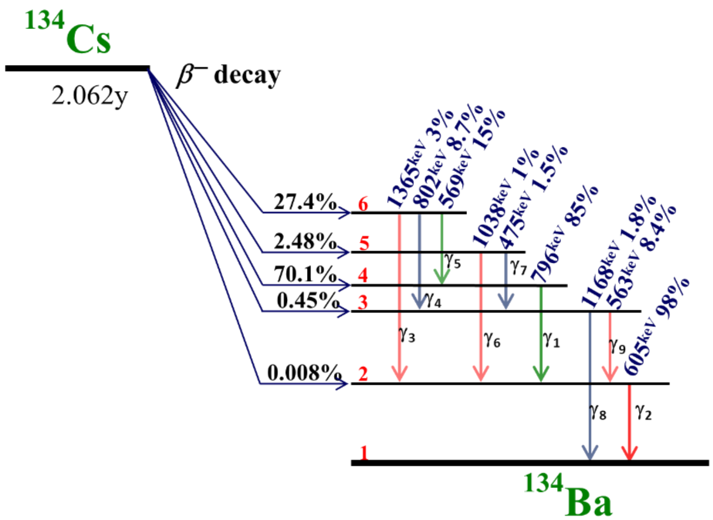


Fig. 1 Decay scheme of ^{134}Cs

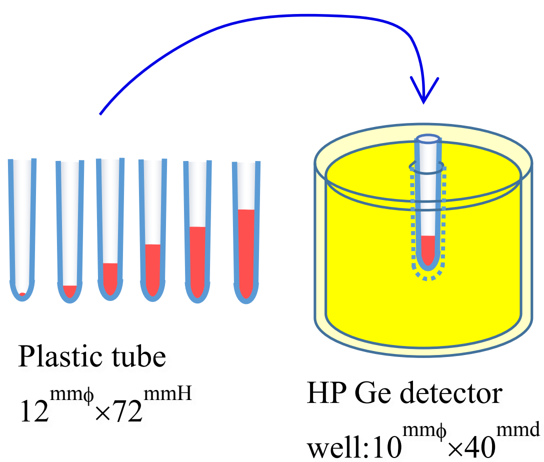
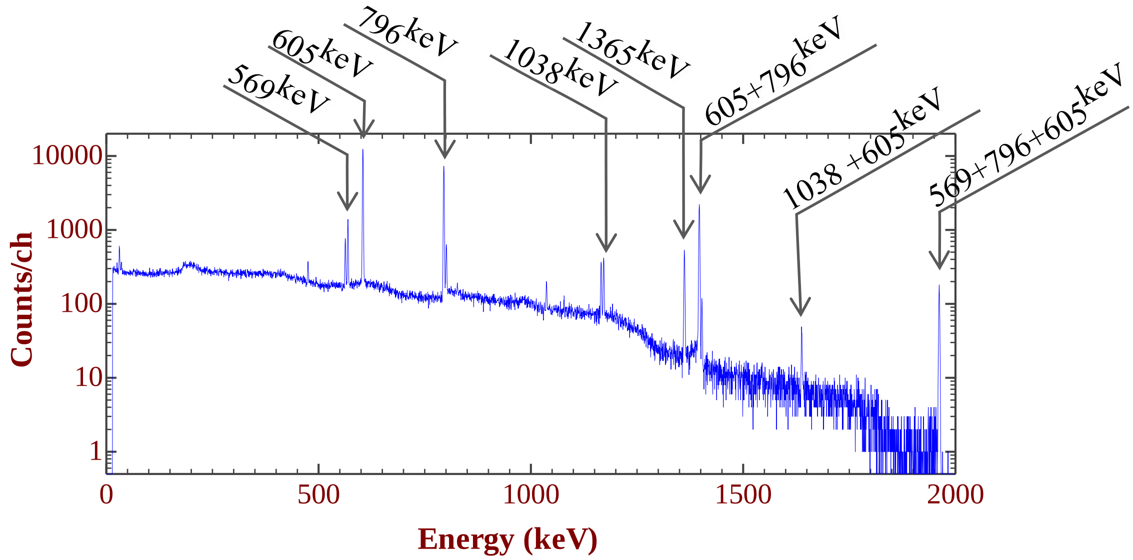
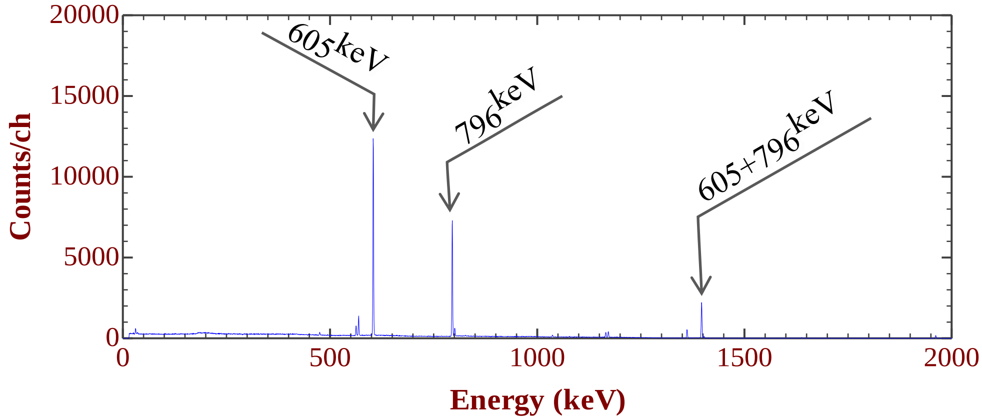


Fig. 2 The schematic diagram of the experiment



(a)



(b)

Fig. 3 Energy spectra of ^{134}Cs; ordinate with logarithmic scale (a) and that with linear scale (b)