

ACTIVITY DETERMINATION IN SAMPLES OF ISOTOPE MIXTURES BY USING SUM PEAKS IN Ge (Li) SPECTRA

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The basic concepts of the absolute activity determination based on sum peaks are briefly discussed. A possible extension of the method to measure the absolute activities of monoenergetic gamma sources is also given.

Introduction

The energies of coinciding gamma rays absorbed in a detector giving energy proportional signals are added up. Besides photopeaks, the spectrum also contains sum peaks as a result of coincidence. These sum peaks facilitate absolute activity measurements.

The basic formula for the absolute activity of a sample giving gamma rays in coincidence (derived by BRINKMAN et al [1]) is:

$$N = R + T = \frac{A_1 A_2}{A_{12}} + T, \quad (1)$$

where N is the absolute activity of the source, A_1 and A_2 are the measured count rates in the coinciding gamma peaks, A_{12} is the measured count rate in the sum peak, T is the area under the whole spectrum divided by the measuring time, and R is defined by Eq. (1). In the scintillation era the measurements were done in the well of well-type scintillators, so the dominant term in (1) was T . The poor resolution of the scintillation detectors made the determination of the peak area uncertain while these detectors have high efficiency by approximately 4π geometry. This technique was applicable only for the measurement of single isotopes. Using semiconductor detectors it is possible to perform the measurements far from the detector, when the dominant term is R , which is completely derived from the measured peaks, and the T term is only a minor correction [2]. This fact makes it possible to use the method for multiisotopic sources.

Let us briefly enumerate some advantages of this absolute activity measurement method, using modern detectors [3]: a) The measurable range covers several orders of magnitude, from about 10 Bq upwards. b) The formula

uses directly the raw measured quantities (A_1 , A_2 , A_{12} , T). c) No standard preparates are required. d) One saves instrumentation and time by getting the coincidence in a single detector, using a simple spectrometer instead of a complicated multiparameter system. e) No efficiency and absorption data are required. f) For pointlike sources the method is rather insensitive to the geometry (distance and angle between source and detector) [4]. g) The method can be used even for extended sources with distributed activity supposing that the source can be arranged approximately on the surface of a sphere, concentric with the detector. h) Due to the good resolution of semiconductor detectors, it is possible to determine the simultaneous absolute activity for several different isotopes in the same sample, provided that all the isotopes give distinct sum peaks. i) The calculation is simple.

Some drawbacks of the method are as follows. a) Unless at least one of the coinciding gamma ray lines is of 100 percent abundance, the basic formula needs correction for branching. b) Angular correlation effect should be considered. c) Clear sum peaks should be available. It is necessary to take the cross-over transition into account and, in any case, a proper dead time and pile up correction are needed against counting losses. d) A fundamental limitation is that the sum peak itself is necessary. This problem emerges with every isotope where the second, third etc. gamma transitions are very weak, or do not exist at all!

The combined sum peak method

In many practical cases the activity of isotopes having only a single gamma-ray should be measured. These isotopes have, of course, no sum peak, consequently, the sum peak method is not directly applicable [2].

In this paper we should like to show that in such cases the activity can be determined on the basis of the sum peak method.

1.) If the sample to be measured has at least one component with coinciding gamma rays, the absolute activity can be determined for this component which may appear further as an internal standard. This procedure compares the intensities of photopeaks, therefore the efficiency and the absorption effects should also be taken into account.

Such a combined absolute-relative method was applicable in the case of the G-2 activity intercomparison measurement organized by the International Atomic Energy Agency. The task was to measure the activity components of five mixed sources, each composed of ^{57}Co , ^{133}Ba , ^{54}Mn and ^{65}Zn , having different activities. While the ^{133}Ba isotope has more gamma rays in coincidence, its absolute activity was easy to determine by the help of the sum peak method. As a second step this activity was used for the relative activity determination by the comparison of photopeak intensities. The relative efficiency of the

Table I
Measured and nominal activities of the G-2 activity intercomparison standards

Source	^{137}Ba		^{54}Co		^{54}Mn		^{65}Zn	
	A_{nom} [kBq]	A_{meas} [kBq]	A_{nom} [kBq]	A_{meas} [kBq]	A_{nom} [kBq]	A_{meas} [kBq]	A_{nom} [kBq]	A_{meas} [kBq]
MIXO8-1	20.5	21.2	15.6	16.8	22.0	23.4	44.6	46.5
MIXO8-2	60.2	63.0	44.1	48.5	62.4	63.0	126	132
MIXO8-3	132	129	96.2	99.3	130	129	264	258
MIXO8-4	198	210	144	162	206	222	427	451
MIXO8-5	270	298	194	226	274	312	562	627

detector, and the probabilities of gamma-ray emission were to be taken into consideration (Table I). As it is seen all measured values are in good agreement with the nominal activities.

2.) If the sample to be measured has no component with coinciding gammas, the task is somewhat more complicated. An external source should be placed close to the sample for standardization. Using the photopeak efficiency calibration curve of the detector, the other activities can be determined relative to that of the standard. This part of the procedure is of course very sensitive to the measuring geometry. If they are significant, absorption corrections also must be made.

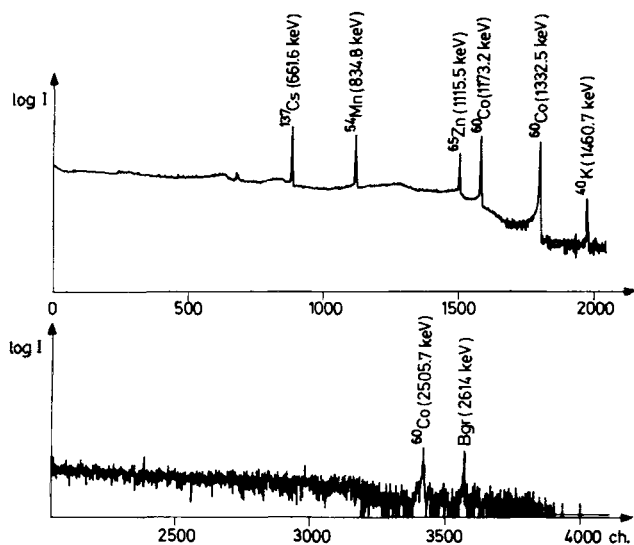


Fig. 1. The complex spectrum of ^{60}Co , ^{54}Mn , ^{137}Cs and ^{65}Zn standard sources.

Table II

Measured and nominal absolute activities using the parallel and the serial method

Isotopes	Nominal activity [kBq]	Measured activity [kBq]		Calculation method
		"Parallel" method	"Serial" method	
^{60}Co	8.89	8.95	9.15	from sum peak
^{137}Cs	9.83	9.31	9.55	relative
^{54}Mn	8.11	7.61	7.47	to ^{60}Co
^{65}Zn	5.22	5.55	5.24	

To test this method we took a collection of standard gamma ray sources* of small volume and put them tightly together, so approximating a point like mixed isotope source, which was measured by a Ge(Li) detector of 12% efficiency. To measure the absolute activity the ^{60}Co spectrum was measured in "parallel" with the spectra to be measured. Fig. 1 shows the spectrum obtained in this way. Table II gives the nominal and mesured absolute activities of the sources.

Table II shows that the measured activity values are approaching the nominal activities quite well. At the same time, the influence of the geometrical uncertainty can be seen in the larger deviation of the value pairs.

In one respect the combined absolute-relative sum peak method explained above needs a relative photopeak efficiency curve (Fig. 2) for the applied geometry. On the other hand, to assess the absolute activity we have to know the T area as a part of the whole spectrum, belonging to the standardising isotope. While it is impossible to determine it from a complex spectrum having more components, it is useful to measure the peak-to-total ratio separately,

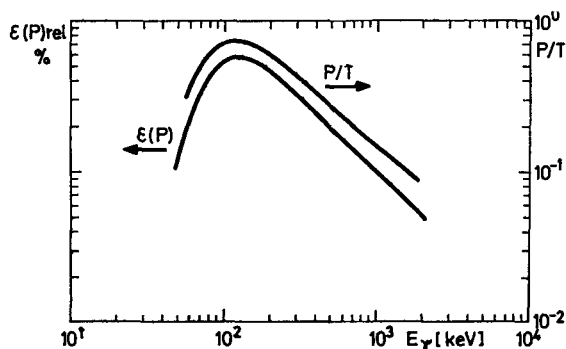


Fig. 2. Peak-to-total and relative efficiency curve of the detector.

* The sources were made available and certified by the National Office of Measures (OMH)

using isotopes emitting monoenergetic gamma rays at different energies. This curve in Fig. 2 may help us to calculate Eq. (1). Otherwise the T term may be measurable, if we have the different isotopes in separated sources, and measure all of their spectra successively in the same, strictly known measuring geometry ("serial method", see last column in Table II).

The estimated error of the measured activity values is generally 5–10%, due to some statistical errors in the evaluation of spectra, positioning errors, the uncertainty of efficiency and peak-to-total curves, angular correlation ambiguities, etc.

Conclusions

The sum-peak coincidence method, using a simple spectrometer consisting of a high resolution, large volume (high efficiency) Ge(Li) detector and a multi-channel analyser, is suitable to measure absolute activity in a flexible way. Coincidence events without any coincidence circuit can be measured by this method by using a single detector only. As an absolute method, it gives the activity of the measured isotopes without any intensity standard sources. It seems to fit well into health physics laboratories, where it is often necessary to measure many kinds of isotopes over a very large range of intensity.

While the original method can only be used for isotopes having two coinciding gamma rays, the combined absolute-relative method discussed in this paper can also be applied to monoenergetic gamma-ray sources. Activity determination is possible in many cases with an uncertainty less than 10%. In health physics practice such accuracy is sufficient in most cases. The method measures absolute activity in a very simple way, relatively quickly and very cheaply.

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