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SOLID COUNTERS: SCINTILLATION COUNTERS

by

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INTRODUCTION

In the early phases of nuclear research, scintillation counters proved valuable for detection of alpha particles. In this technique, as perfected by Rutherford and his co-workers, scintillations produced by the excitation of fluorescent and phosphorescent screens were observed visually. The method has been revived in modern form, principally by Kallman,¹ with the use of photomultiplier tubes for the observation of light pulses, and large, transparent crystals for the conversion of as much of the energy of a penetrating particle into light as possible.

A wide range of fluorescent materials has been tested by various workers.¹⁻⁸ On the basis of reported efficiency and response time, the work at this laboratory was initiated using naphthalene. The first experimental arrangement consisted of a 1P21 P.M. tube mounted in a glass container stuffed with moth flakes and immersed in liquid air as shown in Figure 1. With radium gamma ray source, a ratio of about 20 was observed between the counting rate of this set-up as compared with that of the photomultiplier surface alone exposed to the gamma radiation under otherwise identical conditions.

As a consequence of the sensitivity (about 10 per cent gamma counting efficiency) of even this crude and inelegant device, as well as of its very simplicity, a program was undertaken with the aim of devising a rugged and adaptable basic counter unit. This program received added impetus when it was discovered that the much greater usable light output of anthracene made possible the elimination of the refrigerant. With respect to pulse width, it will be seen that the fortuitous condition exists whereby the minimum, independently observed pulse width coincides with the maximum resolution of simple, fairly conventional electronic recording equipment. Such apparatus does not presently exist in useful quantity; however, it is feasible and economically advantageous to construct adapter units, containing fast discriminator, coincidence and prescaler stages, that would permit use of the greater part of existing scaling equipment.

THE DETECTOR AND AMPLIFIER

In order to further investigate the characteristics of the scintillation counter, a more convenient refrigerated photomultiplier unit was constructed. (Figure 2) The associated electronic equipment was of conventional form as indicated in Figure 3.

Detection Material

Research on fluorescent materials has centered on the basic condensed aromatic (benzene) molecules, such as naphthalene and anthracene. A number of good features may be enlisted in favor of these compounds as compared with the many fluorescent inorganic substances:

- 1) The aromatic molecules (containing only hydrogen and carbon) have but one neutron-induced activity of appreciable cross section, namely, the $n-2n$ carbon reaction, with a 20 Mev threshold, resulting

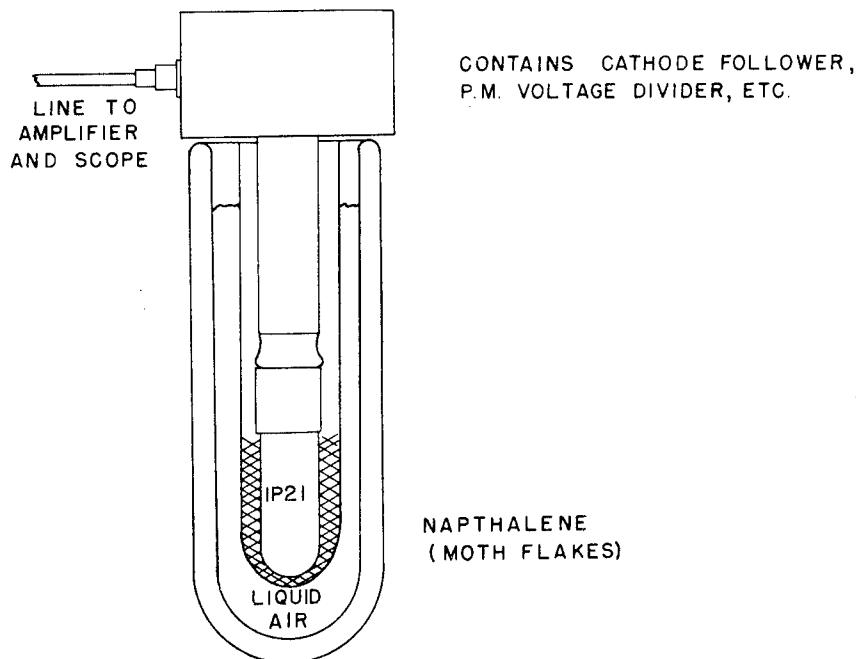


Figure 1.

in a 20 minute beta activity. On the other hand, the inorganic fluorescent materials generally contain heavy nuclei, with many possible activities, which could contribute to the counting background. The hydrocarbons have the further advantage of decaying quickly, hence, restoring themselves to usable form after intense exposures.

2) Most of the suitable inorganic substances would be next to impossible to crystallize artificially in an economic manner.

3) The response time of the hydrocarbons as counters is of the order of $1/10$ microsecond. In contrast, few of the efficient fluorescent inorganic substances are free from phosphorescences persisting anywhere from microseconds to seconds. This is, to a certain extent, a consequence of the more complicated energy level structure of the inorganic molecules, for it is reasonable to expect that partially forbidden transitions giving rise to both fluorescence and phosphorescence would be much more probable in such a molecule made up of atoms of high atomic number. On the other hand, fluorescence in the basic aromatic molecules exhibits properties which vary in a regular way with the degree of condensation. It appears to be characteristic of the ring structure and seems to be a relatively "clean" phenomenon.

4) Anthracene stands out in particular in so far as concerns the conversion efficiency (the ratio of the particle energy to total light pulse energy) for radiation of the proper wavelength to match the photomultiplier sensitivity maximum. We have confirmed the results of Bell⁵ in this regard; the light output of anthracene in this range is of the order of five times that of naphthalene. This is more than enough to eliminate the encumbrances of refrigerating the photomultiplier, as was formerly required to reduce the noise background. The scintillation counter thus attains the status of a practical laboratory instrument.

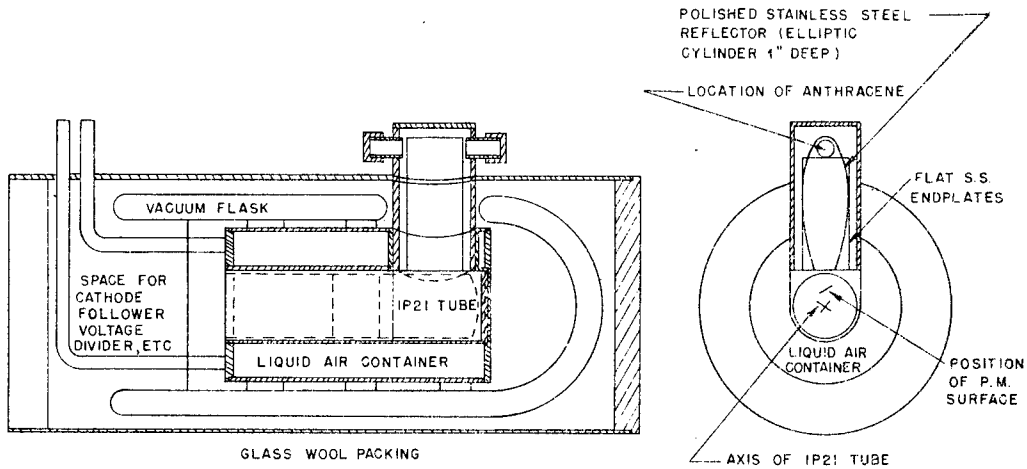


Figure 2.

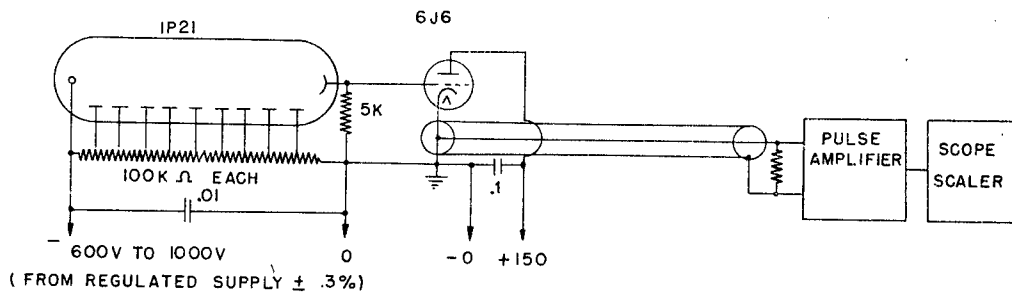


Figure 3.

The wavelength of emitted fluorescence of these condensed aromatic molecules increases with the degree of condensation; the anthracene radiation happens to coincide with the 1P21 sensitivity. Ultraviolet radiation-induced visible fluorescence serves as a rough check of the radiation wavelength and intensity. Anthracene glows a brilliant blue, whereas naphthalene shows a faint purplish glow. The various aromatic isomers of anthracene exhibit about the same behavior as anthracene but might prove difficult to procure in comparable quantities and purity.

Crystallization of Anthracene

A crude test of an impure granulated sample of this material made in the original experimental arrangement of Figure 1 gave initially negative results. In view of the progress made with naphthalene* no further steps were taken until Bell and Davis at Oak Ridge National Laboratory reported their remarkable success with crystalline anthracene.⁵ By a very exacting procedure they obtained clear anthracene of appreciable thickness. We have since duplicated their results, confirming that anthracene of extraordinary purity is required and that extreme care is necessary in cooling the melt.

The procedure adopted here used Eastman Kodak Organic Chemical, Catalog No. 480-X, "anthracene, blue-violet fluorescent." This is sealed in a flask together with about half an atmosphere of pure argon. Since the material has an appreciable vapor pressure at room temperature, liquid air trapping is essential to prevent poisoning of the vacuum system when evacuating and refilling the flask with argon. Considerable quantities of noncondensable gas can be evolved by shaking and "tickling" the contents with a spark coil during evacuation. This entire procedure is necessary to prevent oxidation of the melt in the oven.

An accurately controlled, ($\pm 1^\circ\text{C}$) well-insulated electric oven is used for melting and crystallization. To date we have obtained reasonably clear large conglomerates by cooling through the melting point (218°C) at a rate of 3 to 5 degrees per hour. Sections of such a mass yield crystals of complete transparency up to 3/16 inch thick and as large as 3/4 inch diameter. The behavior during and appearance after crystallization indicates that several modifications might pay:

- 1) Inclusion of a single stage of recondensation for further purification. This could readily be performed "in vacuo" just prior to the argon filling.
- 2) Crystallization in a flat-bottomed, straight-walled (cylindrical) "inert" metal container.
- 3) Even slower, more constant cooling rate, approaching one degree per hour, with a more carefully controlled space temperature gradient across the flask (one degree or so).

The crystalline material is presently extracted by physically removing the flask.

Optical Systems

Numerous means come to mind for collecting and concentrating the fluorescence onto the photomultiplier surface. Except for special applications, the first two general methods described below fulfill the conditions of simplicity and efficiency.

- 1) Direct observation. Intimate contact is maintained between detector and photomultiplier by fastening the anthracene crystal directly to the glass bulb. This simple arrangement makes an easily assembled "standard" for the comparison of optical systems, materials, etc., as well as for general survey and radiation counting purposes in which coincidence schemes might prove unnecessarily elaborate. (See the discussion on "signal-to-noise" ratio.) Sample characteristics of a typical "direct observation" unit are cited below.

For coincidence experiments it would be necessary to accept the slight reduction in light collection efficiency resulting from the small separation required to allow a line-of-sight path between crystals.

*Clear cylinders of naphthalene are easily made by slow cooling of the molten commercial moth flakes.

2) Multiple internal reflection systems. A successful arrangement employing a 1.5 inch diameter silvered glass tube three feet in length between the anthracene detector and photomultiplier has been described by other workers in this field.

Polished Lucite rods of lengths up to 12 feet and diameters ranging from 1/2 inch to 2 inches have been used in a similar manner at this laboratory. For distances up to 3 or 4 feet there is only slight reduction of efficiency over the "direct observation" scheme, for the solid angle subtended by the photomultiplier is almost as great. This method is presently by far the best and most practical for transmission over intermediate distances.

3) Elliptical reflectors. The crystal is located at one focus of an elliptical reflector with the photomultiplier at the other. This was the first true "optical system" tested using naphthalene. For small geometries it is an easy matter to get practically complete light collection. Polished stainless steel surfaces are used in the arrangement illustrated in Figure 2. For small effective counting volumes, and where the maximum of efficiency is desired for short distances, the method is highly recommended.

Reflecting back surfaces for the crystals used in the other methods will in general improve the optical efficiency, depending on the transparency of the material.

4) Lens systems. In one arrangement that employed a hollow probe 20 feet long with parallel focusing 2.5 inch diameter lenses at each end, light was collected from a fluorescent crystal inserted inside the 184-inch cyclotron. The foci were adjusted to coincide with the crystal and photomultiplier. This system showed a light collection efficiency of perhaps 10 per cent as compared with the "direct observation" scheme; the efficiency remained relatively independent of lens separation provided accurate focussing was maintained. For long distance transmission there is no alternative.

The Photomultiplier

The choice of the 1P21 from among the several commercially available types of photomultipliers for the light pulse detector is dictated by the following considerations:

- 1) Highest sensitivity and gain.
- 2) Best construction. The 1P21 is much superior from the standpoint of internal construction, shielding, and insulation. This is particularly important under the high voltage conditions to be described.
- 3) Lowest thermionic noise due to statistical fluctuations in cathode emission. At a given temperature this will be lowest for surfaces having a high work function. This indicates the use of a blue-sensitive tube, and hence blue fluorescent particle detector, such as the 1P21. Red-sensitive surfaces, besides having the higher thermionic emission resulting from their lower work function, will also show photosensitivity due to direct heat radiation.

In connection with thermionic fluctuation noise, the thermionic emission (dark current) of a 1P21 was measured to be about 10^{-13} ampere or 2.5×10^6 electrons sec. Since the resolution time of the amplifier was about 3×10^{-7} sec, the observed noise pulses due to thermionic emission should then appear as pulses corresponding to at most a very few simultaneously emitted electrons. This is not the case. Some isolated noise pulses correspond to the simultaneous emissions of as many as 25 electrons, probably due to the departure from good statistics. The constants of Richardson's equation are measured average quantities; over many seconds these surfaces do obey this relation. However when the time scale is magnified as it is here, it becomes necessary to postulate that the emission of electrons takes place in bursts.

From the standpoint of kinetic theory this is quite reasonable. On the assumption of a momentary statistical concentration of energy and a consequent increase of effective "temperature" in a localized region of the surface, electrons can be emitted profusely at that spot. The "cooling" period of such a local hot spot of dimension such as to be commensurate with the observed emission turns out to be about 10^{-7} seconds. Incidentally, in connection with the use of photomultipliers with naphthalene it was determined that dry ice temperatures are sufficiently low to eliminate the thermionic noise originating in the photomultiplier.

Pulse and Noise Characteristics of the Overall Detector

For purposes of comparison, the operating characteristics of a so-called "direct observation" counter, connected in much the same way as that indicated in Figure 3 might be mentioned.

Geometry: 1 x 1 x 1/4 inch anthracene (translucent) fastened against the glass bulb of a 1P21 P.M. tube immediately opposite the light-sensitive area. (Nonreflecting background)

Photomultiplier operating conditions: (Room temperature) 70 volts per stage (Total gain—200,000) 20 micromicrofarad anode circuit capacitance, 500 ohm anode load resistance.

Radiation: Gammas, from one mg standard radium source.

Pulse Height: Up to .3 volt at photomultiplier output.

Sensitivity: (Discriminator adjusted to eliminate thermionic noise) Counting efficiency of about 40 per cent. Unit readily detects one mg source at 12 feet.

Background: Essentially cosmic ray plus laboratory radiation.

In this arrangement, a simple calculation shows that an observed .3 volt pulse corresponds to about 120 electrons being emitted from the cathode surface. Taking into consideration the approximately 10 per cent conversion efficiency (the ratio of incoming to outgoing flux) of this surface the gamma-induced pulse heights indicate a conversion efficiency for the anthracene of about 25 per cent. This figure checks with the observed brilliance under ultraviolet excitation which is but slightly less than that of a blue-white phosphor having a reputed efficiency of 40 per cent.

If we define "noise" more generally as those pulses which do not originate as a result of the passage of a desired ionizing particle in the fluorescent crystal, it is then possible to identify a number of noise sources.

- 1) Statistical fluctuations in thermionic emission.
- 2) Laboratory background radiation.
- 3) Cosmic ray pulses.
- 4) Induced activity in the crystal or surrounding structures.
- 5) Light leakage in the optical system. Extreme precautions must be taken to exclude all external light from the optical system. In connection with this problem, it should be noted that exposure to relatively weak light with normal voltage applied to the electrodes can result in the destruction of the photomultiplier as a radiation detector-amplifier. The manufacturers suggest a maximum light intensity such that the heat dissipation of the anode will not exceed 1/4 watt at normal operating voltages. Even though the surfaces may not suffer permanent damage, exposure to abnormal light intensity will markedly desensitize the surfaces.
- 6) Electrons released from the photomultiplier surfaces by the photoeffect by an ionizing particle.
- 7) Photoemission due to bombardment of the surfaces by ions in the residual gas.
- 8) Electrons released by means of a process as yet unidentified, by gamma radiation and neutrons of energy upwards of roughly one Mev. Some of the isolated pulses observed with 50 and 100 Mev neutrons are of great height, corresponding to the emission from the first surface of as many as 2000 electrons within 10^{-7} seconds.

High Gain Photomultiplier Operation

In the typical arrangement discussed above, the signal-to-noise ratio is about 5:1 below 120 volts per stage. The gain-per-stage characteristic is illustrated in Figure 4 which also indicates the region in which the signal-to-noise ratio begins to decrease. It was verified that this ratio was dependent only on

the voltage applied to the first two stages. Hence, by applying higher voltages (180 to 200 volts) to the seven last stages, it is possible to raise the total gain sufficiently (up to 25×10^6) to dispense with conventional vacuum tube pulse amplifiers. The pulse can then be observed directly as it appears at the photomultiplier output by connecting the tube to the deflection plates of an oscilloscope (preferably of the 5JP1 style).

The gamma pulses so observed range up to 20 volts in height and have a rise time of about .05 microsecond. This, then, is essentially the duration of emission of light by anthracene, being longer by a factor of 50 than the response time of the photomultiplier. The decay time of the anode coupling circuit is adjusted for minimum overall pulse duration compatible with minimum attenuation of pulse height. Variation of the decrement of this coupling circuit confirmed the above value for the rise time. The optimum decrement satisfying the above criterion appears to be such as to yield a total pulse duration of .15 microsecond.

Since the charge transferred through the last few photomultiplier stages is quite large in this mode of operation, it is necessary to by-pass those later stages at the socket with small mica capacitors. In addition certain precautions must be taken against breakdown, since the total voltage between pins 10 and 11 (anode and cathode, respectively) can be as high as 1800 volts. A slot is cut halfway across the base and socket between these pins, and a small sheet of 1/32 inch polystyrene is inserted snugly into that slot. The locating pin is cut open and the tube base filled with ceresin wax to prevent base and glass seal leakage. The socket and circuit elements are likewise coated with ceresin wax. The amplification of the pulse by the photomultiplier to a height adequate for "interpretation" by succeeding discriminators, scalars and so forth, has, besides the obvious advantage of simplification, the further advantage of eliminating the need for careful shielding and circuit isolation. In addition, for certain specialized applications it makes possible the construction of a complete coincidence counting circuit in a compact unit, as well as portable survey equipment of minimum complexity and very high counting efficiency.

Pre-amplifier

There is thus established the feasibility of supplying information concerning high energy ionizing particles in the form of negative pulses of the order of 20 volts in height and .1 microsecond in width that are fairly proportional to the amount of ionization produced. The "pre-amplifier" is no longer a true amplifier, but is rather an impedance matching device for the purpose of transmitting this information by coaxial cable. The purpose in transmitting the pulses in this way to remote interpreting equipment is to permit the observation of the pulses as they come from the individual photomultipliers, rather than from some distorting devices such as discriminators or coincidence mixers. More extensive use of electronics becomes possible in view of the length of the anthracene scintillation pulses, which are indeed much longer than anticipated.

The "pre-amplifier" must thus transmit the indicated pulses with essentially no distortion in any way, except perhaps in the matter of polarity. In order to accomplish this successfully over lines of 100 ohm impedance, cathode followers (2 parallel 6AG7's) with high peak plate currents will be employed. In order to provide the positive grid driving voltage, a unity gain phase inverter stage (1-6AG7) is to be used. The addition of this stage will provide a comparatively low impedance signal source, which makes it possible to drive the cathode followers to as much as 2 amps peak current. Both phase converter and cathode follower are heavily degenerative, to the extent of reducing the effective tube input capacitance to only a few micromicrofarads. It is expected that this line-matching unit will thus add a delay of at most .02 microsecond to the photomultiplier output pulses.

Procurement

It is generally felt that, from the long-range viewpoint, the problem of anthracene crystallization in quantity could best be served by a commercial organization experienced in the matter of crystallizing widely different substances. Accordingly contacts have been initiated through the Materials and Information Branch of the Atomic Energy Commission to handle this problem.

Inasmuch as the 1P21 photomultiplier will be operated well in excess of its ratings, it is also felt that a specially designed 11-stage photomultiplier with the features listed below would be the ideal light detector.

- 1) Head-on exposure of photosensitive surface (this indicates in-line geometry).
- 2) Same photosurface as on the 1P21 (i.e., blue sensitive).
- 3) Operable in moderate magnetic fields (up to a few hundred gauss).

The mechanical design of the "pre-amplifier" units takes into account their possible modification for use with such a tube. Steps have been taken to insure early development of such a tube by a competent commercial group. Inquiries concerning these two items should be referred to the agency designated above.

The Counting Equipment

The arrangement of the complete "counting unit" using anthracene scintillation counters is shown in Figure 5. The use of coincidence light pulse detectors with either single or double crystals should eliminate the background due to the various photomultiplier-generated noise pulses. The discriminator-coincidence unit is presently in the design stage. It is noted that coincidence mixing will be done directly with the discriminated and clipped pulses. It is felt that a generated, gated pulse would not be narrower than the scintillation pulse, and that the latter is of sufficient uniformity to permit its direct use in such a circuit.

It appears that VR tube regulation will be adequate for the various voltages required, provided that a fair degree of line regulation exists (as might be provided by "sola" type regulators). Regulated supplies are to be used initially, however. The adjustment of individual photomultipliers will be accomplished by individual variable series resistances and appropriate metering.

Applications

The number of applications are legion; the scintillation counter is applicable to all very high energy scattering and absorption experiments for which conventional gas chambers are used. It becomes particularly advantageous because of its high density in the energy region of minimum ionization for heavy particles.

Because of the high hydrogen content of anthracene, it makes an effective neutron counter, being its own proton generator. An anthracene crystal shows visible fluorescence in a high intensity, high energy neutron beam such as that generated by the 184-inch cyclotron. Another interesting application in connection with detection of fast neutrons is the use of the carbon $n-2n$ induced 20 minute beta activity as an integrating detector for a neutron flux of energy greater than the 20 Mev threshold. Such work is presently accomplished by counting the surface beta activity of carbon discs. By counting the volume activity induced in the anthracene by its fluorescence, a considerable gain in sensitivity is possible.

The anthracene scintillation counter also makes possible a portable radiation survey instrument including some form of discrimination control for identifying the ionization density of the radiation as well as its presence. It has the advantage of having an organic composition and density something like that of living organisms.

In connection with its very high efficiency for gamma ray counting, these radiation detectors may make handy probes for biological work, using Lucite light conducting rods with the anthracene crystal cemented at the end of the probe.

These applications seem the most promising of the many which have been suggested by various members of the laboratory staff and do not include variations of the several uses appearing in the literature.

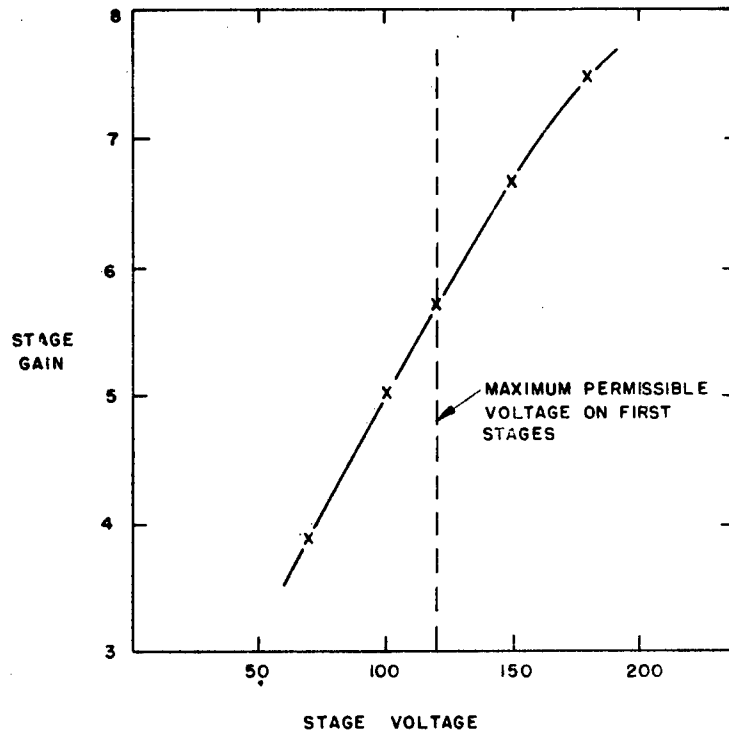


Figure 4. Voltage - gain characteristic 1P21 photomultiplier.

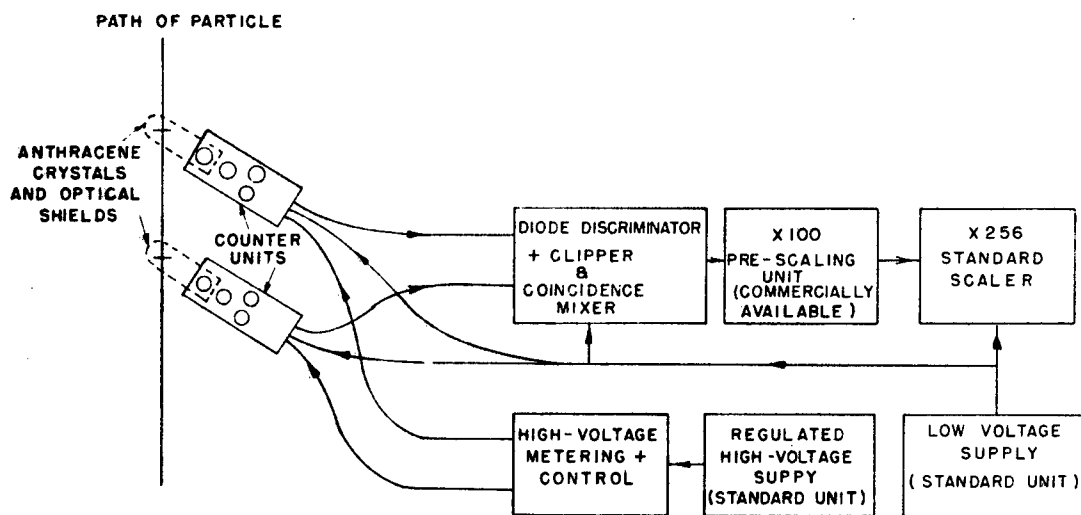


Figure 5.

Acknowledgment should be made to W. Stephan, H. Powell, and A. Ghiorso who were instrumental in developing techniques for crystallizing both anthracene and naphthalene.

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