THE NEW LOS ALAMOS HUMAN COUNTER: HUMCO II*

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INTRODUCTION

A new well-type liquid scintillation gamma counter, large enough to provide 4π steradian counting geometry for the human body and for samples up to several hundreds of kilograms in weight, has recently been placed in operation at the Los Alamos Scientific Laboratory. Called Humco II (short for Human Counter), this counter is the sixth in a developmental sequence of large liquid scintillation counters constructed for biological purposes. Its predecessors are shown in Table 1.

The major objectives in the design of Humco II were increased energy resolution in terms of both peak widths and peak-to-valley ratios, increased useful energy range to include the efficient counting of bremsstrahlung from hard betas (especially Sr-90 and P-32), and electronic improvements including transistorization and automatic data recording and processing with punched cards.

*Work performed under the auspices of the U. S. Atomic Energy Commission.

TABLE 1. PREDECESSORS OF HUMCO II

Name	Volume (liters)	Scintillator Thickness (in.)	Composition	Sample Well Dimensions (in.)	Multiplier Phototubes (No.)	Multiplier Phototubes Dia. Size (in.)	References
"Herr Auge"	200	5	Terphenyl plus NPO in toluene	20 (dia.) x 30	45	2	(1)
K-9	75	4	PPO plus NPO in triethylbenzene	9 (dia.) x 28	8	2	(2)
Humco I	530	6	Terphenyl plus POPOP in toluene	18 (dia.) x 72	108	2	(3,4)
WRAIR Counter	600	6	Terphenyl plus POPOP in toluene	20 (dia.) x 72	30	5	(5)
Genco	280	6	Terphenyl plus POPOP in triethylbenzene	18 (dia.) x 72 (semi-cylinder, 2π)	6	16	(6,7)

DESCRIPTION

An artist's sketch of Humco II is shown in Fig. 1. The sample is introduced into the well by the usual trough and sling arrangement (3,4). Shielding is provided by 7-1/2 in. thick naval armorplate in the form of a room with inside dimensions 8 x 10 x 7 feet high. Use of room-type shielding provides easy access to the counter for installation and maintenance. The tank, which has a volume of 1600 liters, is 72 in. long and provides a minimum scintillator thickness of 12 in. around the 18 in. diameter well. Previous counters have not exceeded 6 in. in thickness. It was hoped that the extra thickness would increase the probability of multiple Compton scattering and would thereby enhance the Compton peak relative to the continuum at low energies.

Scintillation light is collected by 24 multiplier phototubes (DuMont No. K-1328) with nominal 14-1/2 in. diameter photocathodes. Thus, 24 per cent of the total wall area is photosensitive compared with 18 per cent in Genco. This fraction is a measure of primary light collection efficiency. Because of the comparatively poor reflectance of all reflectors when wet and the difficulty of obtaining long optical mean free paths (several meters) in the scintillator, rapid collection of the scintillation light is essential.

Phototubes are balanced individually through the use of a small test tank of 40 liter capacity (16 in. diameter x 12 in. deep). Balancing potentiometers in series with the dynode resistors are adjusted to give the same peak pulse height for each tube when tested with a K-40 source and a supply voltage of 1500.

During most of the testing period, the counter has been filled with a conventional scintillation solution consisting of 4 g/l terphenyl and 0.05 g/l POPOP in toluene. For routine operations, this will be replaced with a solution of low volatility described later.

Figure 2 is a photograph of the partially assembled tank showing the arrangement of the phototubes and the extent of photocathode coverage. The outer wall of the tank is hexagonal in cross section to provide flat surfaces on which to mount the tubes. No windows are used, the phototube being in direct contact with the liquid. Because of the curvature of the glass faceplate and the desirability of providing optical coupling to the scintillator, use of

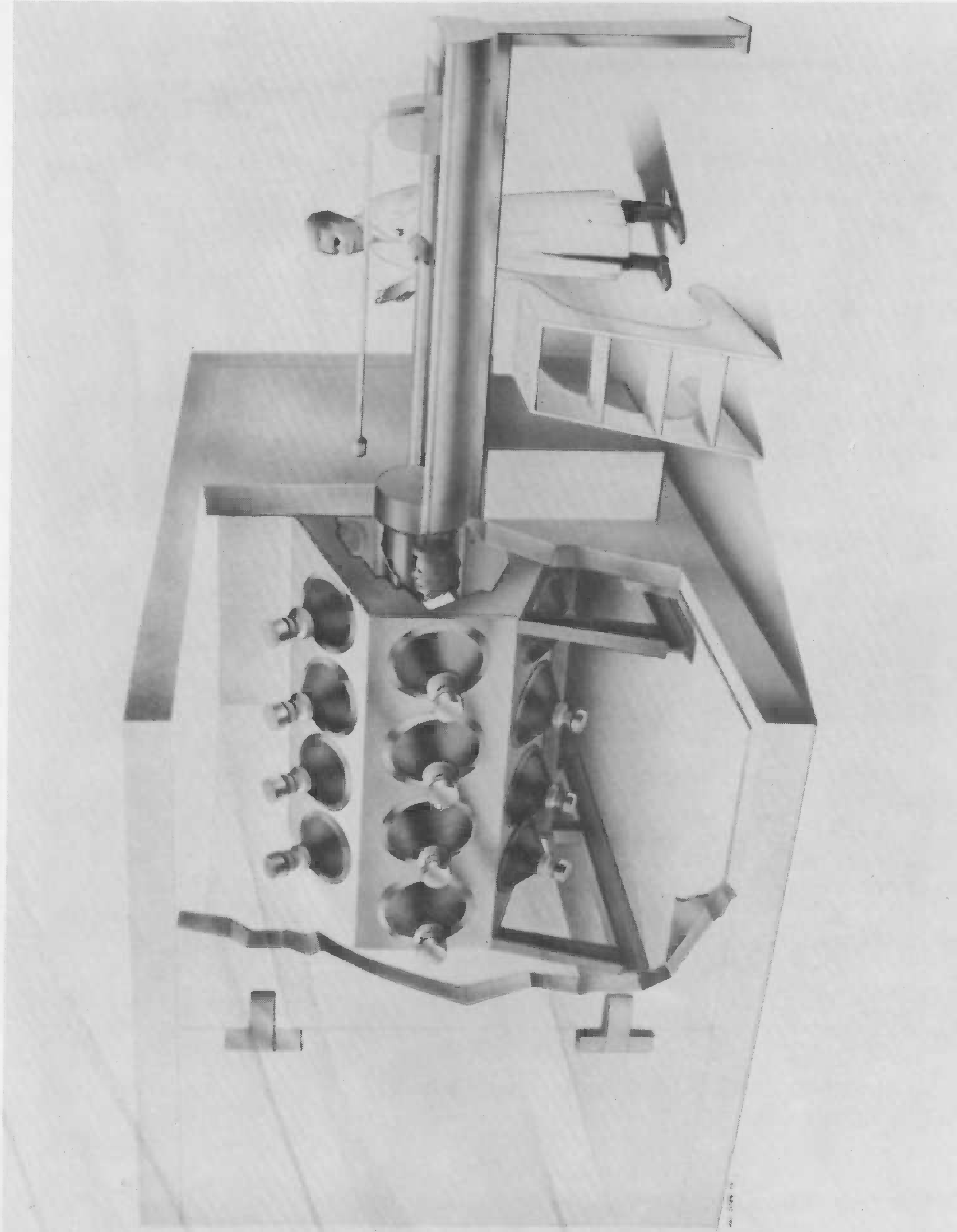

Fig. 1. Artist's sketch of Humco II.

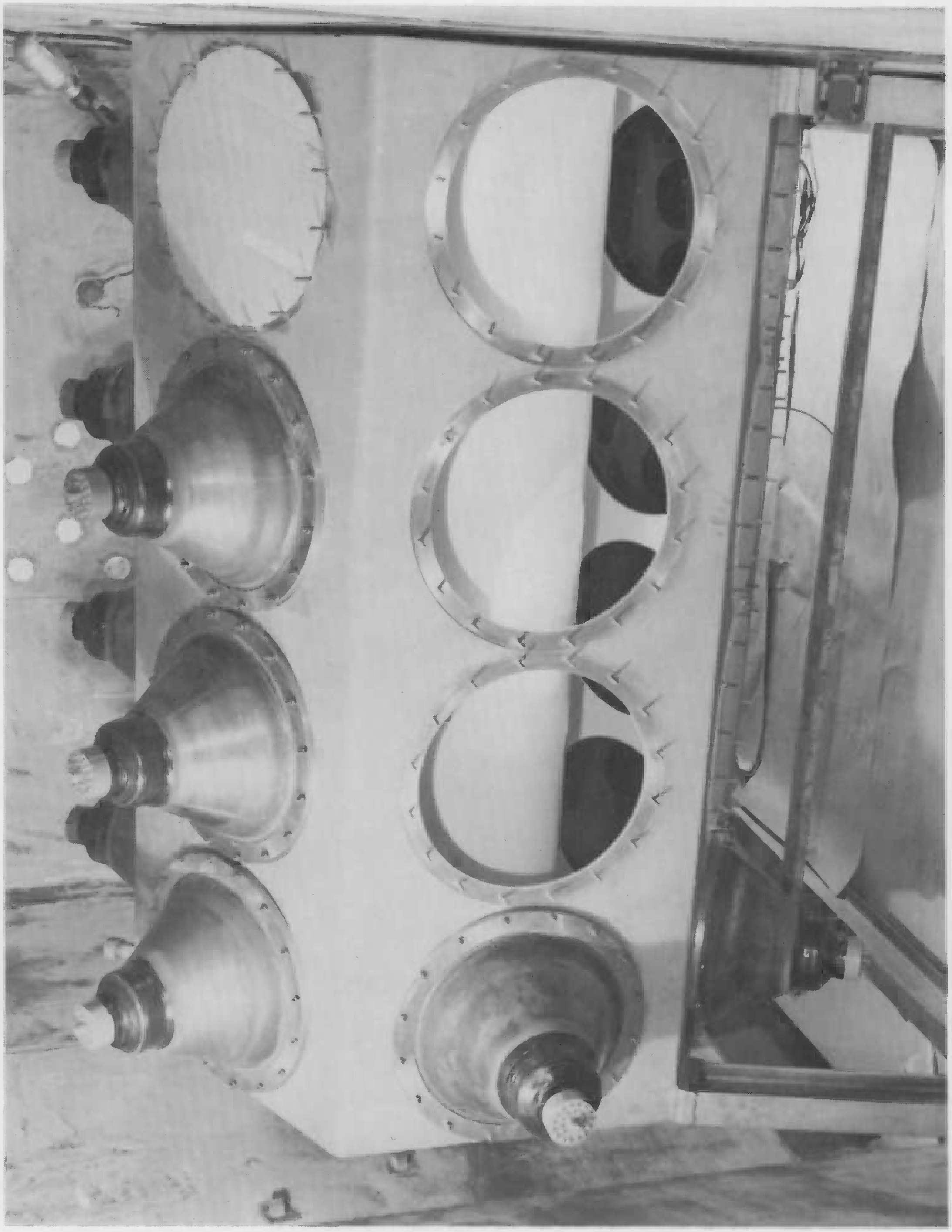

Fig. 2. Photograph of Humco II tank.

windows would be quite complicated. Failure of a photomultiplier is a sufficiently rare event that the necessity for draining the tank to replace tubes is not a serious drawback.

Liquid-tight seals between phototube and tank are provided by cementing the tube to a steel ring with an epoxide adhesive. A silicone-rubber O-ring makes the seal between the ring and flat machined surface on the tank. The ring is clamped by 16 studs welded to the tank; a torque wrench set at 16 in. pound ensures uniform tension. It is very important that the surface of the tank to which the O-ring seals are made should be flat to a tolerance of $\pm$ 0.005 in. Deviations larger than this will cause strains in the phototube faceplate, which may result in cracking of the glass-metal seal. A simple method of obtaining such a flat surface is to cement to the roughly machined tank a flat aluminum ring using Rezolin Epoxy Resin F.* Successful liquid-tight seals have also been made using special silicone-rubber gaskets between the ring and the phototube envelope. This method further reduces the possibility of straining the phototube, but is considerably more complicated than the epoxide cement method.

Figure 3 shows the transistorized electronics for the counter. The chassis in the relay rack, reading up from the bottom, are the pulse height analyzer consisting of 6 plug-in type single channel units, the amplifier, the punch control unit, a data input chassis which permits sample identification information to be entered, a preset timing unit, 3 double-scaler chassis, a digital master clock, and a slave clock which provides the time of day output. At the completion of each count, all data are entered on an 80 column decimal punched card by the IBM 526 punch. Auxiliary typewriter input permits additional information to be entered manually in the last 8 columns of the card. (Alphabetic information can be entered only in these columns.) An IBM 704 computer will perform all data processing, including averaging of replicate runs on a single sample, subtraction of appropriate backgrounds, calculation of calibration runs, and solution of simultaneous equations for the amounts of various selected nuclides (up to 6) present in the sample. A multichannel analyzer is used for energy calibration and for spectral analysis of unknown activities. As will be seen later, the ability of the liquid scintillator to resolve complex mixtures

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*Rezolin Inc., Santa Monica, California.

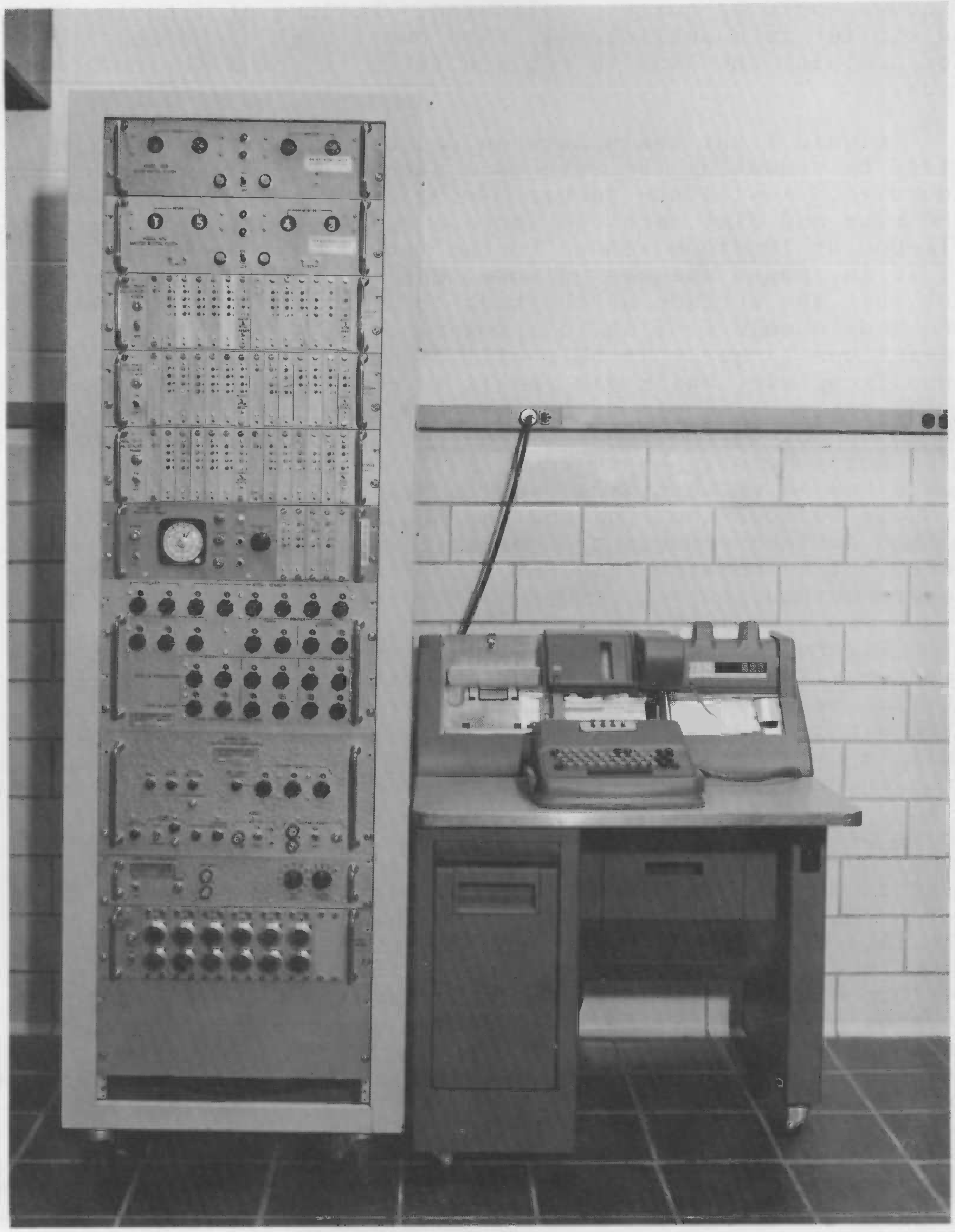

Fig. 3. Transistorized electronics for Humco II.

is limited; however, spectral identification is possible for the components of several simple mixtures which are commonly encountered.

ENERGY CALIBRATION

It is difficult to measure directly the energy actually deposited in a large organic scintillator. As a result of the low atomic number of the material, total absorption of gamma ray energy by the photoelectric process is not an important primary process. The principal interaction over the range of gamma energies of interest is by Compton scattering, in which any amount of energy can be transferred from the incident gamma ray to the recoil electron up to a maximum amount,

$$T_{max} = \frac{2\alpha_o}{1 + 2\alpha_o} E_\gamma,$$

where α_o is the gamma energy in units of m_oc^2, the characteristic rest mass energy of electrons. E_γ is also the initial gamma ray energy, in whatever units are desired for T_{max} (8).

A moderately sharp energy peak results from this process, but the actual peak energy is subject to change by multiple scattering, introducing a corresponding uncertainty in the calibration. Sources of alpha rays or conversion electrons cannot be used for calibration outside the tank, and internal sources of these activities would, if dissolved, contaminate a large and expensive volume of scintillator. If used as encapsulated local sources, it is necessary to move them throughout the tank to obtain a proper space average of the resultant pulse heights. An attempt will be made to perform the latter experiment both for calibration purposes and for measurement of variations in light collection efficiency over the scintillator volume.

It is, however, possible to determine the energy deposition and to estimate the relative importance of multiple Compton scatter. The method is based on the fact that a plot of maximum Compton energy against incident gamma energy is very nearly linear above a few tenths Mev, as indicated in Fig. 4. The intercept of the extrapolated line with the abscissa varies only slightly with the energy range used. Extrapolating the theoretical curve from the range 0.66 to 1.45 Mev gives an intercept of 170 kev. Using 0.66 to 3.0 Mev, the intercept rises to about 200 kev; using 0.4 to 1.56 Mev,

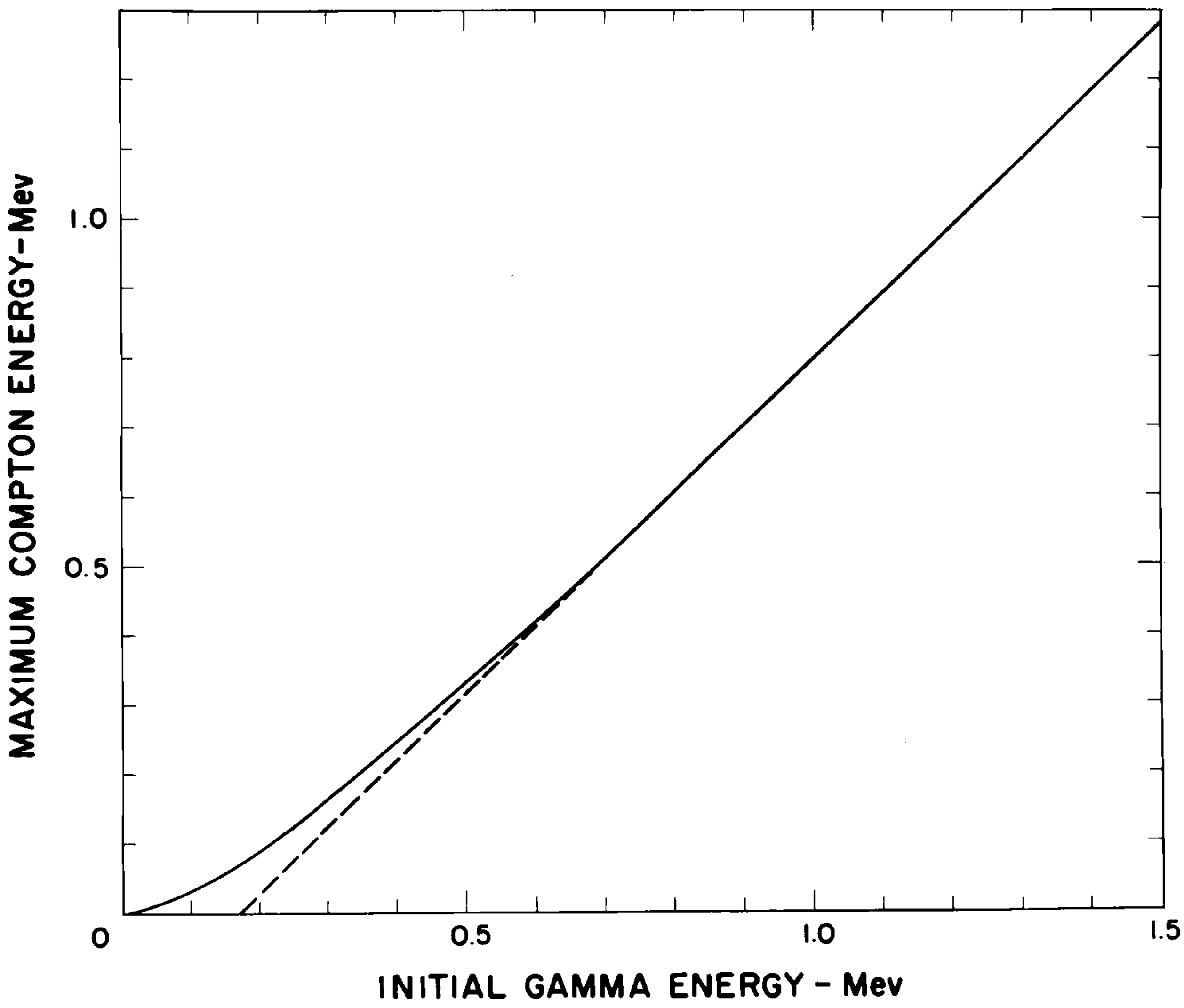

Fig. 4. Calculated maximum Compton energy from single scattering versus gamma energy.

it falls to 150 kev. For small detectors, the plot of observed pulse height of the Compton peak versus full gamma energy should show a similar intercept. This may be the case for the counter whose calibration is given in Fig. 5, a 40 liter test tank 16 in. in diameter by 12 in. deep, viewed with a single 16 in. multiplier phototube. The observed pulse heights from 6 gamma rays 0.51 to 1.46 Mev are plotted against full gamma energy in the lower curve. The intercept is 200 to 220 kev, which is 40 to 50 kev higher than the calculated value. The discrepancy may be real and may be due to an inherent nonlinearity in the scintillator response, possibly due to the deposition of some of the energy at high specific ionization near the end of recoil electron ranges. Anthracene crystals show a similar nonlinearity amounting to 22 to 25 kev (9,10).

If observed pulse heights are plotted against maximum Compton energy instead of against full gamma energy, one would expect a positive intercept of a few tens of kev if single Compton scatter predominates, and a negative intercept if there is much multiple scatter. The upper line of Fig. 5 shows that the former case is again indicated. The energy calibration of the 280 liter Geneva Counter is shown in Fig. 6. The scintillation tank, in this case, is hemicylindrical with a detector thickness of 6 in. so that multiple scatter is unlikely. Relative pulse height plotted against calculated maximum Compton energy yields a straight line with an intercept of 30 to 40 kev.

Figure 7 presents the similar calibration of the new counter, Humco II. Not only is total scintillator volume now much larger (1600 liters), but as a result of the 4π geometry the backscattered photons from a maximum Compton recoil now must traverse the scintillator on the opposite side of the well (the calibration source, of course, being in the sample well). The expectation of increased probability of multiple scatter is confirmed by the calibration curves. When pulse height is plotted against energy deposited by single maximum Compton collision, the line shows a negative energy intercept of 40 kev. When plotted against full gamma energy, the intercept is 120 kev. Plotting against the energy transferred by two successive 180° collisions gives the middle line, which shows the most reasonable intercept of 20 kev. A summary of the evidence regarding multiplicity of Compton scattering in detectors of various sizes is given in Table 2.

It is of interest to consider the apparent pulse heights

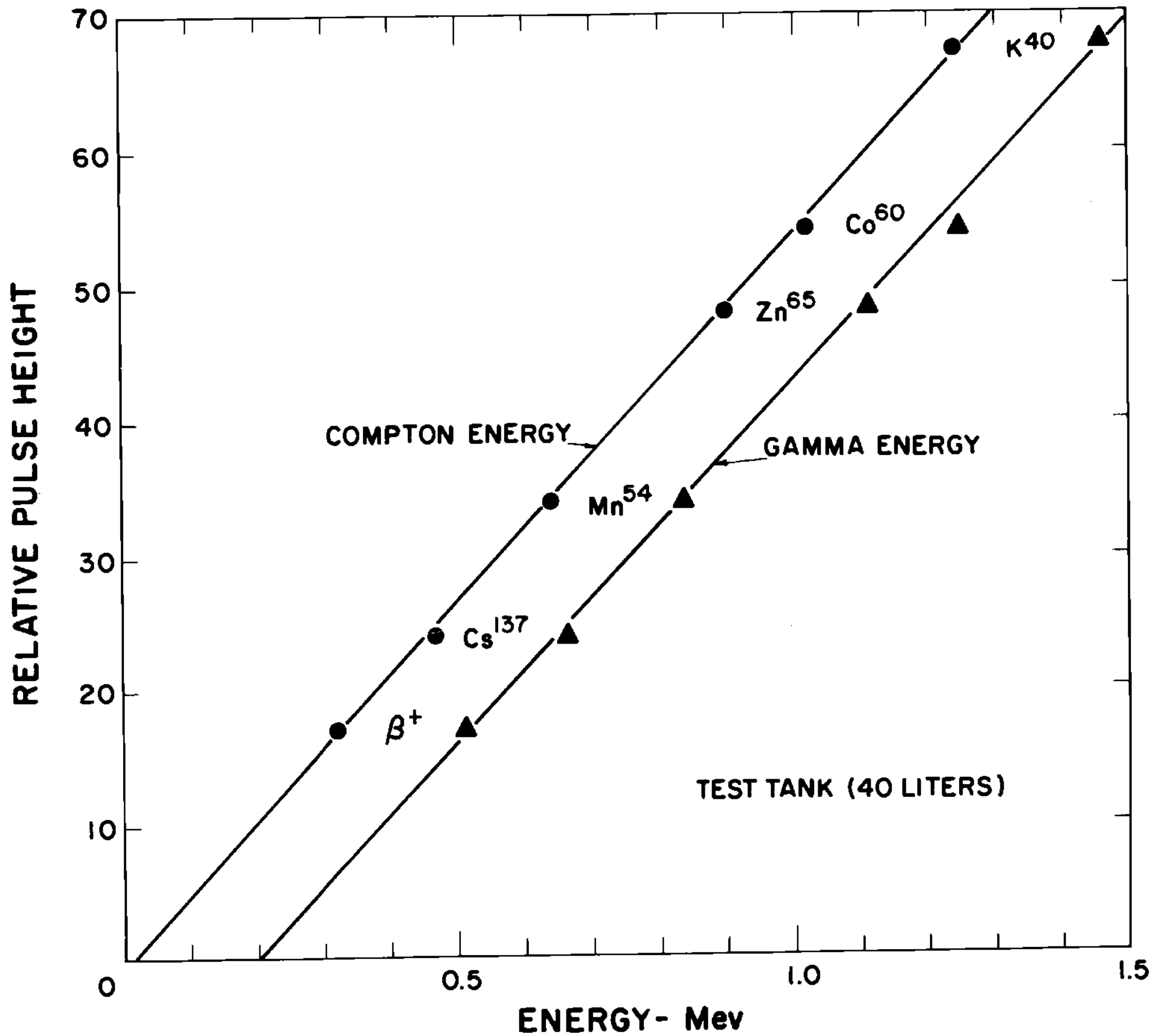

Fig. 5. Test tank energy calibration.

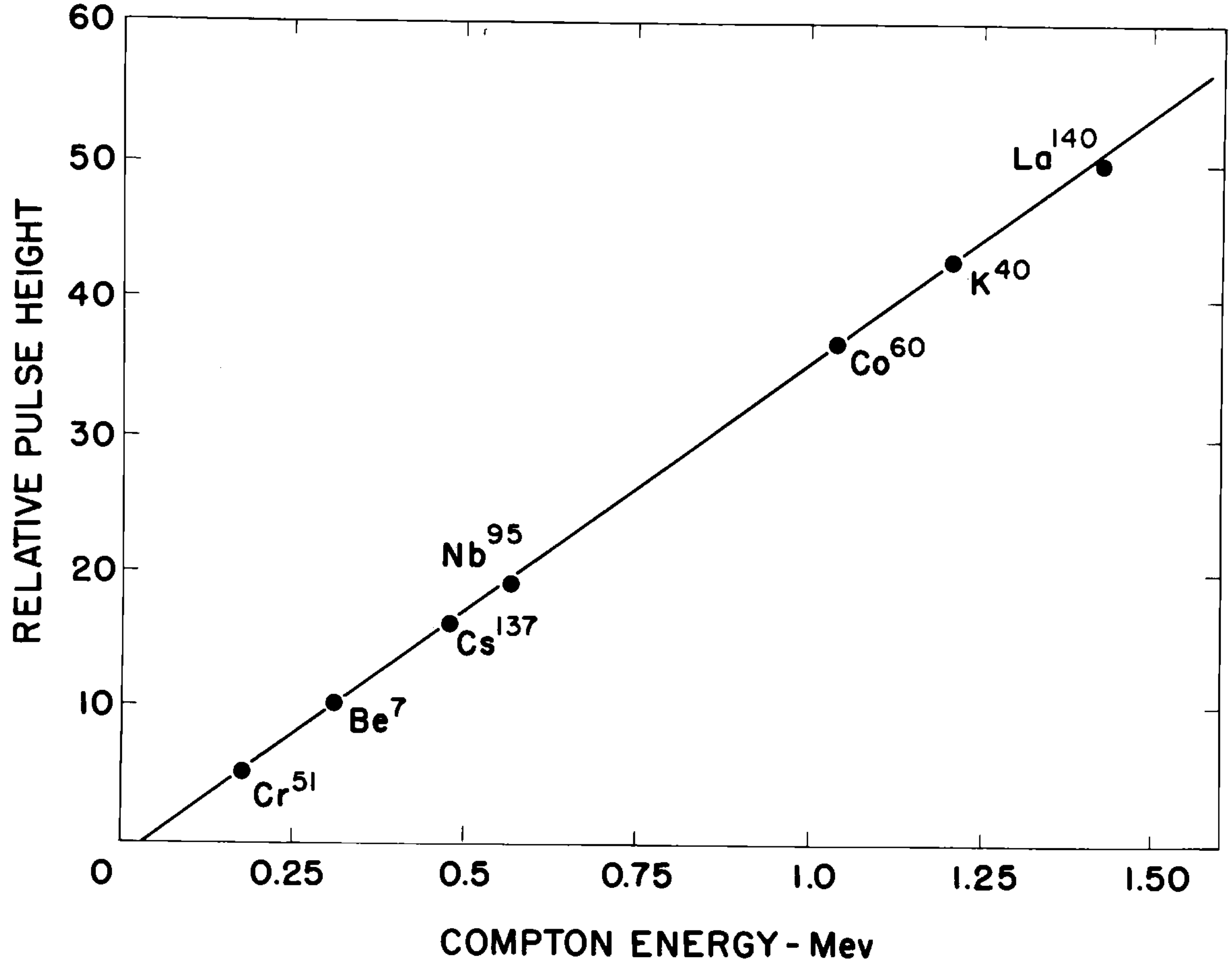

Fig. 6. Energy calibration of Genco.

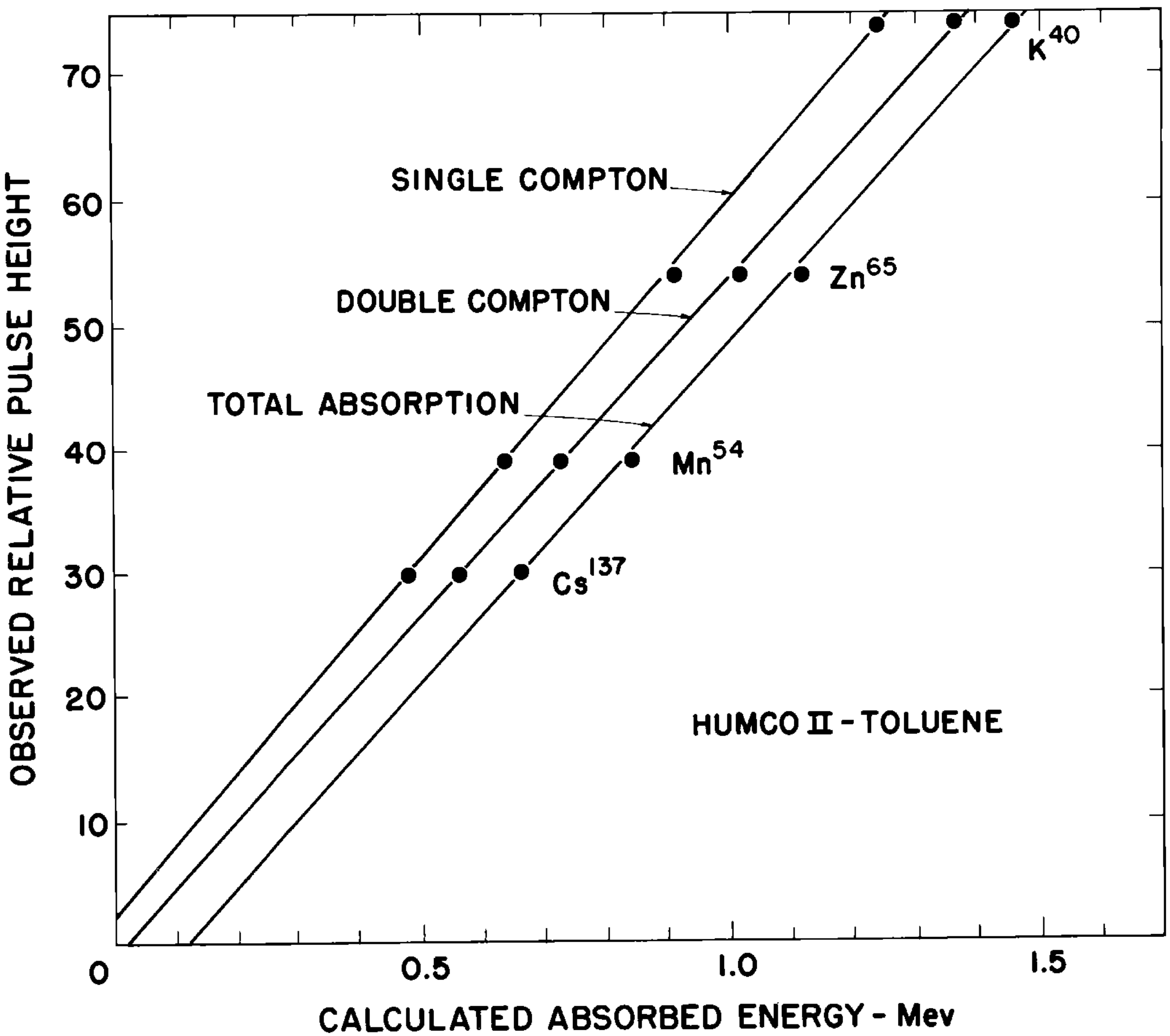

Fig. 7. Energy calibration of Humco II.

TABLE 2. MULTIPLICITY OF COMPTON SCATTERING IN DETECTORS OF VARIOUS SIZES

Detector	Shape	Volume (liters)	Scintillator Thickness (cm)	Zero Pulse Height Intercept (kev)	
				Single Compton E	Total Gamma E
Test Tank	right cylinder	40	30	+20	+200
Genco	2π well	300	15	+30	+170
Humco II	4π well	1600	30	-30	+80
Calculated for single Compton scattering				+24*	+185**

*Observed value for electrons on anthracene.

**Including 25 kev for inherent scintillator nonlinearity.

of compound events occurring when cascade gamma rays are absorbed. Figure 8 is an extended graph of the energy calibration of Humco II, including the peaks resulting from Co-60 and Th-228. The points are plotted against the energy absorption calculated on the assumption of double Compton scatter. The two Co-60 gammas (1.17 and 1.33 Mev) are not resolved but appear as a single peak of height 68. This is significantly off the curve to an extent suggesting that in addition to double scattering of one of the photons, some energy absorption from the other photon has also occurred. The Co-60 sum peak at pulse height 123 also lies off the curve if it is assumed that only one gamma ray suffers double scattering (point marked "either"). If double scattering of both gammas is assumed (point marked "both"), good agreement with the calibration line is obtained.

The case of Th-228 is more complicated. The coincidence involved is that between the 100 per cent 2.615 Mev gamma and the 88 per cent 0.58 Mev gamma of Tl-208, namely ThC'' (11). Only one peak is observed above 2 Mev, and it appears to correspond best with the energy transferred by double scattering of both gamma rays.

A Monte Carlo computation of the actual energy deposition in large organic scintillators is now under way by M. A. Van Dilla of the Los Alamos Scientific Laboratory using a program written by Zerby and Moran (12) of the Oak Ridge National Laboratory. These results will not only provide further evidence on energy absorption, but will also indicate the extent to which resolution is limited by the absorption process.

ENERGY RESOLUTION

Energy resolution obtained with Humco II using a conventional toluene-terphenyl-POPOP scintillator is shown in Fig. 9. The plot is the usual η^2 versus 1/E (13), except that because of the asymmetry of the Compton peaks, the half-resolution has been used instead of the full width. The energies used are the values calculated for double Compton scatter as in Fig. 8. The half-resolution varies from 17 per cent at K-40 (1.36 Mev) to 25 per cent at Cs-137 (0.56 Mev). The results would appear to be explicable on the basis of two resolution terms, one energy independent with a half-width of 10 per cent and the square of the other varying as the reciprocal of the energy and having the value of 14 per cent at 1.36 Mev. The factors limiting the

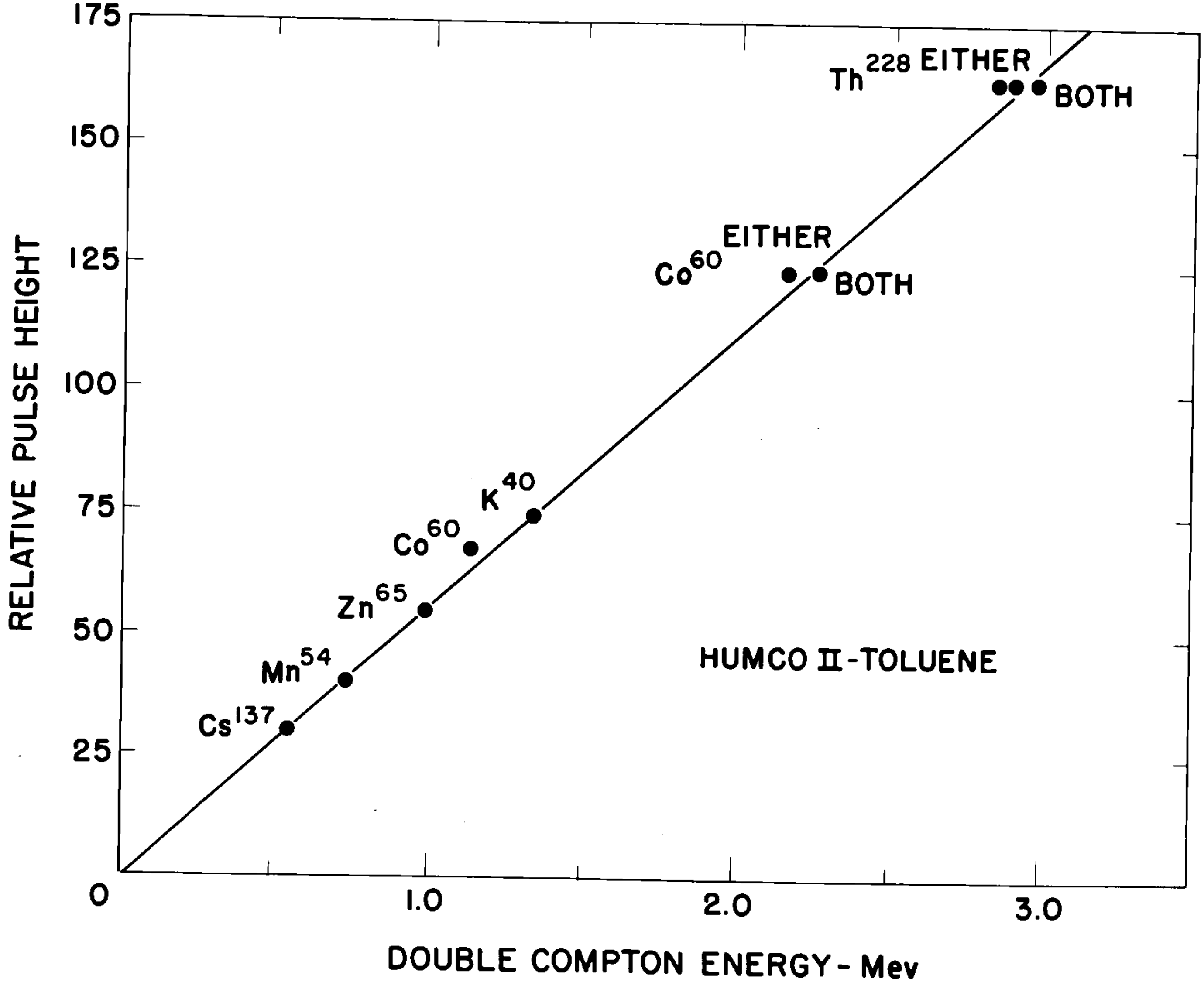

Fig. 8. Extended energy calibration of Humco II.

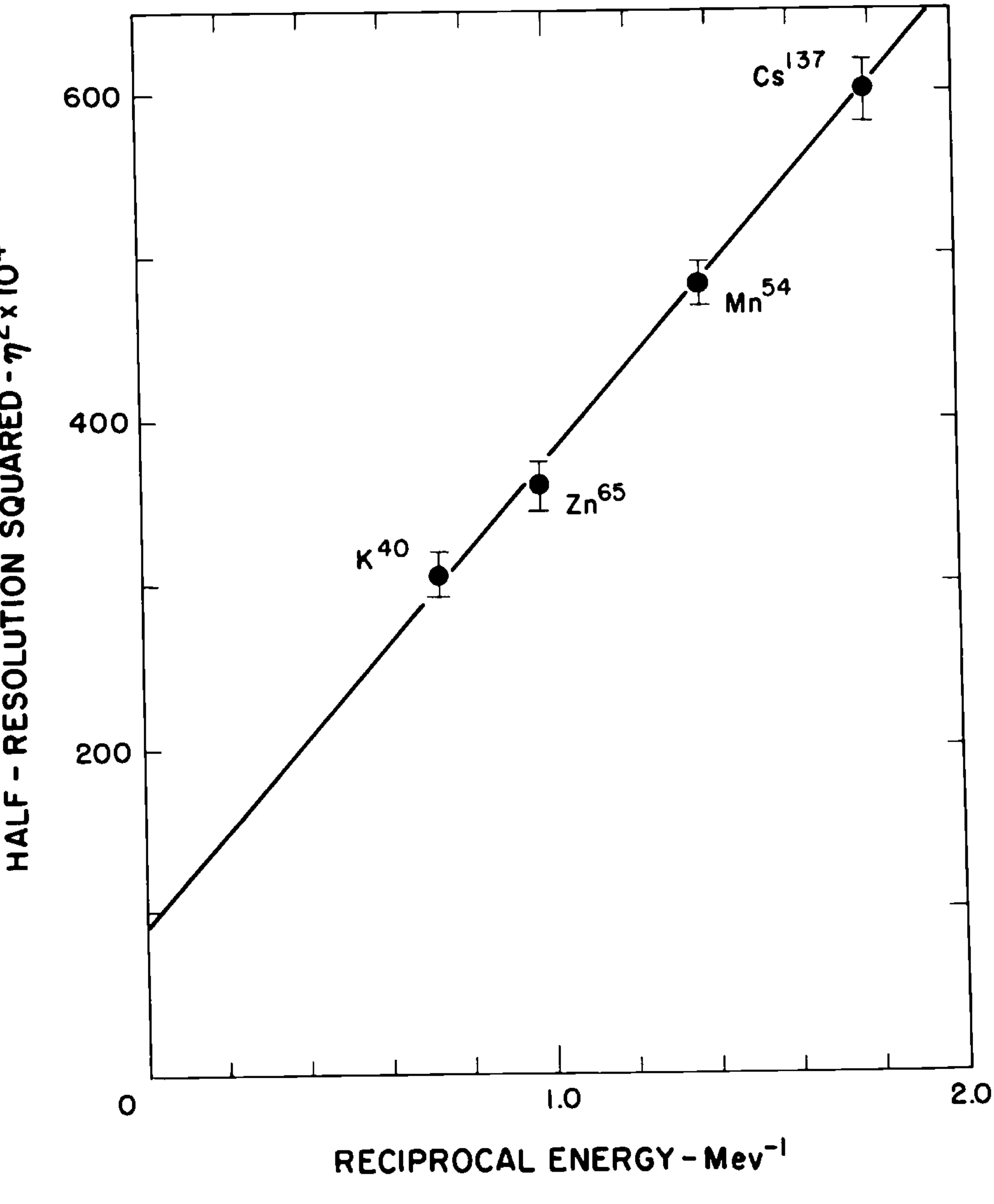

Fig. 9. Energy resolution of Humco II.

obtainable resolution which might explain Fig. 9 are as follows:

1. Inherent scintillator resolution, i.e., variations in the total scintillation light emitted for events of equal energy deposition.

2. Statistical fluctuations both in number of photons emitted by the scintillator and in the photoelectron emission and amplification factor of the multiplier phototube. Since the photocathode efficiency is of the order of 10 per cent, scintillator statistics will be negligible compared with phototube statistics.

3. Variations in the amount of energy deposited in the scintillator by gamma rays of the same energy. This includes escape of scattered photons or of bremsstrahlung, and in the case of organic scintillators the shape of the Compton recoil spectrum as modified by multiple scattering.

4. Variations in the efficiency with which light is collected from various locations in the scintillator.

5. Nonuniformity in the photosensitivity of the cathodes of the multiplier phototubes.

While it is not possible at present to decide quantitatively on the relative contributions of the above effects, some of the factors can be eliminated.

The inherent resolution of organic scintillators is not greatly inferior to that of NaI (Tl). Figure 10 shows the resolution obtained by exciting a small volume of liquid scintillator (terphenyl plus POPOP in toluene) with Cs-137 conversion electrons under conditions of optimum light collection efficiency and photocathode uniformity. The full width at half-height is 11 per cent. Taylor et al. (9) obtained 12 per cent for a small anthracene crystal under similar conditions. At this energy, the resolution of a typical sodium iodide crystal would be 7.7 per cent with an inherent width of 6.5 per cent and a 4 per cent contribution from multiplier statistics (13). If the liquid scintillator light output is about 1/4 that of NaI, then the multiplier contribution would be about 8 per cent and the intrinsic scintillator resolution is 7.5 per cent full width. This effect is small compared with both the width of the K-40 line and the intercept of the extrapolated line in Fig. 9.

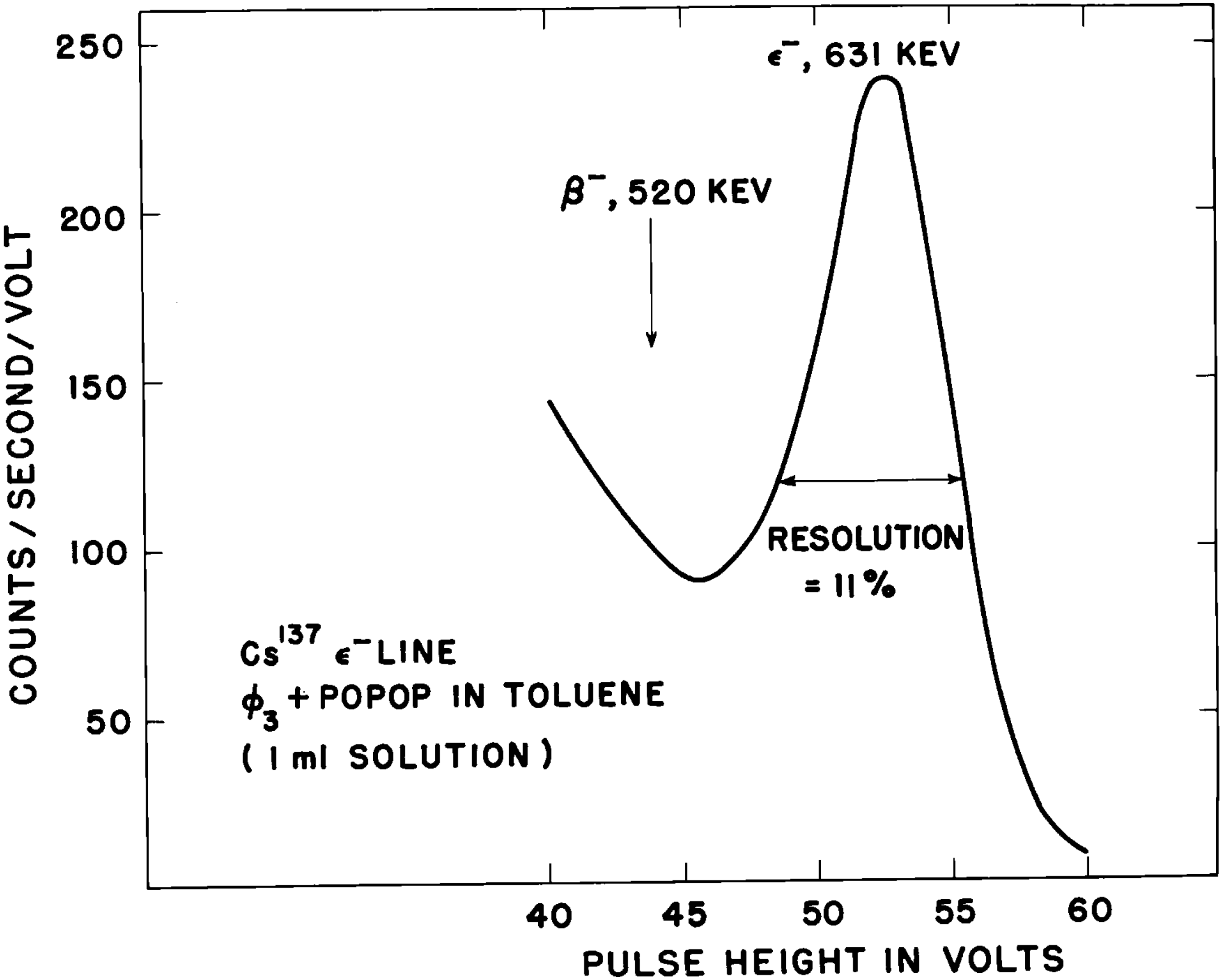

Fig. 10. $Cesium^{137}$ conversion electron spectrum in liquid scintillator.

Multiplier statistics also seem unlikely as the principal source of the observed peak widths. The half width of the Cs-137 line in Humco II is 25 per cent, or 22 per cent if one considers only the portion which has a $1/\sqrt{E}$ dependence. This corresponds to a standard deviation of $\pm$ 19 per cent, requiring approximately 25 photoelectrons. Multiplier statistics contributed an estimated 8 per cent to the small volume full width for the conversion electron line, indicating about 850 photoelectrons for 630 kev (740 ev per electron). With equal light collection and photocathode efficiency, one would expect 750 photoelectrons for the Cs-137 gamma ray. The 25 electrons calculated on the assumption that the entire line width was due to multiplier statistics, therefore, implies an efficiency of 3 per cent relative to the small volume, or 6 per cent if the cathode efficiency of the large phototube is half that of the small tube. Since Humco II has 24 per cent photocathode area, an optical mean free path of 10 meters and a wall reflectivity of better than 90 per cent, one would expect at least 50 per cent efficiency.

The third effect, shape of the Compton recoil spectrum, is certainly an important contributor to peak width. Figure 11 gives the calculated spectrum (8) for a 600 kev gamma ray assuming single scattering only. The 18 per cent full width is certainly an underestimate, since the actual peak height will be less than that of the sharp edge given by theory. The effect of multiple scattering may also operate to broaden the distribution. The Monte Carlo calculations mentioned above will give a quantitative measure of the contribution and its dependence on gamma energy.

The last two factors also contribute to an unknown degree. Both experimental and theoretical study of their importance is planned. Geometrical variations in light collection efficiency would be expected to give a constant η^2 independent of the energy of the event and might contribute to the intercept. The effect of variation in photocathode sensitivity will depend on the averaging effect of random incidence of the photons on the cathode. The efficiency of this averaging process will depend on the number of photons involved, and hence broadening by this effect could be energy dependent.

Figure 12 shows the spectra obtained with Humco II for Cs-137, Zn-65, and K-40. The latter pair could be determined simultaneously, but closer gamma rays could not.

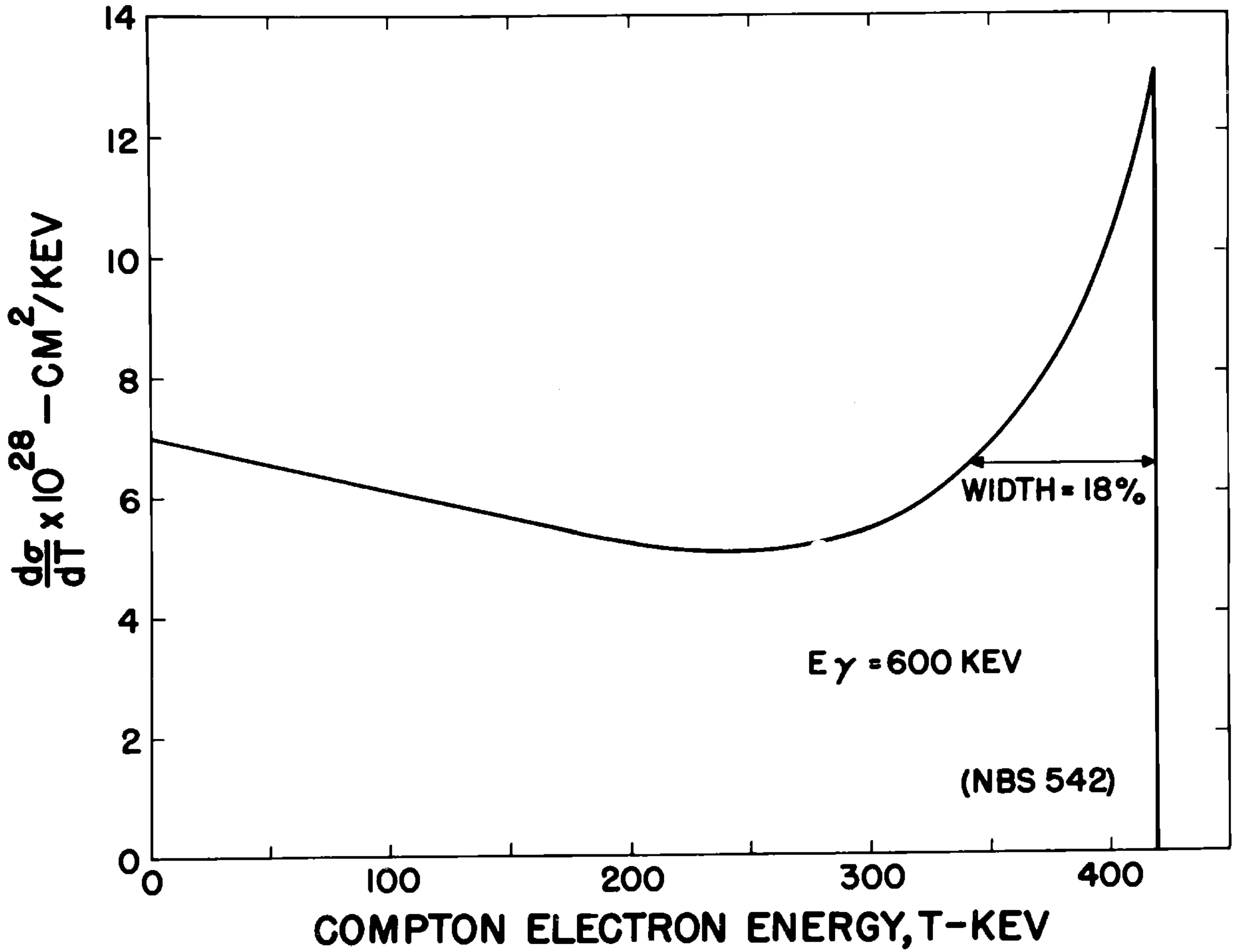

Fig. 11. Calculated shape of single Compton spectrum (600 kev).

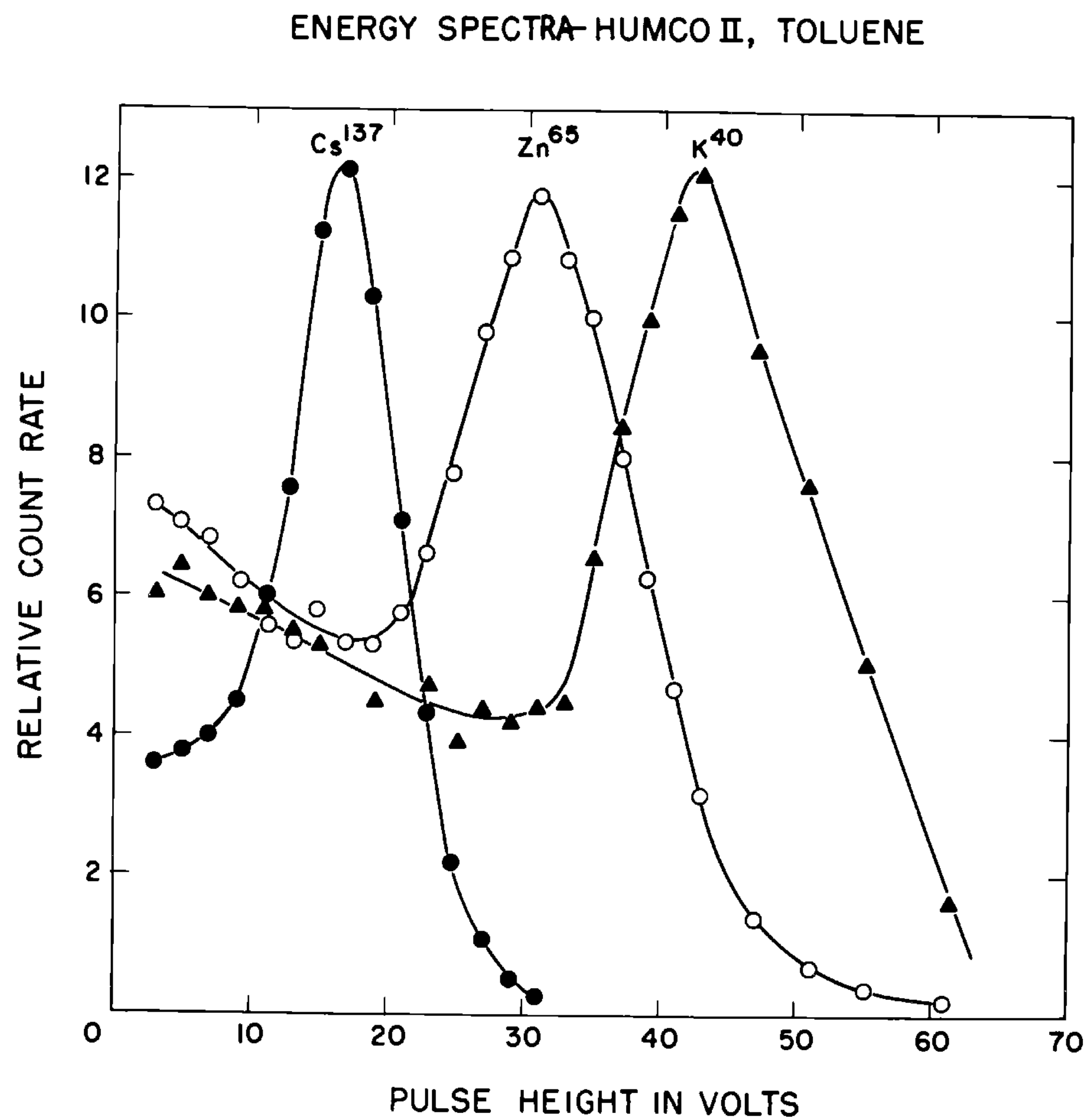

Fig. 12. Humco II spectra.

ENERGY RANGE

Because of the desirability of efficient bremsstrahlung counting (e.g., for tracer use of P-32, monitoring industrial exposure to intake of Sr-90, etc.), an important parameter of counter performance is the apparent energy at which phototube noise causes a significant increase in background. The onset of tube noise is evidenced by a sharp change of slope in the plot of background against energy on a log-log scale, and a quantitative measure of the energy at which tube noise makes a significant contribution to background is obtained from the intersection of the extrapolated linear portions of the background curves.

Figure 13 is such a graph for the Geneva Counter [triethylbenzene filling (6)] and for Humco II operated as a 2π and as a 4π system (toluene filling). For the 2π operation, the Humco II tank was filled half full of scintillator, bringing the liquid level up to the middle of the side rows of phototubes. The lower four rows of 16 phototubes were in place, and the upper two rows of 8 were replaced with aluminum foil reflectors. Only 14 of the 16 tubes were operational, the second tube in one of the bottom rows having gone dead and the second tube in the adjacent side row having become excessively noisy. These tubes were in place mechanically but were out of the circuits electrically.

During 4π operation, Humco II was completely filled with a toluene-terphenyl-POPOP solution. Twenty-three tubes were in place, the remaining position having an aluminum foil reflector. Of these tubes, 5 showed noise levels above 90 kev when tested individually in a 40 liter test tank. Substitution of better quality tubes should improve performance.

Noise levels, as defined above, for the three systems were 60 kev for Genco, 60 kev for Humco II 2π, and 100 kev for Humco II 4π. All energies are given in terms of energy deposition in the system by single Compton scattering. A deposited energy of 60 kev corresponds to an initial gamma energy of 160 kev. Measurement of Sr-90 bremsstrahlung down to this energy should give a detection sensitivity of 0.1 μc or better, or about 5 per cent of the maximum permissible body burden for workers.

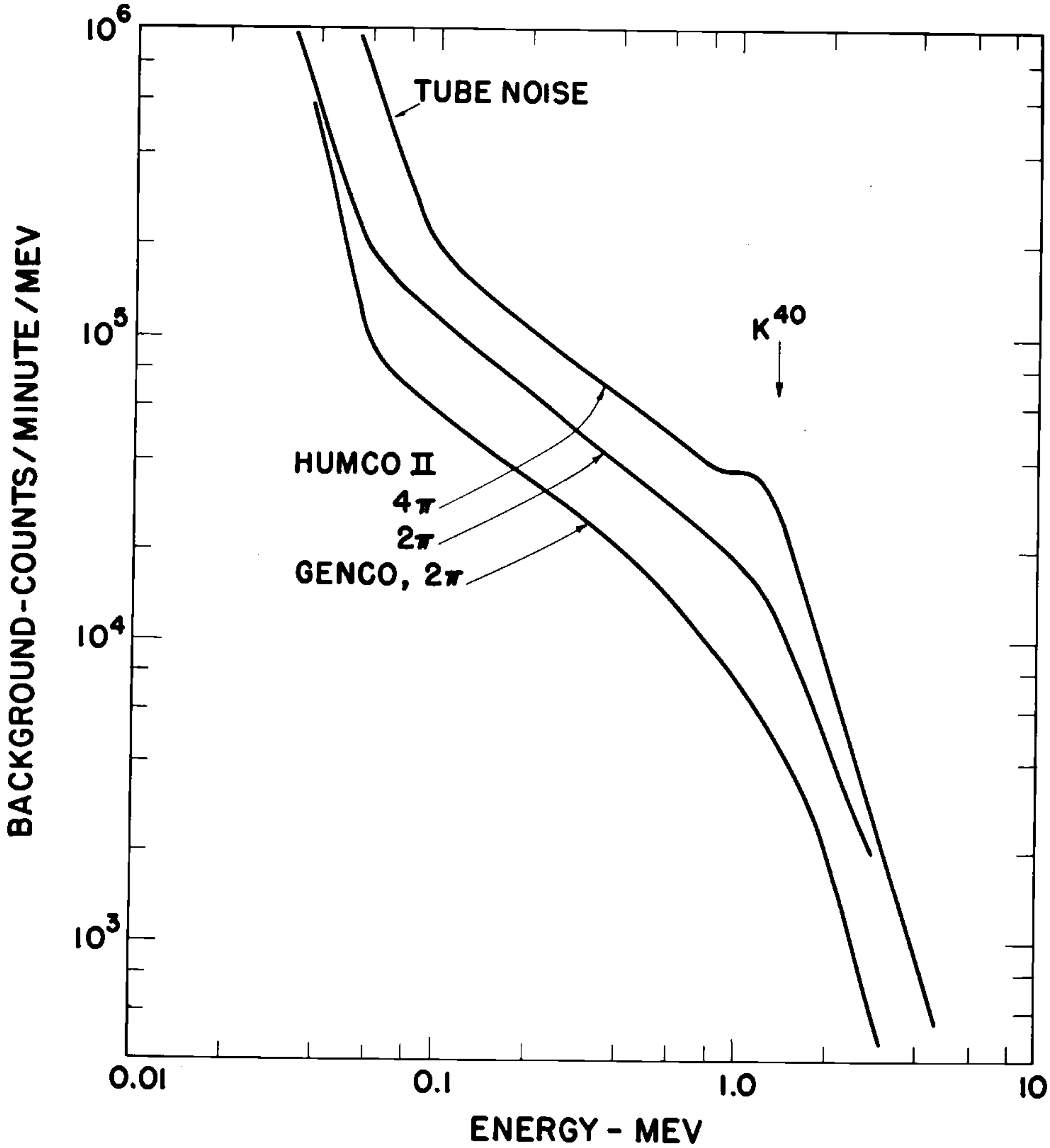

Fig. 13. Background versus efficiency (Genco and Humco II).

LOW-VOLATILITY SCINTILLATOR

While the use of toluene as a solvent has the advantage of combining economy with near maximal pulse height, it has the serious disadvantage of flammability and toxicity. Considerable effort has been devoted to development of a solvent of comparable pulse height and reduced volatility. These requirements have been met, for example by triethylbenzene (6), but only with greatly increased cost, limited availability, and difficult purification requirements. We wish to report a new solvent of low volatility which is free from these objections. The solvent, TS-28M, is available from Shell Oil Company, by whom it is sold as a paint thinner and general-purpose solvent. The composition, according to company literature, is as follows: 12 per cent paraffins, 1 per cent olefins, 25 per cent naphthenes (cycloparaffins, derivatives of cyclopentane and cyclohexane), and 62 per cent aromatics. It has a flash point of 120°F (49°C) and a boiling range of 321 to 398°F (161 to 203°C).

The material was obtained by this Laboratory in our own stainless steel drums, directly from the still at the Martinez, California, refinery. As received, it had a decided yellow cast when viewed through a large thickness. Purification was carried out by absorption of the impurities upon columns of activated alumina. Three polyethylene columns, each 8 feet x 5-1/2 in. in diameter, were packed with 50 pounds each of activated alumina (Alcoa, grade F-20, 80 to 200 mesh) to an effective height of about 80 in. The solvent traveled down the column at a rate of from 1.2 to 1.5 in. per minute, and the output was 100 to 120 ml per minute depending upon the column. These rates were self-determining. The effluent was monitored regularly for transparency and from first to last, all treated solvent was as transparent as or slightly better than reagent-grade toluene from 500 down to 390 mμ. Below 390 mμ, the transparency dropped off rapidly. No saturation of the column by the impurities was noticed, although a through-put of an individual column was up to 180 gallons.

The solubility of terphenyl in TS-28 is limited, a saturated solution at 21°C containing only 3.5 g/l. The solution used to fill Humco II, therefore, contained only 3 g/l terphenyl and 0.04 g/l POPOP to prevent possible precipitation of solute by temperature changes. At these low solute concentrations, the pulse height is reduced significantly below the optimum. Substitution of PPO as the primary solute is, therefore. recommended. Use of methyl-substituted quaterphenyl (14) would also be advantageous.

Measurements of the pulse heights of air-saturated solutions in very small volumes relative to the standard reference solution (15,16) of 3 g/l PPO in toluene gave 1.16 for the toluene filling (4 g/l terphenyl and 0.05 g/l POPOP) and 0.53 for TS-28M filling (3 g/l terphenyl and 0.04 g/l POPOP). The ratio of the relative pulse heights of the two fillings is, therefore, 0.46 in a small-volume system. In Humco II, the ratio (air-saturated) was 0.47, indicating equal transparency for the TS-28M solution. Because of the greater sensitivity of the TS-28M solution to oxygen quenching, the Humco II pulse height ratio of the deoxygenated solutions was somewhat improved to 0.55. The toluene solution pulse height rose by a factor of 1.30, and the TS-28M solution rose by 1.50 following purging of the system with argon gas for 24 hours.

Small-volume tests were performed on the Humco II TS-28M solution to which 3 g/l PPO was added. The air-saturated pulse height rose to 0.98 of the toluene-PPO reference solution, or 0.85 of the toluene-terphenyl-POPOP filling used in Humco II. Assuming equal oxygen quenching factors, one can expect the same pulse height ratio of 0.85 to hold in Humco II. A tube noise level of 70 kev is, therefore, predicted for Humco II using a TS-28M solution of 3 g/l terphenyl, 3 g/l PPO, 0.04 g/l POPOP (deoxygenated), if the complete tube complement is as good as the 14 tubes used in the 2π configuration.

SUMMARY

A new 4π liquid scintillation counter for the measurement of low-level gamma activity in human subjects has been constructed. A counting well 18 in. in diameter by 6 feet long is surrounded by a layer of liquid scintillator 12 in. thick. Scintillator volume is 450 gallons; 24 multiplier phototubes with nominal 14-1/2 in. diameter cathodes provide a cathode-to-total-wall-area ratio of 0.24. Six single channel analyzers cover the energy range 0.1 to 3 Mev. Electronic circuits are completely transistorized, and all data are automatically transcribed onto punched cards. A non-volatile solvent will be used to eliminate fire hazards. Operating characteristics, including energy resolution and sensitivity, are reported.

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