Rapid Analysis of Gamma-Emitters Using Gamma-Ray Scintillation Spectrometer. I. Quantitative Analysis of ^{134}Cs—^{137}Cs *Mixture*

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Introduction

Rapid determination of individual species in a mixture of radionuclides will be required in many cases, such as processing of fission products, activation analysis, and chemical, biological or metallurgical experiments with several kinds of radionuclide.

Since the advent of electronic scintillation counting techniques, the application of scintillation spectrometers has been introduced to the study of analysis. Several investigations[1] on the analysis of radionuclide mixtures were reported, and the quantity of each nuclide was obtained from the area under the photopeak resolved by the spectrometer.

The authors have been investigating the possibility of quantitative determination of two nuclides which cannot be resolved by the spectrometer. This report is the result of the study on ^{134}Cs—^{137}Cs mixture.

Quantitative Measurement of ^{134}Cs—^{137}Cs Mixture

Principal gamma-energies emitted from ^{134}Cs[2] are 0.57 MeV, 0.60 MeV and 0.79 MeV, others being negligible in intensity. The pulse height scan shows two photopeaks at 0.79 MeV and at 0.57 MeV and 0.60 MeV (unresolved).

The gamma-energy of ^{137}Cs is 0.66 MeV; the pulse height scan shows one photopeak at 0.66 MeV.

In the presence of both ^{134}Cs and ^{137}Cs, two peaks appear as shown in Fig. 1. Let A be the area under 0.6 MeV photopeak (0.57 MeV, 0.60 MeV and 0.66 MeV photopeaks being superposed), then,

$$A=A_4+A_7$$

where A_4 and A_7 are the areas under the real photopeaks due to ^{134}Cs and ^{137}Cs near 0.6 MeV respectively. Or it is approximately obtained from the following expression,

$$A=\sum_a^b \gamma \times E$$

where

γ: counts per minute per channel width,

E: increment of energy equal to the channel width,

a and b: minimum points of the pulse height scan (Fig. 1).

So far as the geometry is fixed through out the measurement, and an identical apparatus is used, the pulse height scan normalized to the peak height of the principal gamma-energy is independent of the activity for a gamma-emitter[1]. Hence similar curves will be obtained by the

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1) R. E. Connally and M. B. Leboeuf, *Anal. Chem.*, **25**, 1095 (1953); D. H. Peirson, *Atomics*, Sept. 316 (1956); U. L. Upson, R. E. Connally and M. B. Leboeuf, *Nucleonics*, **13**, No. 4, 38 (1955).

2) J. M. Hollander, I. Perlman and G. T. Seaborg, *Rev. Mod. Phys.*, **25**, 552 (1953).

spectrometer scans for the gamma-emitter, and A_4 is calculated as follows:

$$A_4 = \gamma_{max} \times C$$

where γ_{max} is the height of the 0.79 MeV photopeak, which is not affected by the ^{137}Cs pulse height distribution, and C is a constant; the value of the latter is obtained from careful measurements of ^{134}Cs standard.

A and γ_{max} (A_4) are obtained from the pulse height scan; hence A_7 is calculated. Through the comparison of γ_{max} (or A_4) and A_7 with those of a mixture of ^{134}Cs and ^{137}Cs of a known composition, their quantities will be found.

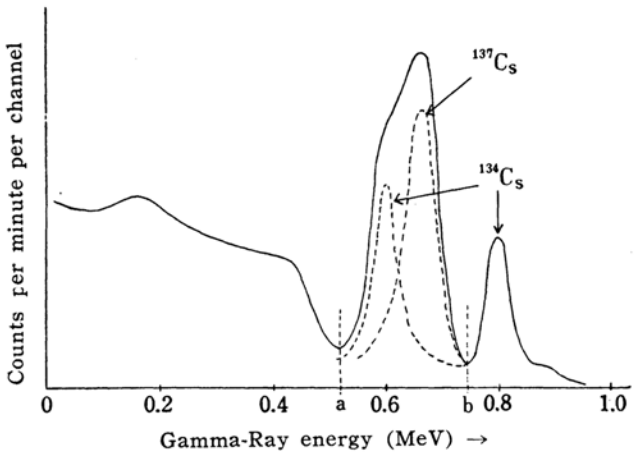

Fig. 1. Gamma-Ray spectrometer scan of ^{134}Cs—^{137}Cs mixture.

Experiment

The block diagram of the gamma-ray scintillation spectrometer (Atomic Instrument Co. Model 513) used in the experiment is shown in Fig. 2.

Before the quantitative measurement, the authors had ascertained the counting loss with ^{134}Cs and the results are shown in Fig. 3. The linearity of the counts vs. the quantity held up to 1×10^5 counts per minute in total counts and 5×10^3 counts per minute per channel width (0.02 MeV) in peak counts.

The chemical form of cesium was CsCl in aqueous solution. It was diluted to about 0.1 μC/ml with demineralized water. Mixed samples of ^{134}Cs—^{137}Cs were prepared as shown in Table I, taken in glass tubes of equal shape and size, and dried up. All residue remained on the bottom. Each glass tube has 15 mm outside diameter, which fits the geometry of the well of the NaI(Tl) crystal (see Fig. 2); it has 0.2g/cm^2 wall thickness. The absorption of the gamma-rays by the glass tube is undoubtedly negligible.

TABLE I
COMPOSITIONS OF SAMPLES

Nuclides	Quantities (ml)										
^{134}Cs	1	1	1	1	1	1	0	2	3	4	5
^{137}Cs	0	1	2	3	4	5	1	1	1	1	1

One ml. portion each of the ^{134}Cs and the ^{137}Cs were taken as arbitrary standards, which were carefully measured three times, The measuring system was switched on about three hours before the measurement for stabilization. Every sample was put into the well of the crystal, when its activity was measured.

The results obtained in 0.02 MeV channel width are shown in Table II and III. As their absolute activities were unknown, they were temporarily described by volume (ml.) of the solutions. The area under 0.6 MeV photopeak was calculated by summing up the counts from

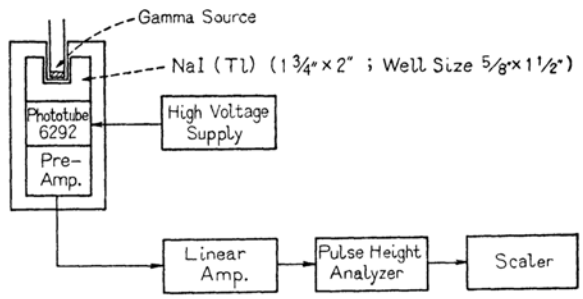

Fig. 2. Block diagram of scintillation spectrometer.

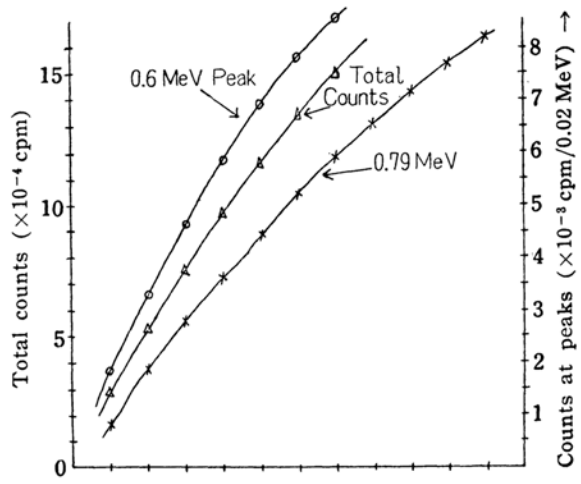

Fig. 3. Counting loss of the spectrometer.

a (0.51 MeV) to b (0.73 MeV) of Fig. 1, both of which were minimum points of the spectra.

Discussion

Concerning a mixture of ^{134}Cs and ^{137}Cs, the 0.79 MeV photopeak of ^{134}Cs is scarcely

TABLE II

ANALYSIS OF THE STANDARDS

Nuclides	Quantity (ml)	γmax (0.79 MeV) (cpm/V)	Area under 0.6 MeV peak (cpm)	$C\left(\frac{\text{Area}}{\text{max}}\right)$
^{134}Cs	1	572±3.2%	9.030±0.8%	17.13±3.3%
^{137}Cs	1	—	9.766±0.3%	—

TABLE III

ANALYSIS OF MIXTURES

Samples ^{134}Cs/^{137}Cs	γmax 0.79 MeV cpm/ 0.02 MeV	Area 0.6 MeV cpm	^{134}Cs A_4 cpm	^{134}Cs Quantity ml	^{134}Cs Error %	^{137}Cs A_7 cpm	^{137}Cs Quantity ml	^{137}Cs Error %
1/1	555	19,230	9,509	1.05	+ 5.3	9,721	0.99	− 0.5
1/2	532	23,126	9,080	1.00	+ 0.6	14,046	1.54	−28.0
1/3	537	32,388	9,165	1.01	+ 1.5	23,223	2.53	−20.7
1/4	633	44,182	10,804	1.19	+19.7	33,379	3.42	−14.5
1/5	505	48,841	8,619	0.95	− 4.5	40,222	4.14	−17.6
2/1	1,071	28,816	18,349	2.03	+ 1.6	10,467	1.07	+ 7.2
3/1	1,621	35,552	27,775	3.07	+ 2.5	7,777	0.79	−20.4
4/1	1,984	45,857	33,998	3.76	− 5.9	11,859	1.21	+21.4
5/1	2,694	56,713	46,160	5.11	+ 2.2	10,533	1.08	+ 8.1

affected by the ^{137}Cs pulse height distribution. It may be considered that this is the reason why the error for ^{134}Cs quantity is small and apt to come on the positive side. (see Table III).

Beta-rays emitted from ^{134}Cs (0.65 MeV: 75%, 0.09 MeV: 25%) and ^{137}Cs (0.51 MeV: 92%, 1.17 MeV: 8%)[2] are almost absorbed by the 0.2 g./cm^2 glass tube wall; about 80–90 % of beta-rays incident on the NaI (Tl) crystal are scattered out before being stopped[3]. Therefore the effect of beta rays would be negligible.

If a pulse height scan of a mixture of two nuclides shows two photopeaks and the high energy one involves two peaks of each nuclide, the quantitative determination will be a little more complicated.

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3) K. Siegbahn, "Beta- and Gamma-Ray Spectroscopy", North-Holland Publishing Co., Amsterdam (1955), p. 134.