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# The Use of Low Level Gamma Scintillation Spectrometry In The Measurement of Activity In Human Beings

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## ERRATA

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L. D. Merinelli

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## THE USE OF LOW LEVEL GAMMA SCINTILLATION SPECTROMETRY IN THE MEASUREMENTS OF ACTIVITY IN HUMAN BEINGS<sup>1</sup>

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### INTRODUCTION

THE development of gamma ray scintillation spectrometry at the Argonne National Laboratory came as the inevitable extension of our use of sodium iodide counters in the place of G.M. detectors formerly used by Evans (1) in the estimate of Ra (B + C) activity in industrial workers.

This substitution, the advantages of which became obvious after Hofstadter's work on the scintillation properties of sodium iodide (2), was enacted as soon as the volume of crystals commercially available exceeded a few cubic inches. At that time, pulse-height analyzers were restricted to single channels of variable width, hence detailed spectra of the signals could not be obtained in a reasonable time in the presence of very weak sources. The early work at ANL was, therefore, limited to estimates of Ra (B + C) based on large bands of pulse-height spectra emitted from patients; and our concern was largely centered on the appraisal and improvement of the one-meter-arc technique and on

<sup>1</sup> This work was performed under the auspices of the United States Atomic Energy Commission.

the use of collimators in the study of lung transport of  $\text{RaSO}_4$  dust accidentally inhaled.

Despite the crude analytical tools available, it soon became evident that the sensitivity and general usefulness of NaI counters could be enhanced significantly by exploiting their spectrometric properties and by reducing drastically the interference of the natural radioactivity of the given surroundings. It became obvious also that both large crystals and multiple channel analyzers were in

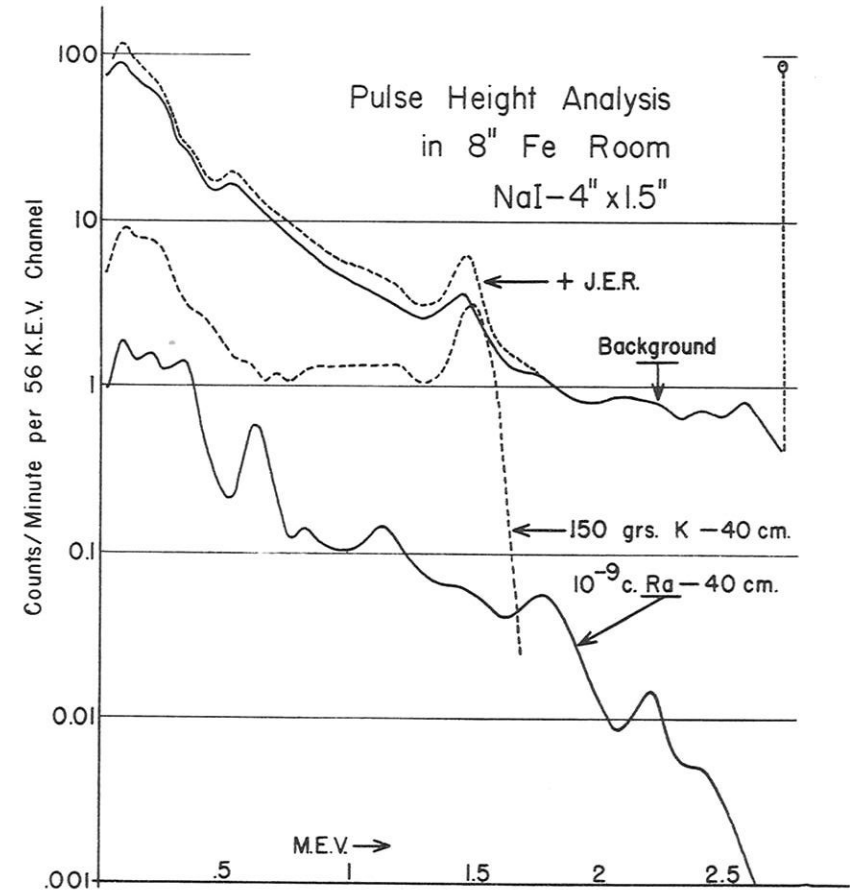


Figure 1. Pulse-height spectra obtained with a 4 by 1.5 in. NaI crystal in an iron shielded room in feasibility study of measurement of gamma ray activity in unexposed human beings. Lower curves were obtained with bare  $\text{K}^{40}$  and  $\text{Ra}^{226}$  sources at the stated distance. Dotted line above background shows spectrum obtained with one of the authors seated near the crystal (1955).

order. Since the cost of both these components was very high, exhaustive mock-up tests were made and these indicated that the tasks were feasible with a 4 by 1½ in. crystal, the largest available at the time. This crystal was connected to a 24 channel pulse analyzer and enclosed in an iron shielded room of roughly 2 M. cube with a wall thickness of 8 in.

Preliminary results obtained in 1955 were sufficient to establish the general features of the spectrum of gamma radiation emitted by normal human beings which, as shown in Figure 1, definitely established  $K^{40}$  as the principal emitter. Since then, many studies have been pursued to improve precision and to ascertain, eliminate or reduce the sources of counter background and to investigate the usefulness of coincidence techniques.

### CRYSTAL DETECTOR ASSEMBLY

Essential limitations of NaI counters are their unadaptability to varied shapes and their high initial cost which precludes their use in four pi geometry.

Besides cost, the choice of size and number of crystals to be used is dictated by the need of achieving satisfactory precision in manageable counting time. If one decides to measure the amount of potassium in the body with a single crystal, a rough estimate of its dimension can be made by considering that this source emits about 28,000 gamma rays per minute. It can be shown by simple calculation that to obtain a precision of 5% in 10 min. a single NaI crystal—situated at about 40 cm. from such a source, and in the presence of a background four times as intense—must have a volume of about 1600 cc. The cost of such a crystal is therefore considerable and attention must be paid to the most economical use of its mass.

If the only aim in the use of these instruments were the attainment of the maximum number of counts in the presence of given types of photons, simple considerations of exponential absorption and of the solid angle subtended by the detector would lead to the conclusion that greatest economy would result from the use of thicknesses small relative to the mean free path of the photons in question; the ultimate choice of the crystal face area would then be based on a compromise between experimental proficiency and the

maximum allowable cost. This simplified approach is rather unrealistic because, in the measurement of gamma rays from the "uncontaminated" human body, one is already faced with the presence of at least two dominant gamma rays of .66 Mev ( $Cs^{137}$ ) and 1.46 Mev ( $K^{40}$ ) in the presence of a continuous pulse background. If, to unravel more complicated situations, the necessity of a crystal spectrometer is conceded and the lack of any sophisticated spectral

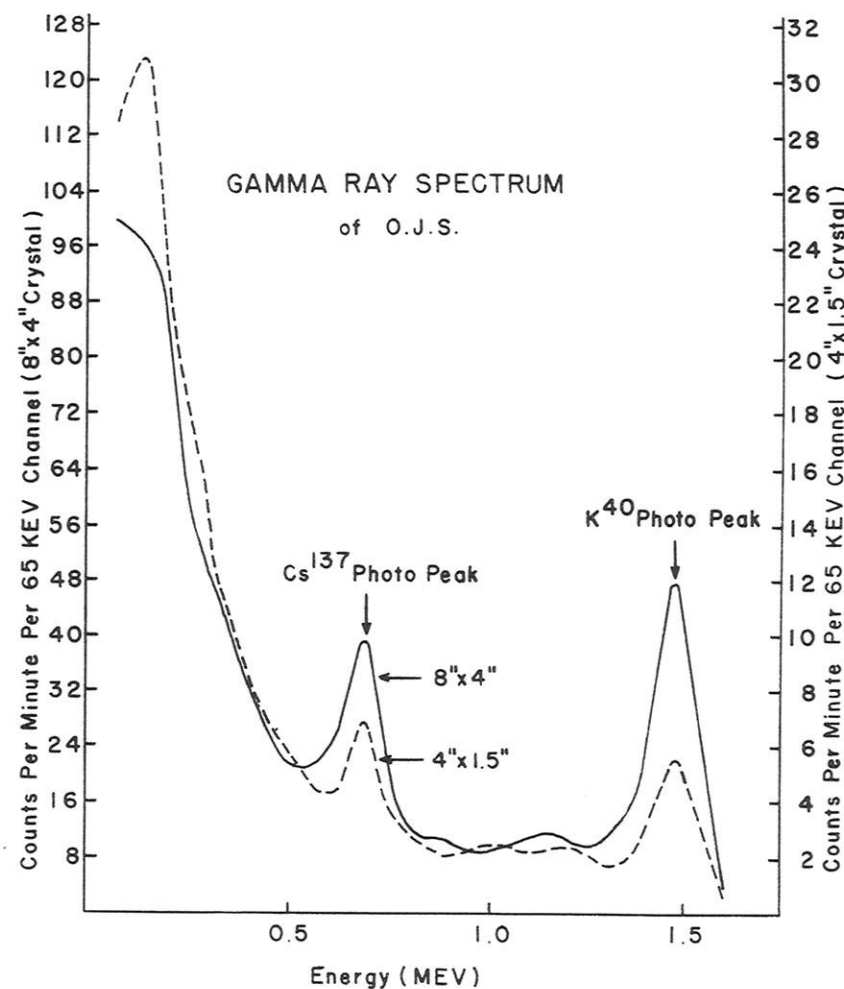


Figure 2. Net spectra of an uncontaminated subject obtained with 4 by 1½ in. and 8 by 4 in. crystals plotted to scales designed to equalize the solid angles subtended by the body at the crystals.

analysis is acknowledged, then one should regard as most important the capability of a large crystal mass to gather within a single photo peak most of the scintillations resulting from monoergic photons striking its face. This fact is illustrated in Figure 2 where spectra from a single uncontaminated individual obtained with crystals for 4 by 1.5 in. and 8 by 4 in., respectively, are shown—the counting rates being normalized to equal frontal crystal area. The curves clearly show that the photo peaks are greatly enhanced and that the Compton continuum is, if anything, somewhat depressed in the spectrum obtained with the thicker crystal. This constitutes a decided advantage in the identification of gamma ray activities smaller than those emitted by natural potassium.

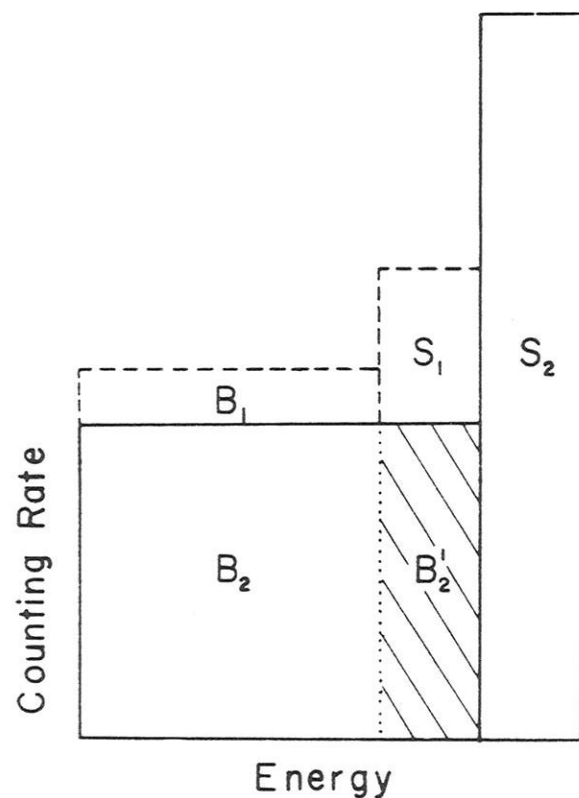


Figure 3. Simplified representation of pulse-height spectra of two gamma rays of about equal energy ( $E_1 \cong E_2$ ).  $S$  and  $B$  refer to the total counts (areas) in the "photo peak" and continuum, respectively.

If one assumes that the spectrum exists as it is roughly shown in Figure 3, (namely, that the background  $B_2'$  is part of the spectral continuum of an adjacent gamma ray of slightly higher energy and of intensity higher by a factor roughly equal to the reciprocal of the photo peak resolution)<sup>2</sup> one obtains as a figure of merit the expression:

$$S \cdot p / (2 - p)$$

where  $p$  is the photo peak fraction and  $S$  is the area under the photo peak.<sup>3</sup>

The photo fraction  $p$  depends upon thickness and total mass of the crystal, but it cannot be expressed as an analytic function of these variables. We have used the values obtained by W. F. Miller (3) and, in order to determine the most *economic* use of NaI, we have normalized the f.o.m. to unit mass of material. One then comes to the conclusion that for crystals varying from 4 to 8 in. in diameter the optimal thicknesses are about 1 in. around 300 keV, 2 in. at about 700 keV and 4 in. from 1.3 to 2.6 MeV radiation.

It should be realized, however, that the single crystal approach to whole body measurements is not the only possible approach. A single large crystal does offer in practice considerable simplification in instrumentation but cannot be used at average distances smaller than 40 cm. from the body. Comparison of our data, obtained with an 8 by 4 in. crystal, with those published by Rundo (4), who employs four crystals of 4 by 2 in. dimension, proves that the multiple crystal can operate at a shorter effective distance from the body and that, in certain cases, it may prove more convenient in a rough approach to localization of contamination in the human body. This advantage, however, is likely to be obtained at the expense of reduced sensitivity in quantitative estimates when the

<sup>2</sup> This may be representative of the situations faced in the presence of contaminations of the order of maximum permissible levels; that is, well above natural background.

<sup>3</sup> We assume: (a) that the resolutions of the two peaks are small, and approximately equal ( $R_1 \cong R_2 \ll 1.0$ ); (b) that the corresponding photo fractions  $p = S / (S + B)$  may also be considered equal; and (c) the intrinsic background is negligible. The usual figure of merit (f.o.m.) is then  $S_2^2 / (S_1 + 2B_2')$  where  $B_2'$  is that fraction of  $B_2$  lying beneath  $S_1$ , that is, of width  $RE_1$ . Hence  $B_2' = RB_2 = RS_2(1 - p) / p$ . Let the intensities of the two sources be given by  $S_2 = n S_1 / R$ . The result follows immediately for the case  $n = 1$ .

source is sharply localized. One must conclude, therefore, that for the optimum in efficiency under diversified contamination it is best to be equipped with a generous set of crystals of various dimensions.

### THE SHIELD

A simple calculation of the solid angle subtended by the human body at the crystal (in units of four pi) at realistic distances suggests immediately that, if the signal and background are to be about equal, the specific activity of the shield should be from 4 to 25% of that of the human body. Since the latter is essentially  $10^{-13}$ c per gram of body weight, whereas that of ordinary building materials is of the order of  $10^{-12}$ c ( $\text{Ra}^{226}$ ), it is obvious that the activity of the surroundings must be reduced considerably. Of the many

CONSTRUCTION DETAILS OF STEEL ROOM

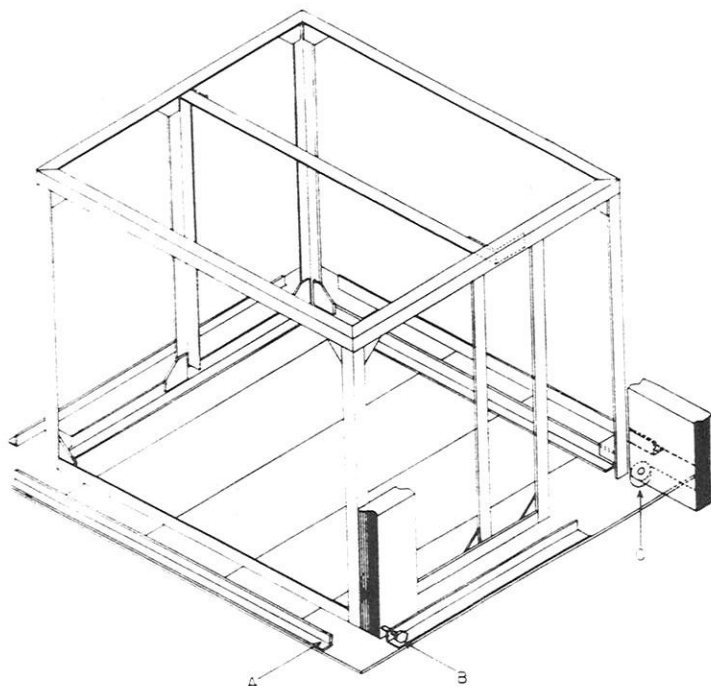


Figure 4. Construction details of steel room. (A) Angle iron used to clamp walls to frame with (B) log bolts. (C) Lower roller bearings which support steel door.

materials mentioned in the literature, we have selected steel because it is relatively clean, inexpensive and suited to economical construction. The generous dimensions of 2.4 by 2.1 by 1.8 M. of our setup were chosen for versatility in experimentation and relative freedom from serious back scatter problems.<sup>4</sup> This structure, as shown schematically in Figure 4, is assembled with iron plates 0.63 cm. thick and of weight not exceeding 72 Kg. each which are staggered and supported by angle irons throughout to overlap the joints between them; despite its large mass it can be dismantled and moved manually. In order to arrive at some definite conclusions as to the optimum in thickness of an iron shield, we had this structure gradually built up to various uniform thicknesses, and scintillation spectra were obtained with the use of our large 8 by 4 in. NaI (Tl) crystal. The results, drawn in Figure 5, show that

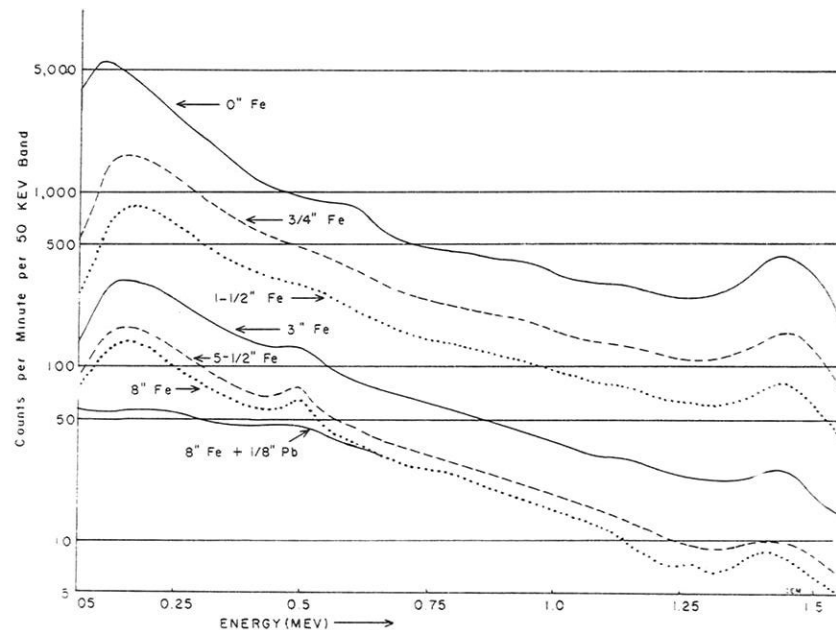


Figure 5. Reduction in gamma ray background of an 8 by 4 in. NaI (Tl) crystal shielded by various thicknesses of iron and lead. Internal dimensions of shield 2.4 by 2.2 by 1.9 M.

<sup>4</sup> Experience has demonstrated that it is wise to check experimentally in a small cave each lot of material which will be part of the shield and any accessory of the experimental setup to be housed in it.

the general shape of the spectrum obtained without the shield remains essentially unchanged but that the counting rate is reduced by a factor of nearly fifty with the use of 8 in. of iron alone.

These curves by themselves do not disclose the sources of the remaining background. Experiments with Pb, Bi and triply distilled Hg have suggested the existence of some radioactivity in the shield without furnishing, however, an accurate index of its absolute magnitude. Empirically, it has been found (5) that with the addition of only 0.32 cm. Pb, a tangible decrease in background counting rate is achieved below 600 kev; the effect can be clearly appreciated from the same figure where a decrease of counting rate by a factor of about two is gained in the region of 100 kev. This spectral modification is consistent with the hypothesis that some of the background is due to the inherent activity of the iron.

Recent theoretical developments (6) have established that in an extended homogeneous medium containing a uniformly distributed source of gamma ray of a given energy it is possible to calcu-

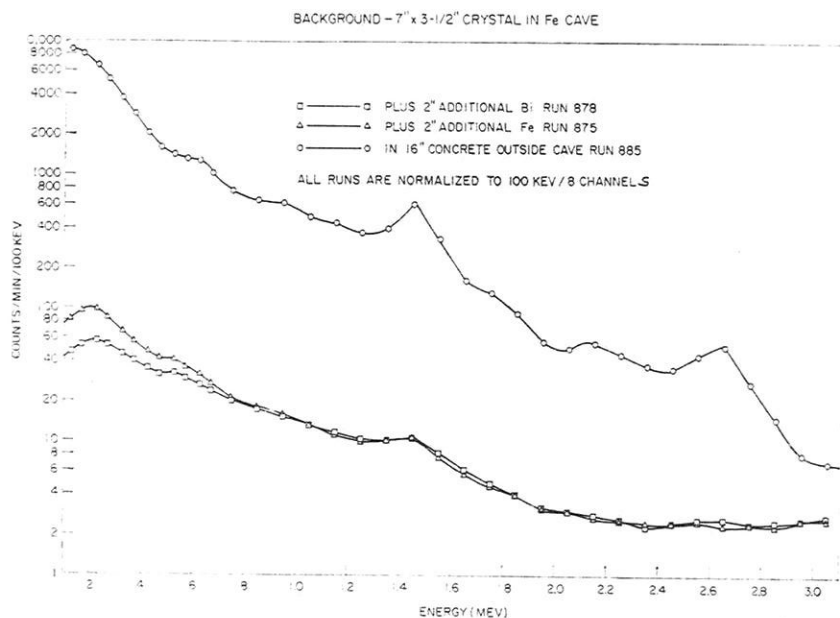


Figure 6. Background of 7 by 3.5 in. crystal in thick shields of concrete (o), iron ( $\Delta$ ) and bismuth ( $\square$ ).

late rather accurately a relationship between the number of gamma rays emitted per gram of material and the spectral gamma ray flux, namely, the number and energy of gamma rays crossing a spherical surface of one cm.<sup>2</sup> in cross-sectional area.

In many instances, it is possible with these parameters to infer from pulse-height spectra, such as those shown in Figure 6, both the nature and magnitude of the radioactivity of matter in bulk, namely, concrete, iron and bismuth. This in turn makes it possible to calculate rather closely the shield contribution to the background. In the case of a very intrinsically clean NaI crystal of 7 by 3 1/3 in. dimension, we have estimated that the specific activity of the steel of our shield (around  $5 \times 10^{-15}$  c (Th + Ra) per gram) cannot possibly contribute more than 30% of the background of our setup. This is information of great practical importance inasmuch as it indicates that there is no point in using shield materials with much lower specific activity without reducing drastically the other sources of background. A more detailed treatment of this subject is to be found elsewhere (7).

## COSMIC RAYS

A source of background comparable in magnitude to the residual activity of the iron shield is due to the presence of cosmic ray radiation composed mainly of photons and ionizing particles (mesons and electrons) of very high energy which interact directly with the crystal and its surroundings.

With NaI crystals of large size, pulses due to high energy mesons are easily recognized from the lower energy pulses (3.0 Mev) due to natural radioactivity because, on the average, a  $\mu$  meson will dissipate more than 4.3 Mev per centimeter of path in the crystal (8), and most mesons will travel through it for considerably longer distances. It is a simple matter, therefore, to discard them *in toto* by pulse-height analysis.

To eliminate counts due to the smaller losses suffered by the ionizing cosmic ray component in the crystal, it is customary to employ anti-coincidence electronic techniques which prevent recording of pulses if the ionizing particles traverse large trays of G.M. counters or scintillator detectors surrounding the crystal. This ar-

angement, however, does not prevent some of the non-ionizing components from interacting with the crystal without activating the overlying detectors.<sup>5</sup> In order to assess the total effect of cosmic radiation it is necessary to shield the apparatus with very great thicknesses of material capable of attenuating  $\mu$  mesons effectively. To properly achieve this, a comparison was made between the background spectra taken at our laboratory and at a tunnel situated 163 M. water equivalent below ground, namely, at a depth sufficient to reduce the cosmic ray ionizing component by a factor of about seventy (11). Readings were taken within shields of 20 cm. Fe and 20 cm. Fe + 5 cm. Bi, and special precautions were

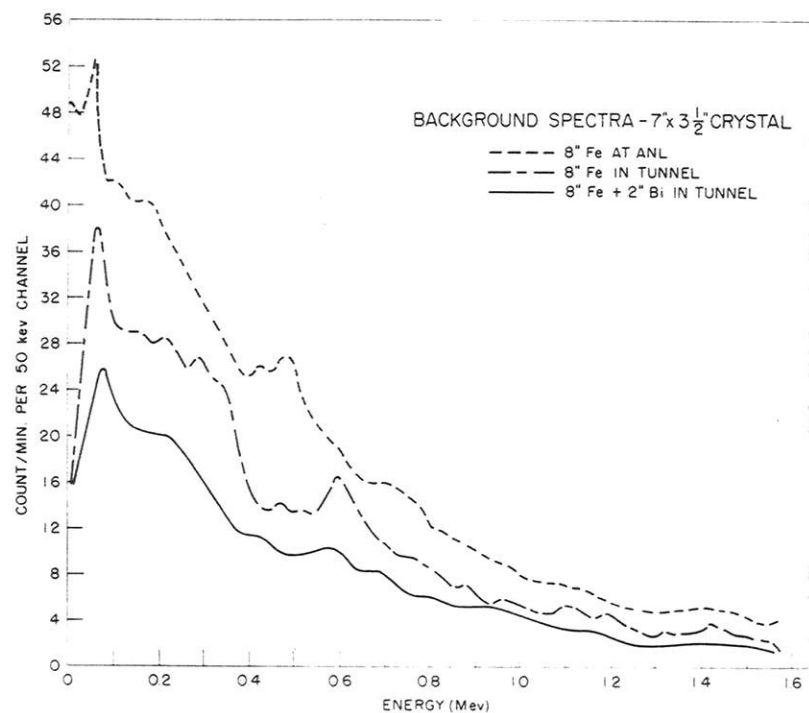


Figure 7. Background in 7 by 3 1/2 in. crystal under various shielding conditions at ANL and a depth of 163 M. water equivalent.

<sup>5</sup> Surrounding the detector with four pi scintillators connected in anti-coincidence has been reported in some low level measurements concerning double beta decay (9, 10). Unfortunately, the suppressive efficiency of this arrangement has never been reported.

taken to keep spurious surface contamination from influencing the results. This experiment showed that the cosmic ray background, as obtained by the difference between the counting rates above and below ground, was of the order of 25% of the background above ground and was not influenced unduly by the shield.

Typical background spectra obtained at 500 ft. above sea level and in a tunnel at a depth of 163 M. water equivalent are shown in Figure 7. This figure is instructive in that it shows, from the sharp decrease of the 511 keV band, that cosmic rays contribute significantly to the annihilating radiation present in the background.

### RADIOACTIVITY OF PHOTO MULTIPLIERS

The remaining source of intense background is the radioactivity of photo multipliers whose contribution is comparable to those mentioned above. Spectral analysis suggests that unless some definite effort is made to manufacture glasses of low potassium content, no improvement in the situation will occur; it seems that either quartz or vycor are eminently suited to the task from the optical and radioactive standpoint. Perhaps a more radical approach may be necessary, such as the use of metal tubes with vycor or quartz windows. An alternative to this would be the development of radioactively clean glass windows and of light pipes which can effectively attenuate the gamma ray emission of the tube. This choice would lead, however, to poorer resolution and would not eliminate the interference of radiation originating in the tube and scattered by test objects closely placed to the crystal; nor would it preclude Cherenkov radiation in the light pipe from being registered under certain conditions.

### CALIBRATION

In order to yield the exact disintegration rates of a radioelement in the body the gamma ray spectrometer, like any other gamma ray detector, should be calibrated with a known quantity of the same element identically distributed in a similar milieu. This goal is attainable only if to the isotope *in vivo* a known amount of the



same can be added and similar distribution can be achieved. Under these conditions the estimate is obtained by straight comparisons of the net counting rates before and after the known amount is given. In practice this exact method is applicable only to those radioelements which have a short effective half life in the body. For the determination of natural potassium, this would be the case if potassium suitably enriched with  $K^{40}$  were available; and it is virtually the case when  $K^{42}$  is used in its stead, provided that the comparison of the pulse spectra is restricted to energies greater than the  $Cs^{137}$  photo peak.<sup>6</sup> Administration of  $K^{42}$  has been used for the determination of total body potassium in several laboratories making use of various types of gamma ray monitors (12), and in our laboratory it has been applied successfully to various subject-to-crystal geometrical arrangements (13, 14).

An alternative to this method (4) is the use of the "wet" phantom which may be applied with confidence to radioelements generally distributed in the body; satisfactory agreement between the various approaches has been found by actual measurement on a few normal individuals. A convenient method of much more general application, which is extensively used at ANL for total body determinations, is the classical meter-arc-technique of Evans (1), as modified later by us (15). A more recent technique developed at ANL (13, 14) is based on matching of pulse-height spectra *in vivo* by suitable arrangement of masonite phantom material. Critical appraisals of both of these methods have not been reported, but they are being undertaken by various laboratories (4, 16, 17).

Interested readers are referred to the original articles and reports.

### CONCLUSIONS

Thus far, the relatively high cost of gamma ray spectrometers (and of ultrasensitive detectors in general) has limited their use to large AEC establishments generally devoid of regular clinical facilities. The installation of such an apparatus in a department of a medical school is a most welcomed step forward, since the ad-

<sup>6</sup> This procedure automatically eliminates the uncertainty, pointed out by Spiers, of the difference in Bremsstrahlung radiation from the two potassium isotopes.

vantages of extending tracer methodology to very low levels are most restrictive. There is every reason to believe that, with sufficient dedication on the part of both physicians and physicists, much needed information will accrue and that many refinements in methods and techniques will follow which will prove rewarding also in other fields of investigation.

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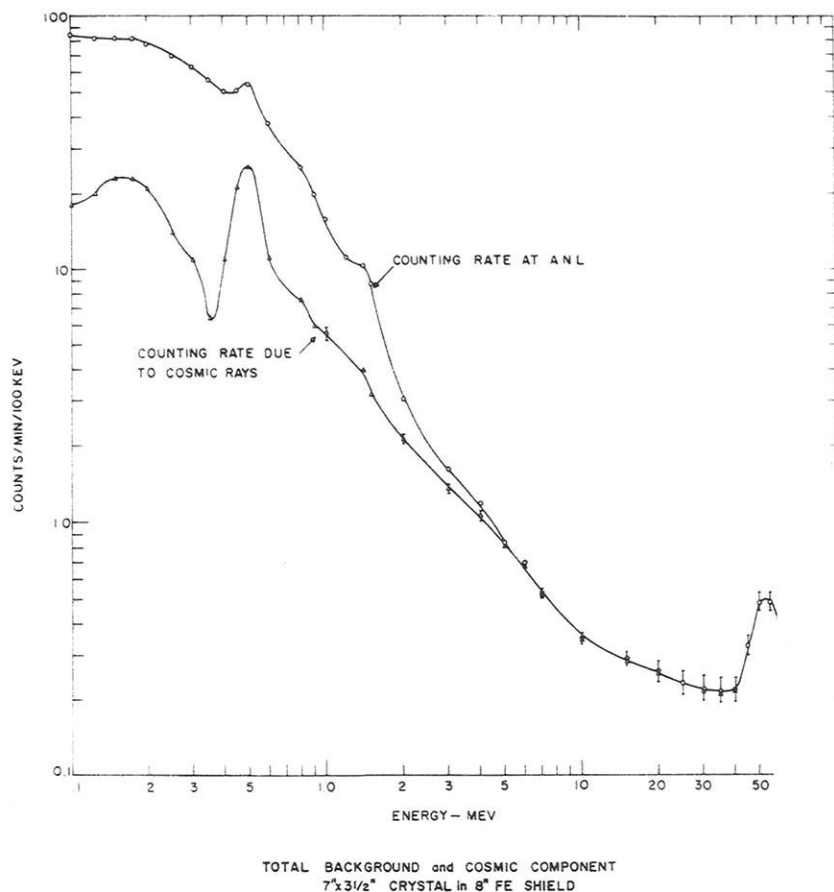


Figure 2. Total background at ANL and cosmic contribution obtained as the difference of two spectra on previous figure.

35% of the original, is of cosmic origin. Surrounding the crystal with extremely pure bismuth bricks removed another  $110 \pm 5$  counts per minute, as is shown in the third spectrum.

The background spectrum at the two locations, extended to 60 Mev, is shown in Figure 1, demonstrating quite clearly that above about 2 Mev the cosmic ray component rapidly becomes the major source of background. The peaks in the underground spectrum, at about 3.3, 4.0 and 5.5 Mev, will be explained later. In Figure 2 the difference between these curves is plotted. These figures show that elimination of the cosmic ray background alone

will be most effective in a relative sense in an energy region which is not of much interest to us. The possible exceptions are in evaluation of the 2.62 Mev thorium line, or in use of summing-coincidence methods (1) for the special case of cascade transitions, such as the 1.17 and 1.33 lines of  $\text{Co}^{60}$  or the 609-770 kev transitions of  $\text{Po}^{214}$  (RaC').

The variation of meson intensity with depth below ground is well-known and may be used to calculate the effectiveness of any underground location or other massive shield. The data of Wilson (2), with which our tunnel experiment was in good agreement, show the following:

Reduction Factor	Depth, meters water equivalent
0.2	15
0.1	40
0.05	80
0.02	160

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Underground locations are seldom available or conveniently investigated the characteristics of large plastic detectors, similar to those employed by the Rossi cosmic shower experiments (3). A disc 24 in. in diameter and 1/2 in. thick was employed (4), viewed by six type Figure 3 shows the efficiency of charged particle discriminator bias, where the efficiency  $\xi$  is defined as the ratio of n-fold coincidence counts to n-fold GM counter background and  $\text{Co}^{60}$  calibration response is shown.

The cosmic ray component varies with angle of incidence as  $e^{-2\theta}$ , where  $\theta$  is measured relative to the zenith. In this experiment the detector located 40 cm. above the 7 by 3 1/2 in. crystal, giving an average solid angle of 1.3 steradians, 50% of the cosmic counts should have been in coincidence. The actually observed reduction in counting rate in the energy range to 1 Mev was about 30% of the known cosmic component, and became less at higher energies. This can be interpreted to mean that the flux of photons and high-energy electrons arising locally in secondary cascade processes is more nearly isotropic. Thus the need for four pi geometry is implied, if 100% removal of the cosmic component

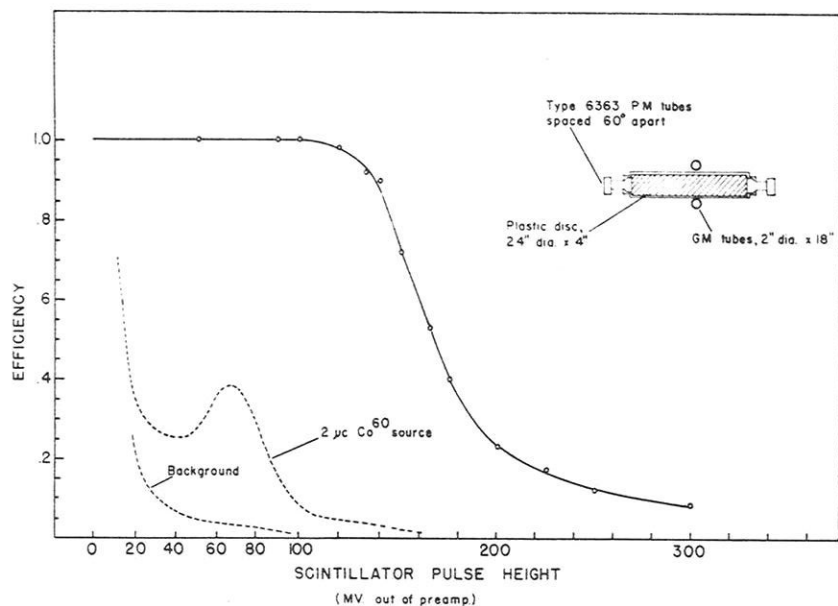


Figure 3. Efficiency of charged particle detection for large plastic detector. Dotted curves are to the same, but arbitrary, ordinate.

is to be achieved. Experiments employing such geometry have been reported recently (5) but their efficiency in eliminating the cosmic ray component has not been sought.

### Shield

We now consider briefly the background contribution from the iron shield. A net difference spectrum, representing the contribution of the iron shield less any radiation penetrating or originating within 2 in. of bismuth, is shown in Figure 4. The broad maximum about 350 keV, together with the more or less well-defined peaks at 600 keV, 770 keV, and 1.1 and 1.2 MeV, is indicative of radium, whereas the peak at 2.62 MeV denotes the presence of thorium. Since no extensive surface decontamination of the  $\frac{1}{4}$  in. steel plates was done prior to assembly of the cave, the possibility of surface activity cannot be completely ruled out. There has been speculation as to whether the solid armor plate used in construction of some iron rooms might not be significantly freer of contamination, but to date we have no experimental evidence bearing on the question.

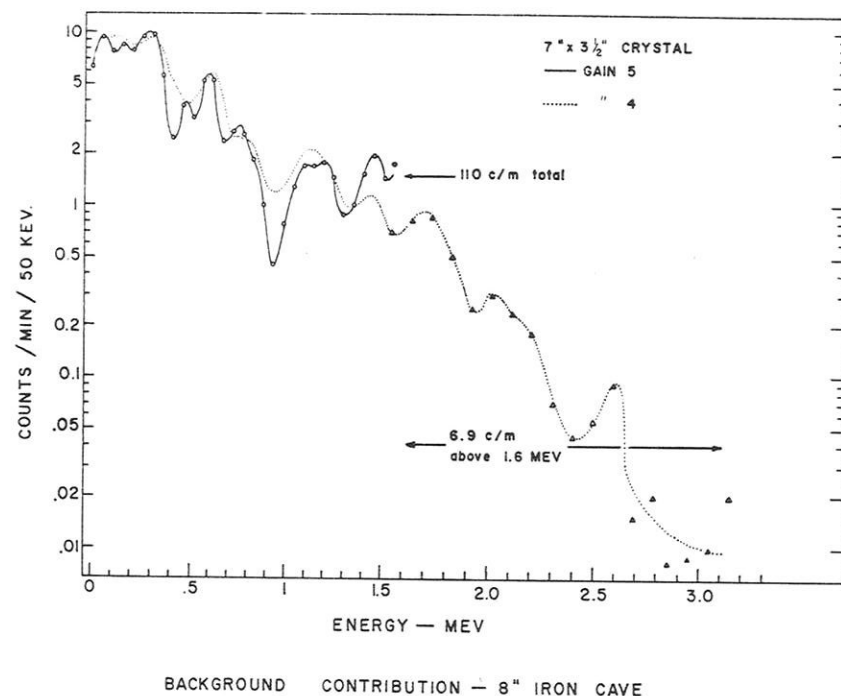
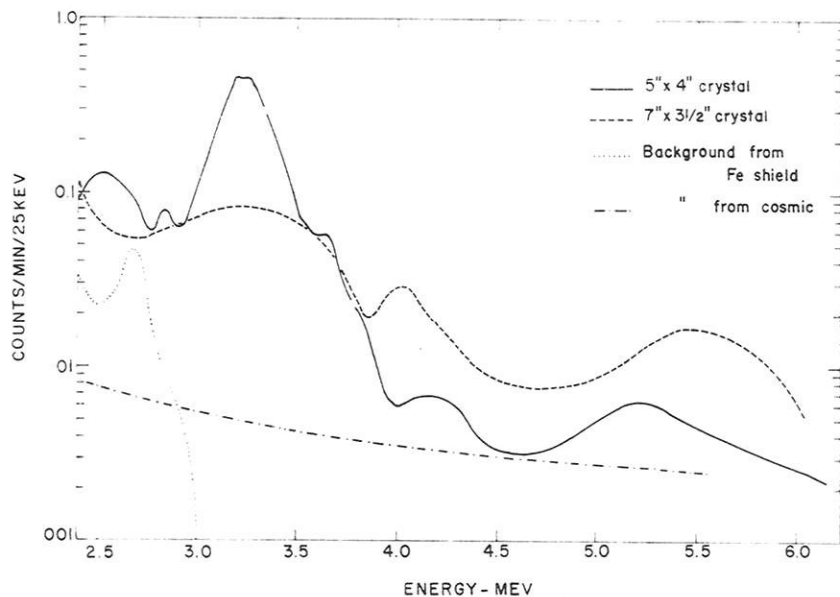


Figure 4. Portion of background due to contamination of shielding material, obtained by differences with very high purity bismuth shielding.

### Crystal

Many people have noticed peaks in their background spectra at energies of 3.2 and 5.5 MeV—the energies being only approximate. These peaks are particularly noticeable in the tunnel runs. Figure 5 shows the relative prominence of these peaks in a 5 by 4 in. and 7 by 3 $\frac{1}{2}$  in. crystal. The estimated contributions from remaining cosmic rays and from the thorium 2.62 MeV line, in the Fe cave, are shown for comparison. We feel quite sure that these peaks are from alpha particles, with energies ranging from 5.4 to 8.7 MeV, emitted by radium and thorium within the crystal. Due to the lower scintillation efficiency of NaI for heavy charged particles (6), these would appear in the observed energy region. In particular, the 5.5 MeV peak is believed to result from the beta decay branch of  ${}_{83}\text{Bi}^{212}$  (Thorium C), followed by the alpha-emitting  ${}_{84}\text{Po}^{212}$  (Thorium C') with an average lifetime of 0.43 microseconds. This is because the Bi $^{212}$  beta spectrum has a most prob-

able energy of 750 keV; the alpha energy of 8.78 MeV is equivalent in light output to a 4.85 MeV electron, and the phototube anode time constant of 2 to 3 microseconds effectively sums these light pulses. The total counting rate between 4.8 and 6.0 MeV, in excess of the assumed cosmic continuum, is only 0.5 counts per minute in the large crystal or approximately  $4 \text{ by } 10^{-13} \text{c}$ . This can give at most



COMPARISON OF ALPHA PEAKS IN TWO LARGE CRYSTALS

Figure 5. Spectra of thorium and radium alpha particles; internal contamination in crystal.

about 1 count per minute below 1.5 MeV, and hence is a completely negligible source of background in the region of interest.

### Phototubes

Radioactivity in the photo multiplier tubes was first recognized by Audric and Long in 1953 (7); additional data and spectra were published by Miller, Marinelli, Rowland and Rose (8). Larger crystals and better analyzers now permit improved accuracy. Table I summarizes many of our accumulated data on seven representative tube types. The probable error in estimates of total

activity is primarily due to calibration uncertainties, rather than counting statistics; the figures shown are believed accurate to  $\pm 20\%$ .

It is found that the use of multiple phototubes is required if the resolution capabilities of large crystals are to be fully exploited, but this usually results in an increased background. If one makes the rough approximation that the improvement in resolution is a function of total photo cathode area (that is, that photo cathode efficiency and electron collection are the same for all cathode surfaces), only then the background increase per unit area of useful photo cathode surface may be taken as a figure of merit in evaluating various tube types. This figure of merit is tabulated in the last column of Table I. Note that the values shown are not distributed at random but fall into four groups, with the 5 in. EMI quartz tubes being clearly the best. Thus, if the resolution achievable with a single 5 in. tube is satisfactory, one should use this tube for low level counting. If one needs the highest attainable resolution and is willing to accept the increased background, the quartz or vycor 3 in. tubes are indicated.

It has been shown, by crushing tubes and counting portions of glass from different regions of the envelope, that the type of glass used in the stem or "press" is consistently more radioactive than the sides or face plate. These Nonex glasses contain a few per cent of potassium, which is apparently necessary for their desired physical properties, as well as varying amounts of radium. Those interested in more detailed analysis are referred to a recent report (9).

## RESOLUTION OF BIG CRYSTALS

### Practical Data

Mr. Marinelli mentioned some considerations determining optimum crystal size. The choice would be relatively simple if the only *practical* limitations were price and availability. One must also consider the resolution capabilities of large crystals, particularly if the thickness is less than about 3 in. While much remains to be learned about the optical properties of large crystals, it is known that if satisfactory performance is to be obtained, every bit of crystal surface not coupled to a phototube should see a good

reflector. An improved method of canning has been developed by Harshaw, in which each phototube has its own individual window and all the area in between is covered with the usual alumina reflector. With a  $9\frac{1}{2}$  by 4 in. thick crystal so packaged, to which four matched DuMont 6363's were coupled, we obtained a  $\text{Cs}^{137}$  resolution of 9%. Figure 6 shows the  $\text{Co}^{60}$  spectrum obtained, with the source about 10 cm. from the crystal face. The ratio of 1.33 Mev peak to valley is 4.35.

In order to obtain such results, it is necessary to match tubes carefully: namely, to obtain matching of their gain versus voltage characteristics whenever changes in high voltage are to be used as fine gain control. Using the split-accumulate feature of most multi-

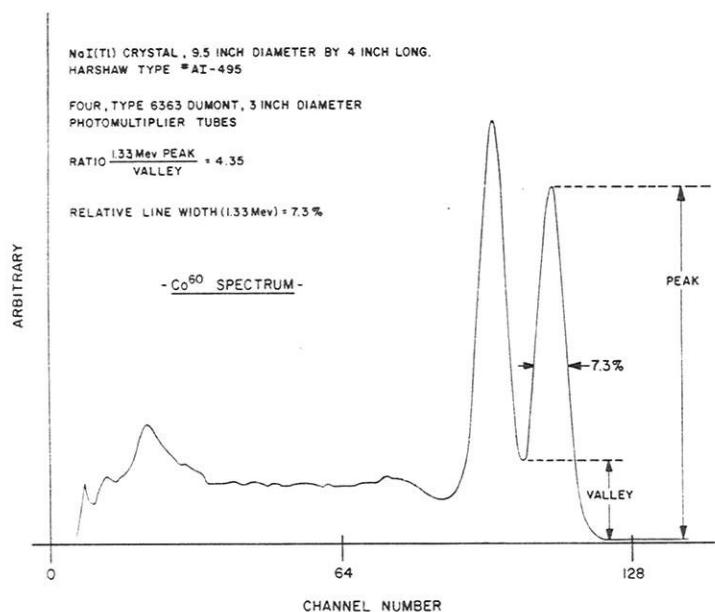


Figure 6. Resolution of large NaI crystal.

channel analyzers, the pulse-height distribution from each tube may be conveniently compared and matched. This distribution depends quite sensitively on the gamma flux distribution about the axis of symmetry, as illustrated in Figure 7. The distributions are those resulting from a single 3 in. tube, when the source was placed near the crystal face, at three locations along a diameter, as shown. Figure 8 shows the distribution obtained with the source on axis, averaged for the four tubes individually, and the result of adding

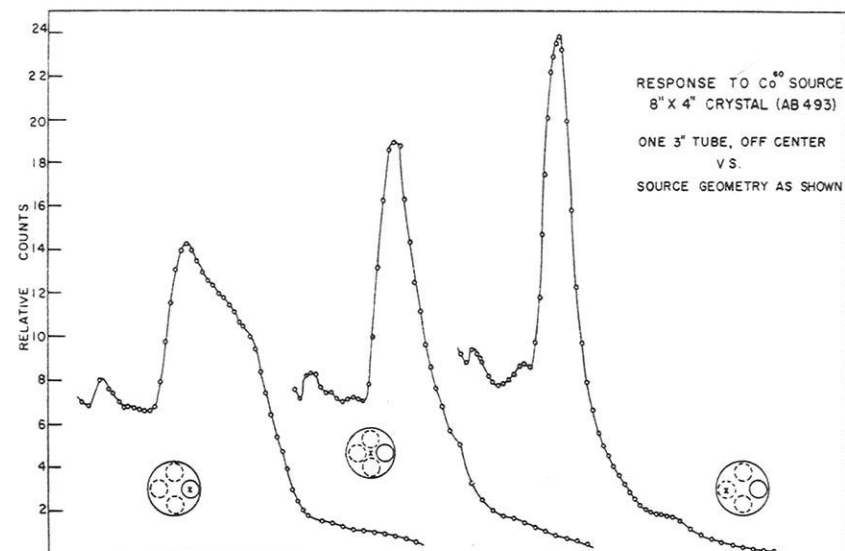


Figure 7. Variation of pulse-height distribution from a single 3 in. tube: source position moved along a diameter, as indicated. Equal counting times.

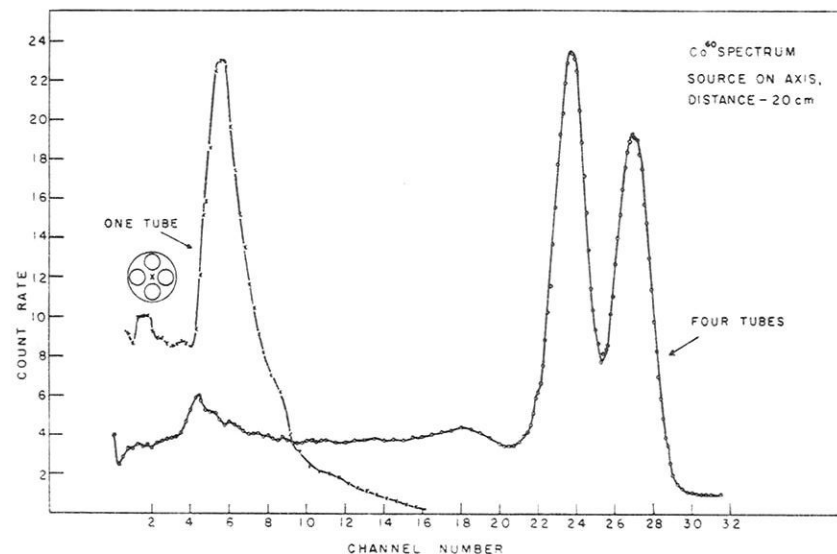


Figure 8. Average pulse-height distribution of single phototubes with source on axis, and the total four-tube spectrum obtained. Peak to valley ratio equals 2.5. (Same crystal as in Figure 7.)

them together in time coincidence. These figures indicate that some care should be used in source placement, or the results obtained may be misleading.

### Theory

The resolution at some specified energy—usually the 662 kev  $\text{Cs}^{137}$  line—is a convenient indication of the performance of a given system but does not tell enough. One would like to have also a figure of merit for the crystal package as a unit. It was shown by Bisi and Zappa (10) following the theoretical treatment by Breitenger (11) that resolution  $\eta$  and energy are related by the expression

$$\eta^2 = \alpha + \beta/E_0$$

The constant  $\alpha$  is determined by broadening of the line width due to statistical processes in the energy transfer from phosphor to

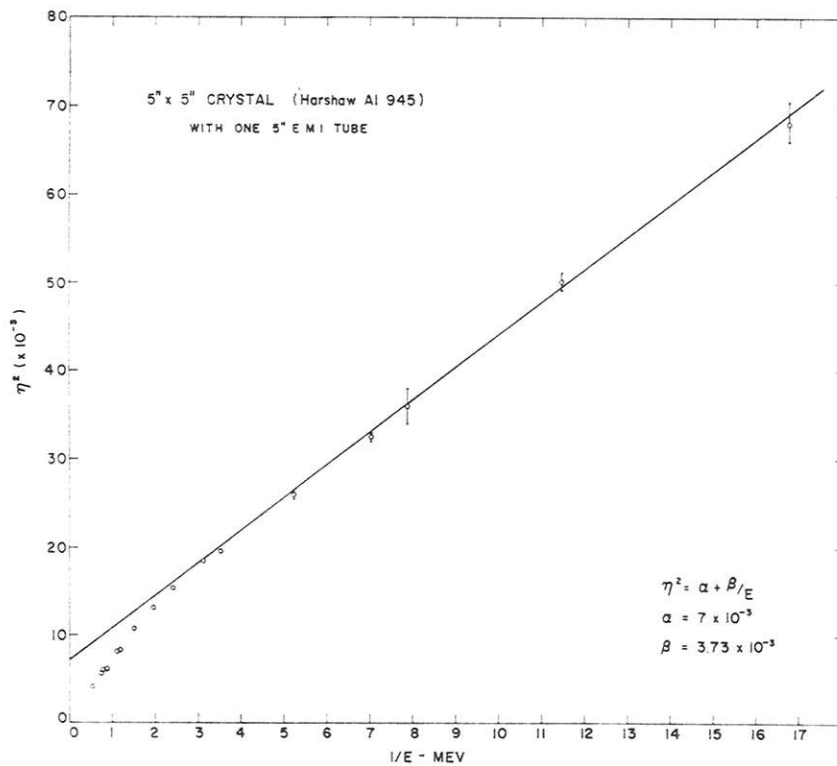


Figure 9

first dynode, whereas the constant  $\beta$  depends upon the statistics of the photo multiplier and the *average* transfer efficiency. Thus the intercept of  $\eta^2$  at infinite energy is more nearly representative of the crystal quality. Bisi and Zappa found that the experimental data from several 1½ by 1 in. crystals were linear for energies below about 500 kev, obtaining the average values:

$$\alpha = .0061 \text{ and } \beta = .0015$$

Data by Kelly of ORNL using a 3 by 3 in. crystal definitely did not fit a straight line. Mott and Sutton (12) discuss these experiments and conclude that there may be a portion of the transfer variance term which is energy-dependent. The experiments of Hickok and Draper (13) also suggest that this is the case, although their evidence does not appear conclusive. The data obtained at ANL with

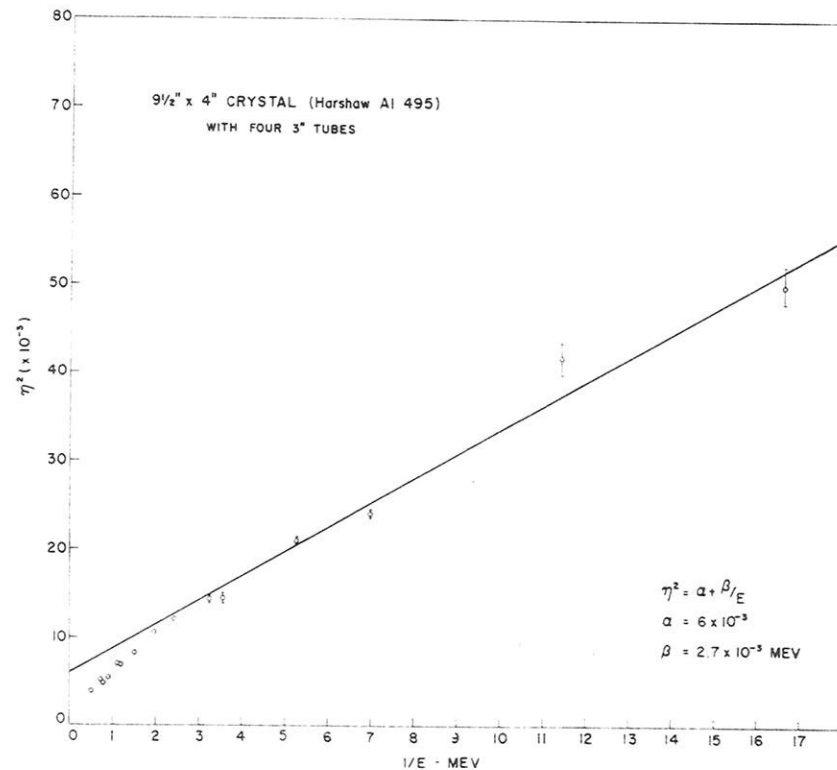


Figure 10

several larger crystals all seem to support the straight-line relationship. Figures 9 and 10 are presented as pertinent.

It is believed that such experimental studies, coupled with analysis of the crystal-phototube optics, will lead to further improvements in the performance of large crystals, justifying their large initial cost. The increased use of large NaI scintillators in routine medical gamma spectrometry is thus anticipated.

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TABLE 1

Phototubes

Mfg	Type	Nominal size	No. of tubes counted		Net counts/min (per tube)	Activity, * curies		Mass, gms (glass only per tube)	c/m/gm of glass
			Individually	Simultaneously		K <sup>40</sup>	Ra		
Part I - Face down									
DuMont	6363	3"		3 (face-plates only)	18				
DuMont	6363			3 (entire tubes)	39				
DuMont	K1785 (Vycor face)	3"	6		40.6			112	.35
				3	25				
				3	27				
			1 (face-plate only)		13.2				
			1		27.7				
			6		24.4				
			mean		26.0 ± 1.5	1.2 × 10 <sup>-9</sup>	5 × 10 <sup>-11</sup>	175	.15
EMI	9531B (Pyrex face)	3½"	2 (avg of 8 runs)		59			180	.33
EMI	9531B (Quartz face)	3½"		3	16				
					25 ± 5			205	.12
DuMont	6364	5"	4		110 ± 5			493	.215
			5		205				
EMI	9530B (Pyrex)	5"	3		184				
EMI	9530B (Quartz)	5"	2 (avg of 6 runs)		18.3 ± 3.2			400	.046
Part II - Stem down									
DuMont	K1785	3"	3		60	4.2 × 10 <sup>-10</sup>	3.8 × 10 <sup>-11</sup>		
EMI	9531B (Pyrex)	3½"	2		85				
EMI	9531B (Quartz)	3½"	4		59	1.0 × 10 <sup>-9</sup>	1.0 × 10 <sup>-11</sup>		
				3	70	1.25 × 10 <sup>-9</sup>	1.6 × 10 <sup>-11</sup>		
EMI	9530B (Pyrex)	5"	2		75	9.4 × 10 <sup>-10</sup>	1.9 × 10 <sup>-11</sup>		

\*Assuming all activity located in stem.