

Chemical Instrumentation

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These articles are intended to serve the readers of THIS JOURNAL by calling attention to new developments in the theory, design, or availability of chemical laboratory instrumentation, or by presenting useful insights and explanations of topics that are of practical importance to those who use, or teach the use of, modern instrumentation and instrumental techniques. The editor invites correspondence from prospective contributors.

LXXV. Nuclear Radiation Detectors (Concluded)

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PEAK RESOLUTION

Energy resolution was defined previously as the ratio of the FWHM to the peak height. When one considers resolution the following questions arise. What causes the line width? What are the reasons for the differences in the resolution between detectors? How does the energy resolution change with the energy of the radiation? Answers to these questions are discussed here.

For the sake of simplicity let us first consider systems such as ionization counters and solid state detectors where the pulse arises from ions (or electron-hole pairs) produced by the incident radiation. The number of ions (or electron-hole pairs) and, therefore, the pulse height is directly proportional to the energy expended in the detector since no multiplication is involved. However, there will be statistical variations in this number, resulting in a distribution in pulse height. This distribution is usually assumed to be Gaussian, and, therefore, the FWHM is given by the standard deviation multiplied by 2.35. The equation for R defined in Eq. 1 can be written as

$$R\% = \frac{2.35N^{1/2}}{N} \times 100 \quad (4)$$

where N is the average number of primary ions produced by the incident particle. In deriving the above equation one assumes that the ionization (or production of electron-hole pairs) is completely random. However, these processes are not uncorrelated and hence Fano has shown that FWHM will be less than that given by the above equation. The half width is actually given by

$$\text{FWHM} = 2.35F^{1/2}N^{1/2} \quad (5)$$

where F is known as the Fano factor. The Fano factor is estimated to be 0.4 for gas counters and is less than 0.2 for semiconductor counters. If one takes for the energy

required to produce one ion pair as 30 eV and an electron-hole pair as 3 eV (slightly higher than the experimentally determined value for Ge), one gets a resolution of 0.8% for ionization counters and 0.2% for germanium counters for an incident radiation of 1 MeV. It is evident from this calculation and from Eq. 4 that the resolution depends on the number of primary ions obtained from the incident radiation. For example, a plastic scintillant, which requires a higher energy expenditure per photon compared to a NaI(Tl) scintillant, is very poor in its energy resolution capability. One can also see that, since N increases with the energy of the incident radiation, resolution of the system improves as energy increases.

Equation 4 gives the theoretical value of resolution which is never attained in any practical system. In gas counters the factors affecting the resolution are: 1) low frequency noise in the chamber and amplifier; 2) source thickness; 3) variation in the rise time of the pulse; and 4) variation in the induced effects of the positive ions. As indicated earlier, the Frisch chamber reduces the effect due to positive ions.

In the case of semiconducting detectors, the electronic noise, especially of the preamplifier, is the most important factor determining the energy resolution of the system. The line width is given by

$$(\text{line width})^2 = (\text{detector noise})^2 + (\text{amp noise})^2$$

Detector noise is the contribution to FWHM by the detector as given by Eq. 4. Use of high impedance field effect transistors at the input of the preamplifiers has reduced the amplifier noise in detector systems. There are commercial Si(Li) detector systems available claiming an FWHM of 175 eV for 5.9 keV x-rays. Special care should also be taken to reduce low frequency microphonics.

In a proportional counter with a cylindrical or spherical shape, the electric field

is concentrated near the anode and, therefore, about half of the ion pairs are formed within a mean path of the central wire. Each primary electron coming to this region of high field produces a small avalanche as the secondary electrons are accelerated to the electrode. These avalanches have no interaction among one another. Hence the number of ions finally collected is the sum of ions in each avalanche and is proportional to the primary ionization. Statistical variations in each avalanche add to the FWHM as well as the already mentioned variations in the primary ions. Figure 7 compares the x-ray spectrum of ^{57}Co obtained from a proportional counter and a Si(Li) counter.

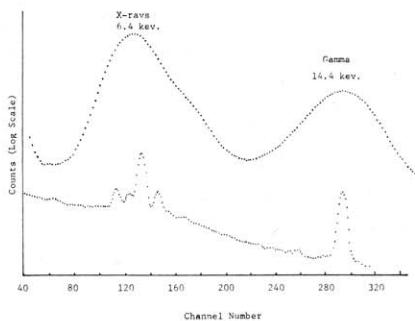


Figure 7. Pulse-height distribution for 14.4 keV and 6.4 keV radiations from ^{57}Co obtained with a proportional counter and a Si(Li) detector. The different groups of x-rays in the 6.4 keV peak are well resolved by the Si(Li) detector.

Discussion of energy resolution in a scintillation counter is much more complicated because of the different processes involved, such as 1) production of scintillant photons; 2) collection of light at the photoelectrode; 3) production of electrons at the photoelectrode; 4) collection of photoelectrons at the first dynode; and 5) electron multiplication at the different dynodes. In addition to the normal variations in each of the factors mentioned above, there are non-normal variations. Some of them are: 1) inhomogeneous luminous efficiency throughout the crystal, mainly due to variations of the Tl concentration; 2) non-proportional scintillation response, that is, luminous efficiency varying with specific ionization; 3) multiple interaction effects; and 4) nonuniform photomultiplier response over the area of the photocathode. A good discussion of the effects of these can be found in chapter 5 of reference (6). A very simple equation for resolution due to Neiler and Good (8) is

$$\Delta E/E = 2.35N^{-1/2}[1 + \beta^{-1}]^{1/2} \quad (6)$$

where N is the number of photoelectrons produced by the radiation, ΔE the half width for the energy E , and β is related to the gain per stage of the photomultiplier. The quantity β takes care of the contribution to FWHM given by the variation in amplification at the dynodes. Since N is proportional to the input energy if β is

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held constant (constant applied voltage), ΔE will be proportional to $E^{1/2}$. The results of a recent study (9) are shown in Fig. 8. It may be pointed out here that the conventional plot is resolution in percent vs (energy) $^{-1/2}$.

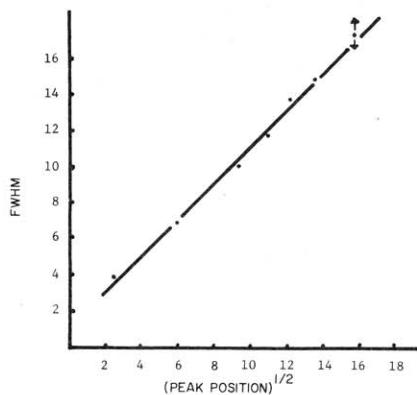


Figure 8. Variation of width at half-maximum for photopeaks (in channel numbers) with square-root of peak position.

The resolution for energies in approximately the 1-5 MeV range for NaI is 7-10%. To obtain the best resolution requires careful matching of the photomultiplier sensitivity, and the scintillant spectra, proper optical coupling between the crys-

tal and the photomultiplier, selection of a low noise photomultiplier and crystal with uniform Tl distribution and suitable reflectors.

The tremendous improvement achieved in semiconductor counters has helped studies in several fields. The effect of this on nuclear spectroscopy is best illustrated by Fig. 9. This figure shows how the nuclear spectroscopic information improved first with the development of scintillation counters and then with the use of Ge(Li) counters (10). Use of Si(Li) counters have also made the x-ray fluorescence spectroscopy a routine laboratory nondestructive testing method. A good description of this method may be found in the article by Liebhafsky and Pfeiffer (11). The availability of large volume Ge(Li) detectors has also increased the capability of activation analysis.

It may be pointed out here that there are methods by which better energy resolution can be achieved. Magnetic spectrometers give resolutions of the order of 0.02% in the keV range for beta rays and 0.6% for particles in the 6 MeV region. Also, it is interesting to note that variation in energy of 1 part in 10^{13} can be detected by the Mössbauer effect even though the precision with which the absolute energy can be measured with a crystal diffractometer is only ~ 1 part in 10^4 . However, from the viewpoint of efficiency, the possibility of simultaneously detecting radiations of different energy, high resolution and simplicity of the whole system, semiconductor counters are in an enviable position.

EFFICIENCY OF COUNTERS

The number of particles counted in most cases is less than the number emitted by the radioactive source. The difference between the two numbers depends on the geometry of the experimental arrangement, thickness of the sample, nature of the backing, absorption in air and in the window of the counter, efficiency of the detectors and coincidence loss due to the overlapping of the pulses. None of these factors except the last two will be treated in this article. Detector efficiency and coincidence loss are discussed in the following section. The detector counting efficiency, defined as the ratio of counts to the number of radiations reaching the active volume of the detector, is sometimes called the intrinsic efficiency.

Geiger counters have an inherent 100% efficiency for charged particles such as alpha and beta radiations. (The coincidence loss due to overlapping of pulses will reduce the counts and hence the efficiency. This loss will be discussed in the next section.) Pulses produced by them are of the same height even for those produced by a single electron-ion pair. Hence radiation particles of energy down to 30 eV will be counted by a Geiger-Müller counter. If a radioactive source can be introduced into the counter, one can determine the absolute activity very accurately. In such experiments corrections have to be made for wall effect (source decaying near the wall). The method is especially suitable

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for low energy beta sources. Proportional counters are also capable of detecting particles with 100% efficiency. For detection of gamma rays gas counters are not suitable: the maximum efficiency one can get is $\sim 15\%$ for 2 MeV gamma rays with proper design of the counter. The counter wall material and its thickness should be selected such that a gamma ray produces an electron, which then reaches the active volume of the counter. Higher efficiencies are obtained in x-ray detection (Fig. 10). Efficiency decrease at the low energy end is due to the absorption in the window and at higher energies, to the decrease in the photoelectric cross section.

The efficiency of scintillation counters for different radiations will now be considered. Scintillation counters such as ZnS(Hg) crystals can detect alpha particles from radioactive sources with 100% efficiency. In beta counting with scintillants,

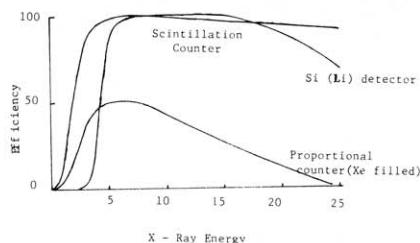


Figure 10. Variation of x-ray detection efficiency as a function of energy for proportional, scintillation, and semi-conductor detectors. The cut-off at the low energy side is due to absorption in the window of the counter.

special consideration should be given to two factors: 1) backscattering and 2) continuous energy distribution for beta rays from radioactive sources. Backscattering, as mentioned earlier, is very small for organic and plastic scintillants. Various 4π -geometry arrangements, such as sources

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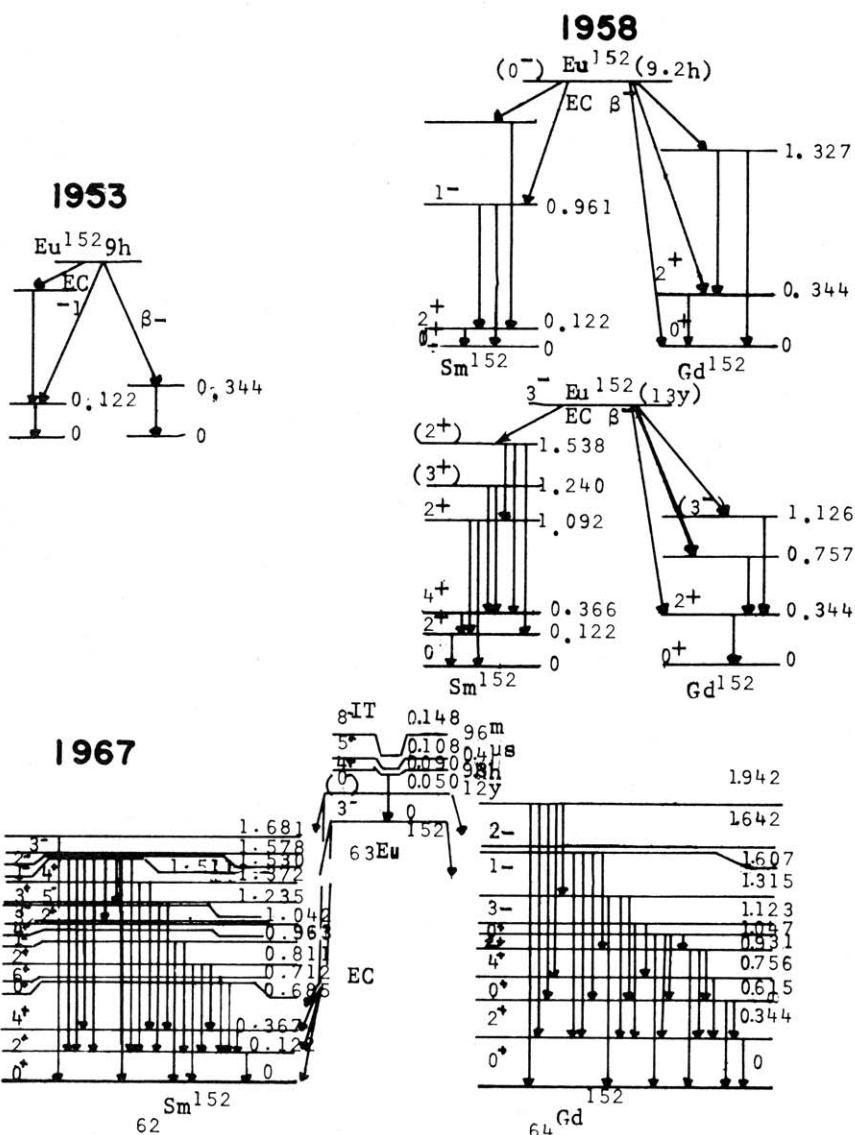


Figure 9. The decay schemes of ^{152}Eu to produce on the one hand ^{152}Sm and on the other ^{152}Gd . Data from the 1958, and 1967 Tables of Isotopes. This illuminating comparison, showing the progress made in nuclear spectroscopic studies with advances in detector technology, first appeared in an article by Hamilton et al. (10). The 1967 data were obtained with a Ge(Li) detector.

located between split crystals, and sources mixed with liquid scintillants, give very high efficiencies. However, counting low-energy beta rays from ^{14}C ($E_{\max} = 155 \text{ keV}$) and ^3H ($E_{\max} = 19 \text{ keV}$) is always a problem. Difficulty in these cases arises from the inability to distinguish between signals due to the few photoelectrons produced by the beta rays and signals due to the thermal electrons from the photomultiplier. Cooling the photomultiplier tubes and looking at the scintillant with two photomultiplier tubes in coincidence are two often used techniques to reduce the noise. There are excellent commercially available liquid scintillant systems utilizing both these techniques which give 90% efficiency for ^{14}C beta rays and 30% efficiency for ^3H beta rays.

Inorganic scintillants, of which NaI(Tl) is the most commonly used, have the best efficiency for gamma detection. Since the absorption vs thickness is an exponential function, complete absorption of gamma rays and 100% efficiency requires an infinite thickness for the detector. However, because of the high values for absorption coefficients for low energies (<300 keV) for a 2-in. radius \times 2-in. thickness NaI crystal, detection efficiency is close to 100%. The detection efficiency, as mentioned earlier, depends on the lower limit of the pulse height selector because of the large number of pulses of lower pulse height in the Compton distribution. To avoid this difficulty, intrinsic peak efficiency is defined as the probability that a gamma ray of given energy will produce a pulse falling in the full energy peak. A pulse may fall in this peak when the gamma ray interacts with the scintillant by photoelectric effect or by multiple interactions involving combinations of photoelectric effect, Compton effect, and pair production. In a complex spectrum, the standard procedure to obtain the efficiency is to break down the spectrum into components by first drawing in the shape of the most energetic gamma component and then obtaining by successive subtractions the lower energy components. Figure 11 shows the variation

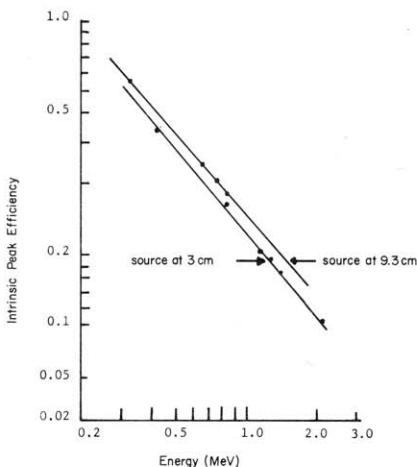


Figure 11. Variation of intrinsic photopeak efficiency as a function of energy (2 \times 2 in. crystal) for two source distances (9).

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in intrinsic peak efficiency as a function of energy for two different source to crystal distances. The intrinsic photopeak efficiency is higher for the larger source to crystal distance because of the increase in the number of gamma rays of near normal incidence.

Semiconductor counters have very good detection efficiency, ~100% for charged particles. X-ray detection efficiency, especially in the range 10–50 keV, is almost 100%. Because of the high atomic number required to get better detection efficiency, Ge ($Z = 32$) rather than Si ($Z = 14$) detectors are used for gamma rays. Detection efficiency for Ge(Li) detectors is about 20% of that for NaI(Tl) crystals.

PULSE WIDTH AND TIME RESOLUTION

The time characteristics of pulses from detectors are very significant. The possible use of a detector in coincidence and in particle identification systems depends on the rise time of the pulse. Another factor of importance is the total width of the pulse which determines the loss in counts due to overlapping. In most detectors except in the Geiger-Mueller counter the total width has some relationship to the rise time of the pulse. The time constant of the detector output stage has to be large enough compared to the rise time to keep the pulse height information.

Time characteristics of pulses from gas

counters are very poor. In ionization counters the drift velocity of the ions is very small because of the low applied voltage. Most of the pulse is derived from the fast electrons, reducing the effect of slow moving positive ions and thereby decreasing the time width. As indicated earlier, this approach has also improved the energy resolution of these detectors. In proportional counters, because of higher applied fields, ions have greater drift velocities. Here the major contribution to the voltage drop is due to the positive ions which travel in a very short time a small distance from the central wire. In this distance most of the potential drop occurs. There is an associated uncertainty in the starting time of these individual pulses if the radial positions of the ions relative to the central electrode are different. This uncertainty could vary from 0.1 μ sec to 1 μ sec, the time increasing with the pressure of the gas in the counter. It is, therefore, not advisable to use proportional counters for coincidence work. For G-M counters, the pulse width is 100 times higher (~100–300 μ sec). Also during the movement of the sheath of positive ions out to the negative electrode the potential inside the counter drops below the limit required to sustain avalanches. An incident radiation during this time will not be registered. Therefore, the resolving time is very high in these counters, giving a higher coincidence count loss.

In semiconductor counters the pulse rise time is the transit time of the charge carrier across the depletion region. The transit time is given by the equation

$$t_0 = \frac{w}{\mu E} \quad (7)$$

where w is the width of the depletion region, μ the mobility and E the electric field. Knowing the value of μ , one can estimate the rise time. The usual range of rise time is of the order of 10 to 100 nsec.

In the case of scintillation counters pulse shape depends on the lifetime of the electronic excited levels in the scintillant and the RC time constant of the resistance and the stray capacitance at the collector. If only one excited level is assumed in the scintillant, one obtains the equation

$$V(t) = \frac{N_p M e}{C} \frac{RC}{RC - \tau} e^{-t/RC} - e^{-t/\tau} \quad (8)$$

where N_p is the number of electronic states excited by the incident radiation (which is equal to the number of photons), M the multiplication at the different stages including the photoelectric conversion factor, e the electronic charge, R resistance, C capacitance and τ the lifetime of the excited state. The rise time of the pulse depends on τ . For plastic scintillants, $\tau = 3.2$ nsec; for stilbene, $\tau = 6$ nsec; and for NaI(Tl) $\tau \approx 0.2$ μ sec. Therefore, in experiments where fast rise times are required, such as, short lifetime measurements, plastic scintillants are preferable. There are more than one electronic level in all the scintillants with varying time constants. The levels excited by dif-

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ferent radiations differ and, therefore, the nature of the particle determines the shape of the pulse, especially on the rising side. This fact has been utilized in identifying the incident particle.

DETECTION OF NEUTRONS

Neutrons interact with matter mainly through two processes: elastic collisions and nuclear reactions. In elastic collisions energy loss depends on the angle of collision. The maximum energy after collision is the energy of the incident neutron, T , and the minimum energy is given by

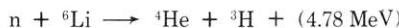
$$T_{\min} = T \left(\frac{M_1 - M_2}{M_1 + M_2} \right)^2 \quad (9)$$

where M_1 is the mass of the nucleus; M_2 the mass of the neutron; and T the energy of the neutron before collision. Neutron-proton elastic collisions are used for the detection of neutrons. The neutron is allowed to strike a thin foil of hydrogenous material on the wall of the counter. Protons will then emerge from the foil with energy between 0 and T .

Organic scintillants are particularly suitable for neutron detection because of the high proton content. By correcting for the nonlinear energy response of the scintillant one can also determine the energy of the neutrons. The presence of gamma radiation sometimes complicates these measurements. However pulse shape discrimination methods are found to be successful in discarding the effects of gamma

rays. This is very successful when stilbene crystals are used as scintillants.

Of the neutron-induced reactions we are interested in those which produce charged particles. The most commonly used reactions are



All these reactions have high cross section for low energy neutrons and are, therefore, especially suitable for the detection of thermal neutrons. The counters are usually designed such that the reaction products lose all their energy in the active volume of the counters.

The gaseous compound BF_3 is suitable for use in gas counters for thermal neutron detection. This counter is usually used in the proportional region enabling the smaller pulses from gamma rays to be readily discriminated. Fission reactions resulting in the release of very high energy of 200 MeV are used in ionization chambers. Lithium iodide crystals activated with Ag or Eu are used in slow neutron detection. With proper discrimination one can count only thermal neutrons since the pulses produced by them form a distinct peak and are well separated from the pulses produced by background radiation or by other reactions in the crystal. For a 95% enriched $\text{Li}(Eu)$ scintillant, close to 100% neutron detection efficiency can be obtained for a 0.5 cm crystal.

As in other types of detectors, semiconductor detectors of neutrons use neutron-proton elastic scattering or one of the reactions, ${}^6\text{Li}(n,\alpha){}^3\text{H}$ and ${}^3\text{He}(n,p){}^3\text{H}$. In most of the neutron counting systems, a thin layer of the reacting material is kept between two surface barrier detectors so that the reaction products fall on these detectors. The sum of the pulses will be proportional to the energy of the neutron plus the energy Q released in the reaction.

RECENT DEVELOPMENTS

There have been several interesting developments in detector technology in recent years, such as position-sensitive proportional counters and position-sensitive semiconductor counters, multi-wire proportional counters, particle identification by pulse shape analysis in scintillation counters, use of scintillant gases to obtain fast rising pulses, etc. Principles behind some of these developments are discussed below.

One of the main differences between the proportional counter and the Geiger counter is that in the proportional counter ionization is limited to a small region around the path of the incident particle while in the Geiger counter it spreads throughout the counter. Therefore, it is possible to get some information about the position of the incident radiation in the proportional counter. Figure 12 is a block diagram of a position sensitive proportional counter system. The anode is a high resistance wire (usually a quartz fiber coated with carbon). Assume the particle incident at position X produces ions in the vicinity of the anode. These ions are collected on the anode and cause current flows in both directions through the anode. The amount of

current flow in each direction depends on the resistances of the two paths. Because

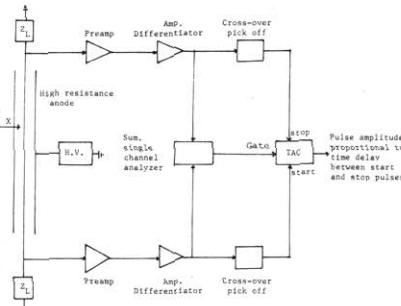


Figure 12. Block diagram illustrating the operation of a position-sensitive proportional counter.

of this difference, the pulses produced at the ends of the detector will differ in their height and rise time. Differences in the rise times, because of the difference in the time constants, are generally used in obtaining information about the position. The pulses are amplified and doubly differentiated. The cross-over pick-ups produce pulses at the cross-over points. The cross-over points change with the rise time and, therefore, the difference in the cross-over points will reflect the change in the resistance along the two paths of the anode. The time-to-pulse amplitude converter produces a pulse with an amplitude dependent upon the time difference between the pulses going into it and, therefore, the height of the pulse will be a measure of the position of the incident radiation. Position resolution of the order of 0.1 cm has been obtained for a 20 cm long counter (12).

Counters with several anodes capable of giving two dimensional information are widely used today. Such multi-wire proportional counters are expected to replace photographic films in x-ray diffraction and radiographic studies in a few years. One major advantage of such a system is that studies can be done much faster with weak x-ray sources.

Position sensitive semiconductor detectors, known as nuclear triodes, are commercially available (13). Figure 13 is a schematic diagram illustrating the principle of these detectors. A surface barrier diode is formed on a long strip of n -type material with a conducting gold surface. On the back side is a resistive layer with two contacts, one grounded and the other connected to a charge-sensitive preamplifier. The charge flow through these contacts will be divided according to the resistance from the point of incidence to the respective contacts. Therefore, the pulse height E_x will have information about the distance between the point of incidence and terminal 3. Pulse height will be a maximum for zero distance. The charge collected at terminal 1 will be proportional to the total energy of the radiation. Counters with position resolution of 1% of the total length for a 5 cm long counter are easily available.

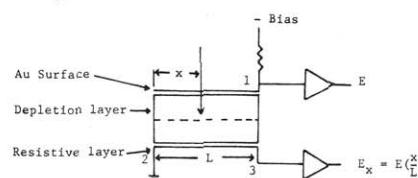


Figure 13. Schematic diagram of a position-sensitive "nuclear triode."

Another recent development is that of intrinsic germanium detectors. These detectors can be cycled to room temperature repeatedly without damage to the crystals. This cannot be done in Ge(Li) detectors because Li diffuses out of Ge at room temperature. Also the intrinsic germanium detectors are practically windowless.

PARTICLE IDENTIFICATION BY PULSE SHAPE DISCRIMINATION

This process depends on the fact that the pulse shape will differ for different particles in organic scintillants such as stilbene and liquid scintillants (14). Each scintillant decays with a short and a long time constant. The fraction of light produced in these two groups depends on the exciting particle. Therefore, when the signals are amplified and integrated, they will have different rise times. These pulses when doubly differentiated cross the zero level at distinctly different times from the start of the pulse. This time is measured in terms of the pulse height produced in a time-to-amplitude converter. The start and stop pulses are produced respectively at the start of each pulse and the cross-over point. (See Fig. 14). The shapes of the signals at different points are indicated in the figure. This method is especially suit-

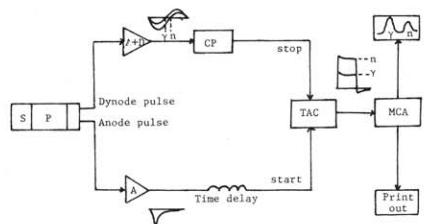


Figure 14. Block diagram illustrating the principle of pulse shape discrimination. Neutrons and gamma rays can easily be separated by this method using organic or liquid scintillants. S, scintillant; P, photomultiplier; A + D, amplifier and differentiator; CP, cross-over pick-off; TAC, time-to-amplitude converter; MCA, multi-channel analyzer.

able for distinguishing between neutrons and gamma rays in the same energy range.

Totally depleted semiconductor detectors are successfully used for particle identification. In this method a totally depleted thin crystal is kept in front of a second thick detector. The sum of the signals from the two detectors gives the total energy, E , of the radiation and the signal from the first detector gives the part of the energy, dE spent in the crystal of thickness dx . Thus knowing dE/dx and E one can obtain information about the nature of the incident particle.

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