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SPECTROMETRY AS APPLIED TO THE CALIBRATION
OF STANDARD SOURCES.

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A B S T R A C T

A brief discussion is presented of the determination of absolute gamma emission rates for monoenergetic gamma emitters using a single-channel scintillation spectrometer. Examples are given for Cr^{51} , Cs^{137} , Nb^{95} , and Zn^{65} . Sources of error are discussed and steps being taken to improve the accuracy of the method are described.

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I. INTRODUCTION

The use of scintillation spectrometers for the detection of radiation of all types is finding increased application. While this type of equipment has been generally restricted to use as a fundamental research tool, the advantages to be gained through the application of scintillation phosphors to routine counting and monitoring problems have long been recognized. In particular, the use of the energy-sensitive response of these phosphors permits identification of particular radionuclides by beta or gamma energy determination. Knowledge of the response of these phosphors to monoenergetic gamma radiation permits the measurement of absolute gamma emission rates as well as positive isotopic identification.

This report is a brief discussion of absolute-gamma counting with a NaI (Tl) scintillation spectrometer, together with a graphical presentation of the physical constants of the detector. As an illustration of the application of these techniques, a series of monoenergetic gamma-ray standards has been prepared for use in the calibration of the high pressure ionization chamber in the MTR counting room.

II. EXPERIMENTAL PROCEDURE

Point sources are prepared either directly from irradiated solid materials or by dilution of stock solution in the case of materials obtained from Oak Ridge. These sources are prepared on standard sample cards to insure reproducible geometry. A gamma spectral analysis may then be made for each source using a single-channel gamma scintillation spectrometer. The data obtained from the spectrometer are in the form of pulse-height spectra obtained by analysis of the amplified output of the detector. The information plotted is the counting rate per unit pulse height interval as a function of pulse height, where the pulse height is in arbitrary units proportional to energy. (See Figures 4-7.)

The scintillation detector produces a number of photons which are indicative of an amount of energy liberated within the crystal. In the detection of gamma rays, this energy is the kinetic energy of the electrons produced by the interaction of photons via the photoelectric, Compton or pair processes. The photomultiplier tube converts these light pulses to a voltage pulse spectrum, which may be amplified and analyzed to represent the

energy liberated by each individual event occurring within the crystal.

In the detection of a monoenergetic gamma ray whose energy is below the pair production threshold, the resulting pulse distribution includes a group of pulses representing the full energy of the incident gamma (interaction by photoelectric process) and a "smear" of pulses representing Compton recoil electrons of all energies up to an energy $E_{\max}$ given by:

$$E_{\max} = \frac{E_{\gamma}}{1 + \frac{mc^2}{2E_{\gamma}}},$$

where E_{γ} = incident gamma energy and $mc^2 = 0.511$ mev.

The number of electrons in this distribution per unit energy increment is fairly uniform, rising somewhat near the end point, then dropping sharply to zero at the maximum electron energy.

At gamma-ray energies above the pair production threshold, interaction by the pair process results in pulses corresponding to the kinetic energy of both the positron and electron. These pulses will correspond to an energy 1.02 mev less than the energy of the incident gamma. Additional pulses are observed corresponding to energies from 0.511 mev less than the full incident energy up to the full energy due to detection of one or both of the quanta from the annihilation of the positron.

Also present in each of these gamma-ray spectra is a pulse-height distribution resulting from radiation scattered into the crystal by the surrounding material (lead shielding, photomultiplier tube, etc.). The peak of this distribution will correspond to the energy of the secondary gamma produced by 180° Compton scattering of the incident gamma ray, and is referred to as the "back-scatter peak." This extraneous distribution does not represent a true contribution to the response of the detector for the incident monoenergetic radiation.

The accumulation of a large number of pulse-height spectra for various monoenergetic gamma sources in several source-detector configurations permits the synthesis of a series of "shapes" for the response of a given detector to the monoenergetic gamma radiation from the source only.* This requires determination of the spectral shape of the background radiation and scattered Compton gammas. These contributions to the gross spectra must be accurately determined and subtracted. Once these spectral shapes have been determined for a standard size crystal and various source-detector configurations, they may be used in the analysis of complex gamma spectra by successive subtractions from the gross spectrum.

* For a theoretical discussion, see: "Line Shapes of Monochromatic Gamma-Ray Spectra in Scintillation Spectrographs," Maeder, Muller, and Wintersteiger, Helv. Phys. Acta, 27, (March, 1954).

III. CALCULATION OF THE INTEGRAL COUNTING RATE

The total number of gammas from the source detected by the crystal can be obtained by dividing the area under the true pulse-height spectrum by the unit pulse-height increment (window width).

In the simple case of radio isotopes emitting only one gamma ray, it is possible to determine the total number of gammas detected by consideration of the "photoelectric peak" only. From experimentally determined shapes discussed above, a quantity P may be determined. P is defined as the ratio of the area under the photoelectric peak to the area under the total pulse-height distribution for "pure" spectra. The value of this quantity will be a function of both geometry and gamma energy. If a gamma photon experiences a series of interactions in the crystal, the light pulses will occur within the light collection time of the phosphor and will result in one voltage pulse liberated in the crystal. Since the probability for photoelectric interaction increases with degradation of the gamma energy, the usual result is a transfer of pulses from the Compton distribution into the full energy peak. Hence, P is not simply a ratio of the primary Compton and photoelectric cross sections.

Measurements of P, the "peak-to-total ratio," have been made by Heath at ORNL.⁽¹⁾ The results of these data are shown in Figure 1. These measurements consider only the case of a point source at a distance of 1.0 cm on the central axis of a standard 1 1/2" diameter x 1" cylinder NaI (Tl) crystal. Since the probability of the occurrence of multiple processes is a function of source-detector configuration, the peak-to-total ratio will also vary with source position.

Since the photoelectric peak from a monoenergetic gamma pulse-height distribution follows a Gaussian distribution, as a result of photomultiplier statistics, the area under the photo peak (A_p) is given by:

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1. R. L. Heath, P. R. Bell, R. C. Davis, Physics Quarterly Progress Report, September 20, 1952, ORNL-1415, p. 10.

Also see: R. Stephenson and P. R. Bell, Physics Division Semiannual Report, ORNL-1705, p. 29, and Maeder, et. al., op. cit.

$$A_p = 2.129 N_m a; \quad (1)$$

where N_m = the peak counting rate in counts/second;

a = the 1/2 width at 1/2 maximum of the photo peak in pulse-height units (PHU);

and A_p is expressed in units of counts/second*(PHU).

The integral counting rate for a single gamma ray can now be obtained as follows:

$$I = \frac{A_p}{W P} = \frac{2.129 N_m a}{W P}; \quad (2)$$

where I = integral counting rate in counts/second;

W = measured width of the "window" of the single-channel analyzer in PHU;

and P = peak-to-total ratio.

IV. CALCULATION OF THE GAMMA DISINTEGRATION RATE

It is possible to calculate the detection efficiency of a cylindrical NaI crystal for a point source of monoenergetic gamma radiation located on the axis of the crystal. It is assumed that no losses occur between the source and the crystal and that the radiation detected by the crystal is directly proportional to $1-e^{-\gamma x}$ where x is the distance the radiation travels through the crystal and γ is the appropriate linear absorption coefficient for NaI. The detection efficiency of the crystal is given by:

$$T(E) = 1/2 \int_0^{\tan^{-1}(r/h)} \left[1 - e^{-\gamma(E) x(a)} \right] \sin a \, da \quad (3)$$

where a = angle between a ray from the source and the perpendicular from source to crystal;

r = the radius of the crystal;

t = the thickness of the crystal;

h = the source distance;

and

$$x(a) = \frac{t}{\cos a}, \text{ for } 0 \leq a \leq \tan^{-1} \left(\frac{r}{h+t} \right);$$

$$x(a) = \frac{r}{\sin a} - \frac{h}{\cos a}, \text{ for } \tan^{-1} \left(\frac{r}{h+t} \right) < a \leq \tan^{-1} \left(\frac{r}{h} \right).$$

Equation (3) can be evaluated by numerical integration. This has been done⁽²⁾ for the case:

$$t = 2.54 \text{ cm};$$

$$r = 1.90 \text{ cm};$$

$$h = 0.2, 5, 1.0, 2.0 \text{ and } 5.0 \text{ cm};$$

$$\tau = 600, 10.0, 2.0, 1.0, 0.5, 0.3, 0.2, 0.15, 0.13 \text{ and } 0.123 \text{ cm}^{-1}.$$

Curves have been prepared (Figure 2) displaying T as a function of τ for the above source distances.

The value of $\tau(E)$ chosen should be the total absorption coefficient corrected for the contribution due to coherent (Rayleigh) scattering which results in no energy loss in the crystal and, hence, does not contribute to the detection of the photon. White⁽³⁾ has calculated the total absorption coefficient for sodium iodide and her values have been plotted on the solid curve of Figure 3. Bell, Heath, et al⁽⁴⁾ have measured the absorption of monoenergetic gammas in NaI absorbers. Their values are shown on the dashed curve of Figure 3. Their experimental setup, however, was such that some of the forward coherent scattered photons could reach the detector. In order to compare their values with the total absorption calculations of White, the coherent scattering cross section must either be measured or computed and the correction to be added to the measured values must be estimated from the geometry. This correction has been estimated to be of the order of one

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2. Unpublished data, ORNL math panel. (Note added in proof) Tables of this function have recently been prepared for various source distances for 1 1/2" x 1" and 3" x 3" NaI crystals, and are available from F. K. McGowan, ORNL.
 3. G. R. White, X-Ray Attenuation Coefficients from 10 Kev to 100 Mev, NBS-1003, (May 13, 1952), p. 76.
 4. P. R. Bell, et al, Physics Quarterly Progress Report, September 20, 1952, ORNL-1415, p. 8. Also see: ORNL-1705, op. cit., Howland and Kreger, Phys. Rev. 95, 407 (July 15, 1954), and H. S. Paul, Gamma-Ray Absorption Coefficient at 6.13 Mev, HW-31123.

or two per cent and, when made, leaves a discrepancy of several per cent between measured and calculated values of the total absorption coefficient. Until such time as this discrepancy can be resolved, we shall use values based on the experimental curve since the theoretical values, even if correct, would still require correction for coherent scattering;* whereas the experimental values should be closer in that respect.

In the series of measurements herein described, the crystal used was not the standard 3.80 cm diameter by 2.54 cm thick on which calculations of efficiency were based, but was 3.82 cm by 2.66 cm. Because of the difficulty in performing the calculations for efficiency, an approximation has been made to correct for this systematic error. The average path lengths of photons through the standard and the larger crystal were calculated to be 1.91 and 1.98 cm, respectively. The ratios

$$\frac{1-e^{-1.91}}{1-e^{-1.98}}$$

were calculated for each source and used as correction factors.

V. RESULTS

Each of the sources was measured using 1/16" of Polystyrene and the 0.028 cm of the aluminum crystal can as beta absorbing material. The spectra obtained from each source are displayed in Figures 4 through 7. The data obtained and the calculated numbers are shown in Table I. The assigned errors shown in Table I are estimated probable errors for all the original data, except that for N_m , which is the statistical probable error. Errors in calculated values were compounded by the usual method.

It will be noted that a major contribution to the error calculated is due to the assignment of a relatively large error to the value of γ , the absorption cross section. As discussed previously, the disagreement between measured and calculated values of γ remains a point of some concern. The large error in the Cr^{51} measurement is due to the assignment of a greater error to P, since a source distance of 5.1 cm was used; whereas all determinations of P were made at 1.0 cm.

* Since this report was written it has come to our attention that such corrections have been applied at ORNL (private communication from F. K. McGowan).

TABLE I
DATA AND CALCULATED RESULTS

Source	E (mev)	N _m (c/sec)	a (PHU)	P	I (c/sec)	γ (cm ⁻¹)	T(E)	T'(E)	$\frac{R}{\gamma}$'s/sec x 10 ⁻⁴	Time of Measurement
Cr ⁵¹	.320	525. ±5.0	38.0 ±1.0	.75 ±.08	3800. ±400.	.490 ±.030	.0175 ±.0005	.0175 ±.0005	22.0 ±3.0	1300 1-12-54
Cs ¹³⁷	.663	675. ±7.0	36.0 ±1.0	.34 ±.02	10300. ±640.	.275 ±.015	.074 ±.004	.076 ±.004	140.0 ±1.2	12-7-53
Nb ⁹⁵	.758	61.0 ±0.6	34.0 ±1.0	.32 ±.02	930. ±57.	.232 ±.014	.069 ±.004	.071 ±.004	1.3 ±0.1	0900 11-9-53
Zn ⁶⁵	1.11	86.0 ±0.9	40.0 ±1.0	.22 ±.01	2250. ±140.	.192 ±.010	.057 ±.003	.059 ±.003	3.8 ±0.3	11-10-53

E = gamma energy

N_m = peak counting rate

a = half width at half-maximum of photopeak (pulse-height units)

P = ratio of area under peak to area under "pure" total spectrum

I = integral detected counting rate (calculated)

γ = linear absorption coefficient

T(E) = detection efficiency

T'(E) = detection efficiency corrected for crystal size

$\frac{R}{\gamma}$ = gamma emission rate

w = window width of single channel analyzer = 15±0.3 PHU

FIG. 6

11-9-53
 NB95 γ SPECTRUM
 SOURCE DIST. 1.1cm.
 1/16" POLY. ABS.