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INSTRUMENT RESEARCH AND DEVELOPMENT DIVISION
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PHOTOMULTIPLIERS (R. K. Swank and J. S. Moenich)

The procedure for measuring the signal to noise ratio of photomultipliers has been improved. In the procedure previously described (ANL-5068) it was necessary to turn off the photomultiplier voltage and expose the tube to light while the scintillation pulse source (the "standard" signal) was removed. In the new pulse source assembly a shutter is provided. In this way the test source may be introduced or removed at any time without exposure to room light. This new device makes possible much greater accuracy in noise measurements as a result of the fact that exposure to light (even though the high voltage is first removed) causes marked changes in the dark pulse distribution of a photomultiplier. The exact source of this hysteresis is not known. But it has been observed that about one half hour is required for the background of an RCA type 5819 photomultiplier to reduce to a stable value. A much longer time is required in the case of a DuMont type 6292 photomultiplier. Furthermore, as will be shown below, there appears to be an even longer term drift in the dark pulse rate which may involve storing the tube for several days in the dark before final equilibrium is obtained. However, after one half hour of operation, the dark pulse rate of the type 5819 photomultiplier is sufficiently low so that no significant change is observed during the subsequent two hours. It is under these conditions that most of the data presented here was taken.

The photomultiplier to be tested is placed in a light-tight assembly in which the pulse source and shutter are mounted. The glass bulb is well insulated and guard ringed so that a high voltage stress across the cathode end of the bulb is avoided. The voltage divider is designed so that the voltage between cathode and first dynode as well as the voltage between the last dynode and anode may be varied independently of the multiplier voltages. A glass diffusing disc is used to distribute the light over the surface of the multiplier cathode and simulate the optical conditions present in an actual scintillation counter.

As previously stated, the "signal to noise" ratio data given here is the integral noise counting rate measured at various pulse amplitude levels, these levels being expressed in terms of the energy needed by a fast electron in NaI(Tl) to produce an equivalent average pulse size. This standardization is carried out with our very best NaI(Tl) crystal, 1-1/4 inch diameter by 1/2 inch thick, canned with an MgO powder reflector and placed in optical contact with the photomultiplier, using a minimum of intervening optical material. It is thus not possible for workers in other laboratories to

duplicate our results exactly, but an agreement of $\pm 20\%$ does not seem unlikely. As will be shown, much larger variations can be observed with various photomultipliers or with a given photomultiplier under various conditions.

The first investigation deals with the effect of the anode voltage on signal to noise ratio. The results for three tubes are given in Figures 1, 2, and 3. All tubes were operated at a voltage to give the same output pulse size (approximately 25 millivolts for the 36 Kev calibrating signal). Three other tubes investigated gave similar data. It is interesting to note that the behavior of the noise counting rate is different at different pulse height levels. In nearly all cases a minimum occurs in the low energy (i.e., small pulse height) counting rate in the vicinity of 30 - 50 volts on the anode. However, at higher pulse "energies" the noise counting rate may increase or decrease with anode voltage. The whole effect is small, however, except for low energies and very low or very high anode voltages.

An investigation was also made of the dependence of signal to noise ratio on the voltage between cathode and first dynode. The results for the same above mentioned three tubes are given in Figures 4, 5, and 6. This parameter appears to be more closely related to the noise phenomenon, since a strong dependence of the noise level on it is observed in every case. Minimum noise is observed at approximately 200 volts for low pulse energies. At higher pulse energies the minimum is much less pronounced and occurs at a higher cathode to first dynode voltage. It is significant that the voltage between cathode and first dynode for optimum signal to noise ratio is lower than that required for maximum pulse height resolution. This suggests that a scintillation counter which is to be used at one time for spectrometry and at another time for low energy counting should have a switch incorporated for changing the value of the cathode resistor. A value of 2 - 3 megohms is desirable for low energy work whereas 5 - 8 megohms is required for spectrometry (assuming dynode resistances are 1 megohm each).

A comparison of Figure 1 and Figure 4 shows significantly better performance in the latter figure even when conditions are identical with those in Figure 1. This discrepancy seems to be the result of a long term hysteresis resulting from the fact that the data for Figure 4 were taken the day following the taking of data for Figure 1. In the intervening period the tube was left in the dark. In the case of Figure 2 and Figure 5, where the data were taken the same day, no such discrepancy is observed. In the case of Figure 3 and Figure 6 the two tests were run in reverse order with an overnight dark period intervening; in this case the discrepancy is reversed. Attempts will be made in future experiments to follow this long term hysteresis in more detail.

A few preliminary measurements made on the Du Mont type 6292 photomultipliers indicate signal to noise ratios comparable to the RCA type 5819 when the shield plate is at first dynode potential (this connection is made internally in the 5819). However, when the shield plate is made negative with respect to the first dynode, the noise level increases markedly.

One Du Mont tube under test has shown a steady decrease in noise counting rate for three days in the dark, so that the final signal to noise ratio may not have been reached in the tubes of this type examined so far.

A photomultiplier with lithium antimonide cathode and beryllium-copper dynodes has been received from Switzerland. It will be examined for noise, transit time spread, and pulse height resolution.

SCINTILLATION COUNTER TECHNIQUES (R. K. Swank, J. S. Moenich)

During this quarter 9 light-tight photomultiplier assemblies were constructed, and 3 sodium iodide crystals of different sizes were canned.

One of the crystals canned was the first of 16 to be used in a Dumond-type gamma ray spectrometer under construction at CP-5 by the Physics Division. The crystal, a 2-1/4 inch cube, is canned in a cubical can of such size as to allow sufficient space for MgO to be packed around all sides, leaving only a circular hole in one face where the photomultiplier and light piper are inserted. Optical contact is made with Dow Corning DC-200 fluid, which also acts as a hermetic seal at the tube opening. A secondary seal and retainer for the fluid is made of EC-801 rubber sealer (Minnesota Mining and Mfg. Co.). At the crystal face opposite the photomultiplier, the can is fitted with a tight lid sealed with R-313 (Carl A. Biggs Co.), after which a second seal is made with thin sheet aluminum and EC-801 sealer. The resulting assembly can pass through a rectangular opening only slightly larger than the actual crystal size. In this way a battery of such assemblies can be stacked to present a large detection area.

SCINTILLATION DECAY TIMES (R. K. Swank)

A program was initiated last month for the investigation of scintillation decay times of organic scintillators. So far all measurements have been confined to a small group of anthracene crystals. A measurement by Kelley and Goodrich¹ for anthracene is believed to be reliable. They give 35 mμs at 25° C for the decay time and a temperature dependence which is approximately directly proportional to the absolute temperature. The three samples under study at the present time have shown decay times* of 32.4, 34.7 and 38.0 mμs. The last value is unexpected since there is no mechanism known by which the decay time can be longer than the decay time of a pure sample. The presence of quenching impurities will reduce the decay time and the fluorescence yield by the same

¹G. G. Kelley and M. Goodrich (ORNL) Phys. Rev. 77 138 (1950).

*Although the measurements were made in a range from 28°C to 32°C the values given are corrected to 25°C from Kelley's curve of decay time vs. temperature.

amount. Although the above results suggest that Kelley's sample may not have been absolutely free of quenching impurities, pulse height comparisons with anthracene samples made at Oak Ridge National Laboratory indicate that their anthracene is of very high quality indeed. However, differences of the order of 10% in impurity quenching might be offset by differences in light absorption. The latter would affect the pulse size but not the decay time.

Although the above decay time values are averages of many determinations, the reliability cannot be precisely stated at this time. A careful analysis of the experimental procedure indicates that a precision of approximately $\pm 1\%$ is obtainable. Most of the determinations do in fact lie within 1% of the average value. However, for causes as yet unknown, an occasional value has been obtained which differs from the average by as much as 10%. The only plausible explanation so far advanced is that a shift in oscilloscope sweep calibration may have occurred between the time that the scintillation pulse was photographed and the time the calibration was photographed. In the determinations in which large errors occurred, only one time calibration was used for as many as 6 scintillation exposures. The present procedure calls for a time calibration to be made immediately after each scintillation exposure.

The time calibration has been established by driving the oscilloscope with a pulse fed into an air dielectric coaxial line, one end of which is shorted and the other end open. Although a single pulse is fed into the line, a series of echos is displayed on the oscilloscope resulting from repeated reflections from the open and shorted ends. The spacing between pips is twice the transit time of the line. The end effects are corrected by measuring two identical lines of different lengths. The lines have also been compared with a calibrated sine wave oscillator.

PLASTIC SCINTILLATORS (W. L. Buck, L. J. Basile, R. K. Swank)

In our report for the preceeding quarter² it was mentioned that a plastic scintillator consisting of polyvinyltoluene containing 4% p-terphenyl and 0.02% 1,1,4,4-tetraphenylbutadiene was found to show unusually high transparency for the light of its own scintillations. To verify this observation the following experiment was performed. A plastic scintillator of the above composition was prepared in the form of a long, highly-polished, straight rod. Scintillations were produced at or near one end of this rod by a beta emitter placed adjacent to this end. A fraction of the light from these scintillations was "piped" down the rod by total internal reflection; the remainder was absorbed by a band of dull black optical paint applied to the surface of the rod for a suitable distance from the source end. The

²ANL-5068, Argonne National Laboratory, Instrument Research and Development Division, Quarterly Progress Report, March - May, 1953.

amount of light reaching the opposite end of the rod was measured with an RCA-5819 photomultiplier. After each such measurement the rod was shortened by cutting off a section from the end opposite the beta emitter, and the light measurement was then repeated, thereby determining the manner in which the intensity of the "piped" light decreased along the length of the rod due to re-absorption by the material of the scintillator.

As expected from the theory, a plot of the logarithm of the light intensity vs. the length of the rod gave a curve which approached a straight line for large lengths. From the slope of this straight-line portion of the curve it was determined that the effective absorption coefficient of this polyvinyltoluene scintillator for the light of its own scintillations was about 0.005 cm^{-1} (referred to the base e). By comparison, stilbene crystal is reported³ to have an effective absorption coefficient of 0.20 cm^{-1} . This result further emphasizes the suitability of the plastic scintillator for applications requiring large volumes of thicknesses.

A plastic scintillator of the above composition, in the form of a large disc weighing approximately one pound, has been furnished to Dr. F. N. Hayes of Los Alamos Scientific Laboratory for use in a proposed experiment of mutual interest to LASL and ANL.

It was found that substitution of about 0.1% p,p' -diphenylstilbene in place of the tetraphenylbutadiene as a spectrum-shifter in the above polyvinyltoluene composition results in a plastic scintillator with about 5% greater pulse height and an appreciably shorter decay time. The diphenylstilbene was obtained from Pilot Chemicals, Inc.

Another new organic fluor, 2-(1-naphthyl)-5-phenyloxazole, obtained through the courtesy of Dr. F. N. Hayes, was found to be slightly inferior to tetraphenylbutadiene or diphenylstilbene for use as a spectrum-shifter.

Plastic scintillators consisting of polyvinylbiphenyl and of polycyclohexylstyrene, and of copolymers of these materials with vinyltoluene, activated with p -terphenyl and/or tetraphenylbutadiene, were found to give scintillation pulse heights somewhat smaller than those obtained with similar polyvinyltoluene scintillators. Since the purity of the vinylbiphenyl and cyclohexylstyrene monomers used for these samples was considered questionable, efforts were later made to effect additional purification of these monomers. The vinylbiphenyl monomer, a silvery crystalline solid, was purified by sublimation under high vacuum. In the case of cyclohexylstyrene monomer, a colorless, oily, high-boiling liquid, distillation at very low pressure was employed, considerable loss of product due to polymerization being experienced. Several additional samples of polyvinylbiphenyl and polycyclohexylstyrene scintillator, made from these more highly purified monomers, are in preparation or awaiting test.

³E. C. Farmer and I. A. Bernstein, Nucleonics 10, No. 2 54 (1952).

Efforts in the past have been directed toward preparing plastic scintillators for beta, gamma, and fast neutron detection. It would be desirable if a plastic scintillator for the detection of thermal and epithermal neutrons would be developed. Thus it was decided to incorporate decaborane, $B_{10}H_{14}$, in a polyvinyltoluene scintillator and determine its performance as a scintillator counter.

Preliminary studies showed that decaborane dissolved readily in the vinyltoluene monomer to the extent of 5% by weight. However, the resulting solution soon developed a reddish-brown color, this effect being accompanied by the evolution of heat and a gas of unknown composition. Such a reaction could have been due to impurities in the vinyltoluene or the decaborane, amongst other reasons.

To eliminate the possibility of an impurity in the vinyltoluene some monomer was purified in the following manner. A quantity of triple distilled vinyltoluene was redistilled twice over Drierite at low pressure, 5×10^{-5} mm Hg, in hopes of further outgassing and drying the monomer. The product was then passed through an activated alumina adsorption column and finally distilled from a greaseless system directly into vials containing decaborane. The vials were then sealed off under high vacuum. However, these samples reacted in much the same manner as described previously which is interpreted as a strong indication that this effect is not due to any removable impurity in the vinyltoluene.

A third sample was prepared containing purified decaborane. Some decaborane was sublimed, under high vacuum, into a vial and some pure vinyltoluene distilled into the same tube. Again the vial was sealed off under high vacuum and slowly allowed to warm up. At 0°C the decaborane readily went into solution with no discoloration appearing. The vial was then placed into a furnace and the temperature slowly raised in order to observe at what temperature the discoloration would appear. Up to 130°C no discoloration appeared in the solution. Proceeding ahead with polymerization, after a period of 36 hours, the solution did discolor with only a slight amount of polymerization taking place. It seems that the decaborane is reacting with the vinyltoluene at this higher temperature.

PHOTOELECTRIC COMPARATOR HEAD (H. Bruce Phillips)

The optical parts for the comparator head were completed and assembled. Some rough experiments were done in order to test the behavior of the system. The signal from the detector was displayed on an oscilloscope as the head was moved across a photographic line. A deviation of 10 microns was visually discernible, and with a synchronous detector this should be reduced to 5 microns or less. As a further improvement, a slit of adjustable length was installed in front of the beam splitter.

One complication has arisen. When the unit was being tested, it was found that the driving motor fell out of synchronism with the 60 cycle line voltage. This meant that the line voltage could not be used to synchronize the detector, and therefore another means of obtaining a reference signal must be employed. A commutator was installed, but the brush noise and wear of the copper commutator was excessive. A hardened, polished commutator is to be made. In the meantime other possible ways of obtaining reference signals are being investigated.

INSULATORS (Raymond J. Munick)

Measurements of electric currents through lucite were made with a vibrating reed electrometer. A modified dynamic capacitor apparatus to measure high resistance was built and given preliminary tests.

The subjects of the current measurements were three disks of Lucite, each 3 inches in diameter and $1/8$ inch thick, on the flat faces of which were evaporated circular electrode and guard rings. To prevent surface leakage across a guard ring gap, the small volume enclosed around it was connected to a dessicator containing phosphorus pentoxide during the measurements.

Specimen IV, on which measurements are in progress, is kept in an oil bath, which maintains a temperature of $25^{\circ}\text{C} \pm 0.2^{\circ}$. Specimens II and III were maintained at room temperature, nominally 75°F in an air-conditioned laboratory, which was found to vary as much as 2 or 3 degrees Centigrade during the day. The changes in room temperature appear to affect the converter unit of the electrometer, producing a background current that rises occasionally as high as 10^{-15} amperes.

The quantity that is measured directly is the voltage drop that is produced across a 5×10^{11} ohm Victoreen resistor connected in series with the insulator specimen. Under these conditions a voltage that is applied to the electrometer input decays exponentially with a time constant of about 16 seconds. Currents (or the equivalent voltage drops) are recorded as a function of time on an Esterline-Angus recorder.

The electrometer input is short-circuited when the voltage across the specimen is altered. After from 10 to 30 seconds the shorting key is opened. The voltage drop across the current resistor rises to a maximum in a few seconds and then decays over a period of several hours to either a steady current or to background. The transient part of the current, which is sometimes referred to as the absorption or electrification current, will here be called the polarization current, since phenomenologically it corresponds to the time rate of change of the electric polarization of the dielectric.

On a graph of the logarithm of the current versus the logarithm of the time a typical plot for either specimen II or III of the polarization current data for from 60 to 10,000 seconds is linear. For a given specimen the charging

current is proportional to the voltage, and changes of current obey the superposition principle with respect to changes in voltage. These results agree with those of earlier investigators on other materials.

The apparatus was arranged in such a way that weights of 15 and 26 pounds (lead bricks) could be added to the low impedance sides of specimens II and III. The addition of either weight to specimen II produced a positive current of about 2×10^{-16} amperes, which decayed to zero in about half an hour. Removing the weight produced a similar negative current. No effect was noticed with specimen III, but the background was so much worse that to be noticeable any change would have had to be greater than 5×10^{-16} amperes. A simple calculation shows that the forces of these weights are enormous compared to the electrostrictive forces of the voltages used.

Volume resistivities of 5×10^{17} ohm centimeters have been obtained for specimens III and IV; for specimen II a value of 5×10^{18} was obtained before the weights were put on, but one month after the removal of the weights the value was about 10^{18} . These measurements for specimen II were taken between the times of those for III and IV. Although the volume resistivities for specimens II and III differed so greatly, the ratio of the polarization current of III to that of II both before and after the weights were applied was less than two.

The modified dynamic capacitor apparatus differs from that described in the last quarterly report in that the variable capacitance and contacts to the insulated electrode are operated manually and the signal is coupled by an additional separate electrode to an electrometer. The first tests indicate too much leakage conductance along the connection to the electrometer. Steps are being taken to eliminate the leakage.

ELECTRON MOBILITY (J. C. Bowe)

Measurements of electron drift velocity have not been started due to difficulties encountered in locating small leaks in the gas purifying chamber. Leaks which could not be located by immersing the chamber under water were readily found with the aid of a helium leak detector. No success was obtained in forming a leak free joint, using the low melting point silver alloy solder (50% Ag, 15-1/2% Cu, 16-1/2% Zn, 18% Cd). A new chamber has been constructed of stainless steel and soldered with 60% Ag, 25% Cu, 15% Sn alloy. The entire system has been checked with the helium leak detector and is now leak free.

The brass cathode has been replaced with a tin coated plate glass disk. Tin was found to be a more efficient electron emitter than brass.

Negotiations for the procurement of two new light sources have been completed with the Kemlite Laboratory. An attempt is being made to produce a more concentrated and brighter source of light.

FLUORESCENCE PHOTOMETER (F. J. Lynch)

A second fluorescence photometer has been delivered to the Industrial Hygiene and Safety Division. Several changes were made. A phase shifting network, which permits phase adjustment with little change in amplitude, was added to the standard signal amplifier. An uncoated AH5 lamp and an ellipsoidal mirror have been substituted for the aluminized lamp and spherical reflector with a resulting twofold increase in optical efficiency.

The photometer is independent of lamp intensity over a large range, showing no change in reading when the intensity is increased 60% by changing the lamp input voltage from 100 to 130 V. However, a long term drift has been troublesome.

Because of geometry, changes in arc shape and intensity affect both samples alike; changes in spectrum, however produce a change in excitation of one sample relative to the other due to the transmission and reflection characteristics of the dichroic mirror. It is believed that the blower used to protect the filter cools the mercury arc lamp to an extent that all the mercury is not vaporized. Deflecting part of the air from the blower away from the lamp onto the filter reduces the drift. It is expected that a further reduction in the lamp ventilation will virtually eliminate the drift.

The over-all optical efficiency is such that approximately 10^{-7} amp is produced in the 929 phototube by the fluorescent light from a $1.0 \mu\text{g}$ uranium sample. The signal to noise ratio is such that on the most sensitive scale the readings fluctuate less than 0.2% of full scale (equivalent to about 2×10^{-12} g of uranium). The background fluorescence of the instrument is equivalent to about 1.5×10^{-10} g of uranium. This, of course, can be balanced out with the zero adjustment. The disparity between readings on different scales is less than 0.2%.

The shop work has been completed on the remaining four instruments. The electronic circuits are nearly finished on one instrument. Further work is being done on improvement of the ultraviolet source. A larger ellipsoidal reflector has been made by the shop for use with the AH5 lamp. The 100 watt sealed beam mercury arc spotlight, type CH4, has been tested in the second instrument and found to be as efficient as the AH5 and small ellipsoidal reflector. Further comparisons will be made on the third instrument in regard to uniformity of illumination and stability. Although the rated life of the CH4 is only one third that of the AH5, it does not require an auxiliary reflector and uses only 40% of the power; it will be incorporated if it is found to have no other disadvantages.

ROTATING CONDENSER ELECTROMETER

The condenser plates of the rotating condenser with internal drive were electrolytically plated with gold. Although the contact potential is reduced and is more uniform, additional gold will be evaporated on the plated gold to improve it further.

Copper plates have been fabricated to conduct a series of tests on contact potential of various surfaces. The plates will be electrolytically coated with different metals; where feasible the plates will also be coated by evaporation with the same metal.

An investigation of bearings suitable for the rotating condenser has been started. It is required that the bearings exhibit low friction without organic lubricants and show good electrical conductivity with low electrical contact noise. Three bearings are being considered: high speed tool steel ball bearings lubricated with barium (such as is used in the rotating anode X-ray tube), a permanent magnet suspension (such as is used in the new G.E. watt hour meter), and synthetic sapphire bearings with electrical contact provided by liquid gallium.