RADIATION COUNTING EQUIPMENT

AND ITS USE IN

VAPOR PHASE COUNTING OF RADIOSULFUR

by

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degree of Doctor of Philosophy

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INTRODUCTION

The increase in availability of radioisotopes has led to their wide use as tracers in chemistry and other scientific fields. Consequently methods for detecting and measuring radioactivity have become increasingly important.

Although many techniques are available, tracers are very generally measured by means of an electronic circuit actuated by a radiation-sensitive detector responsive to individual particles or photons. The most widely used combination is the scaling circuit and Geiger tube.

While the commercially available scalers have been improved since this work was started, they are still not entirely satisfactory with respect to stability and ease of maintenance. This is particularly applicable to the decade scalers, which subdivide the count rate (to a value within the range of mechanical registers) by factors of ten, and are more convenient to use than binary scalers which subdivide by factors of two.

An inherent defect of Geiger tubes is their inefficiency in responding to radiation of low energy. This arises from the fact that the thinnest window strong enough to withstand the pressure differential (about 650 mm) stops a large fraction of low energy incident radiation such as that from radiocarbon and radiosulfur. While other detectors can be used, there are advantages in the use of Geiger tubes.

The difficulty has been surmounted in some cases by introduction of the radioactive substance as a gas into the

Geiger tube. This technique has been used with tritium (as H_2 and H_2O) and radiocarbon (as CO_2). These are both beta emitters, and it is desirable to apply this method to radiosulfur, another weak beta emitter. The major difficulty in developing methods for gas phase counting with Geiger tubes is due to the fact that few substances can be introduced into the tubes without at least partially destroying their useful characteristics.

OBJECTS OF THE INVESTIGATION

The primary objects of the investigation were:

1. To design and construct an improved scaler;
2. To develop a technique for gas phase counting of radiosulfur with Geiger tubes.

RESULTS OBTAINED

A Wynn-Williams thyratron scale-of-eight was constructed for use in the preliminary stages of the investigation.

A wide range amplifier was built for use with a synchroscope in the study of pulses produced by experimental Geiger tubes.

A new type of radiation counter, based on N-scale rings, was developed. Three instruments of this type were built, one a scale-of-twenty-five (two rings of five stages each), and two decade scales-of-one hundred each comprising two rings of ten. These instruments are outstanding in stability and reliability.

In developing a technique for gas phase counting of radiosulfur it was found that methyl and ethyl sulfides are as good quenching agents in Geiger tubes as alcohols and ethers. Consequently it would be possible to introduce radiosulfur into the Geiger tube as alkyl sulfide without adversely affecting the operation of the tube, since the radiosulfur-carrier would also serve as the quenching agent. Less complex compounds, including CS_2, SO_2, and H_2S showed little promise, although H_2S might be used with careful control of the operating conditions.

A standard Geiger tube pattern was developed for use in gas phase counting. The design finally adopted included an electrolytically polished stainless steel cathode. Reproducible results could be obtained with tubes of this type on repeated filling and evacuation.

It was also established that erratic behavior of Geiger tubes can be caused by a trace of oil or grease whose source may be an oil diffusion pump or a greased stopcock. This accounts for lack of reproducibility in results obtained in the early stages of this investigation.

SUMMARY AND CONCLUSIONS

An initial objective of this work was to construct radiation counting instruments for use in the radiochemistry laboratory and in subsequent stages of the investigation. The most important instrument required is the scaling circuit or scaler.

The scaler is an assembly providing the following: the high voltage needed to polarize the Geiger tube; an amplifier to increase the size of small output voltages of the tube; a circuit to subdivide the count, by a known factor, to a slow rate; some sort of electromechanical register to totalize the disintegrations detected; and power for all the components, most of which are electronic.

Results of experiments with the commonly used Eccles-Jordan circuit, in which vacuum tubes are used, were not too promising. Apparently careful matching of components is necessary with this circuit.

The nature of the Wynn-Williams thyratron circuit is such that little difficulty was to be expected in its construction, and since a scaler was needed for the other work contemplated, a scale-of-eight of this type was built. This, with its associated input and output circuit and power supply, proved entirely satisfactory in the exploratory work under way at the time. It was also used for a considerable period in the radiochemistry laboratory course, and in an investigation with radiophosphorus undertaken by a graduate student in zoology.

A scaling factor of eight is not large enough, however, for most purposes, and moreover, thyratron circuits

are inherently slow because of the deionization time of the rare gas present at low pressure in the tubes. Consequently it became necessary to build scalers of some other type.

A decade scaler of some sort appeared to be most desirable. While a binary scale-of-sixteen Eccles-Jordan circuit can be made to recycle at every tenth pulse, the results are not too satisfactory, and as has been mentioned, experiments with the Eccles-Jordan type circuit gave results which were not encouraging.

A search of the literature disclosed that the N-scale ring used in the "Eniac" computer has ideal characteristics for application in scaling circuits. For such use, it must be equipped with appropriate input and output circuits and a high voltage supply for polarizing the Geiger tube.

A counter of this type was designed, and a scale-of-twenty-five utilizing two rings-of-five was built. Since the performance of this counter was entirely satisfactory, two decade scales-of-one hundred were constructed.

These three scalers were used in all the rest of the work. They were found to be completely stable and reliable, even when operated constantly for days at a time, and equally well adapted for use with Veeder-Root and Cyclotron Specialties registers. While some scalers require selection and matching of tubes and other components, components of commercial tolerances are entirely satisfactory in these circuits. In fact, any tube in either ring in any of these instruments can be exchanged with any other, and the plate voltage can vary over a wide range, with no effect on performance.

A scale-of-one hundred is usually all that is needed

in radiochemical work. However, if a higher scaling factor is required, two counters of the type mentioned can easily be connected in series by a simple circuit. Thus, for example, the two one hundred-scalers were operated in series to give a scaling factor of 10,000.

A synchroscope was used in the study of Geiger tube pulses. A succession of similar pulses produced at a satistical rate can be displayed as a single pattern on the screen of this instrument. The pulses, however, are not large enough to actuate the synchroscope circuit, and it was therefore necessary to construct a wide range amplifier to increase the size of the pulses without distortion.

In early experiments with Geiger tubes with standard fillings, it was found that repeated evacuations and fillings caused a progressive and rapid deterioration in performance, i.e., the slope increased and the plateau length decreased. Results with experimental fillings would obviously be without significance unless this change could be prevented.

Several variables were investigated without resulting in a solution of the problem. Tubes were filled with various mixtures at various pressures. Cathodes of several materials were tried, and some of these were given several kinds of surface treatment.

After these and other variations were tried, it was suspected that the source of the difficulty was contamination of the tube by oil or grease. A new vacuum system was built to eliminate the contamination. The oil diffusion pump was

replaced with a mercury pump, and the stopcocks were replaced with packless brass valves. With this system, Geiger tubes were repeatedly evacuated and filled with the standard argon-alcohol mixture with reproducible results.

To demonstrate conclusively that grease was the cause of the trouble, Geiger tubes of a special design were used. Each of these was equipped with a side arm containing a sample of pump oil or stopcock grease, with a break-off tip between the sample and the Geiger tube. The tube was filled with the standard mixture and its plateau was determined. Next the break-off tip was broken, by means of a slug enclosed in the side arm, to permit entry of vapor from the sample into the Geiger tube. Subsequent testing of the tubes showed the deterioration of characteristics previously observed with tubes filled on the original vacuum system.

Since it was now possible to make Geiger tubes with consistently good characteristics, experiments with various sulfur compounds were undertaken. The objective was to find a compound which could be introduced as a gas or vapor into the tube without seriously affecting its operation, and of a material which could conceivably be quantitatively prepared from almost any sulfur compound.

The latter criterion led to a consideration of simple inorganic compounds, namely H_2S and SO_2. These, however, adversely affect the characteristics of the tube, in the sense of increasing the slope and decreasing the

length of its pleateau. The effect is less with H_2S than with SO_2; if the quantity were limited and the voltage on the tube were carefully controlled, H_2S might be useful in gas-phase counting. Carbon disulfide also gave poor results and thionyl chloride was too corrosive to the metal parts of the system.

Next the alkyl sulfides were considered. These are the sulfur analogues of the ethers which are commonly used quenching agents. On the basis of the analogy, and the fact that the alkyl sulfides have ultraviolet absorption in the range required of the quenching agent, these compounds seemed logical choices.

Tubes filled with argon and either methyl or ethyl sulfide had operating characteristics substantially identical with those of standard tubes in which alcohol or ether is used as the quenching agent. It was not difficult to make tubes utilizing the sulfides as quenching agents with the following characteristics: starting potential, 1000 volts; length of plateau 200 volts; slope of plateau, 3% per hundred volts. This contrasts sharply with the situation existing with gas-phase counting of radiocarbon in which special precautions must be taken to counteract the deterioration of tube characteristics caused by the carrier, CO_2.

In the course of this work, it was found that cathodes made of stainless steel tubing are very satisfactory. Characteristics of Geiger tubes with such cathodes

are markedly affected by the nature of the inner surface of the cathode. While quite satisfactory results can be obtained with cathodes finished by turning in the lathe, plateaus are longer and flatter when the cathodes thus treated are electrolytically polished.

In conclusion, it may be stated that the problem of gas-phase counting of radiosulfur has been solved to the following extent: the techniques of making and filling Geiger tubes for this purpose have been developed; it has been shown that sulfur may be introduced into the Geiger tube as methyl or ethyl sulfide. The next steps in the solution would involve preparation of the alkyl sulfide tagged with radiosulfur, testing of Geiger tubes containing the tagged sulfide, and finally the development of a standard procedure, as universal as possible, for quantitatively obtaining the sulfur in sulfur compounds as alkyl sulfide. Such a procedure should be much more satisfactory than those used with radiocarbon and tritium, since a quenching circuit and close high voltage control are not required with Geiger tubes containing alkyl sulfides.

RADIATION DETECTION AND MEASUREMENT

Many devices have been used to detect radiation. When radioactivity was discovered, only the visual observations of the effects of radiation on a sensitive material were used. This was due to the more limited knowledge existing in many fields at that time. These original methods are tedious and time consuming. They are still used, however, but only in those special cases where the attributes outweigh the disadvantages.

Simultaneously with the progress in electronic art, detectors yielding small voltage transients were developed and gradually the convenient and more highly sensitive electronic detectors displaced the older methods. The newer methods permitted each disintegration to be recorded, and also removed many of the human factors from the observations. In chemical tracer work, the most commonly desired data are assays of the radioactive material which is present. Electronic detectors are the most useful methods employed for these assays.

A brief summary of each detecting device will be given, although all the methods of detecting radioactivity possess some similarity, and, therefore, cannot be completely separated. The Geiger region, at present, is that system most widely used in detecting devices because it presents the least problems. It, therefore, was most widely used in the research and will be most widely discussed.

VISUAL METHODS

Photographic detection

Photographic methods were used by Becquerel in his discovery of radioactivity in uranium in 1896. This was based on the fact that all radioactive emminations will affect an unexposed photographic film to some extent and, upon development, will show up as a blackening of the film or, in some cases, will show the path of the particle or ray through the emulsion. Although this process is sensitive to small quantities of radioactive materials (1), there are disadvantages. The time that is required to make an exposure of the film to the radioactivity may be quite long (2), and in addition, time is required to develop and inspect the film. Another disadvantage is that many materials combine chemically with the silver halide of the film emulsion. This necessitates an interposed protective film between the emulsion and the subject being analyzed (3); thus, some of the sensitivity of the process is lost.

This method seems to be the best method when used in two ways; one, in experiments in which the material is stratified and the locations of the radioactive material is thus readily shown; and two, as an accumulative exposure recorder for the protection of personnel exposed to penetrating radiations (4). In the latter use, small film containers are worn by the people subjected to exposure and periodically developed under precise conditions.

The length of the photographic process and the slowness of the recording limit this method very greatly. Also, variations in density of the exposed emulsion, cause

considerable trouble (2). Therefore, this system is not so widely used as some of the electronic detectors.

Electroscopes and Electrometers

Electroscopes have been used to detect radiation almost as long as radioactivity has been known (5). An electroscope is a device that contains an easily deflected conductor, set so that the conductor will be repulsed by anyother part of the same conductor under the influence of an electric charge. Ionization of the air or other gas surrounding the conductor will cause leakage of the charge to occur. Due to the decrease in repulsion of the two portions of the conductor as the charge is reduced by the ionization, the rate of electrode closure is an indication of the quantity of radioactivity present. As the initial charge is very small, the conductor must be well insulated from other conductors. The effect of surface leakage on this insulator can be very troublesome at high humidities.

Electrometers also depend on electrostatic repulsion for their operation. These instruments detect small currents by their effect on a movable electrode in an electrostatic field. Their rotation in an electrostatic field is similar to that of the D'Arsonval or moving coil galvanometer in an electromagnetic field. Many electrometers use an optical lever in a manner similar to the galvanometer. Since electrostatic fields are required, additional sources of voltage are used in their operation. The currents that they measure are usually obtained from an ionization chamber containing the radioactive material and two electrodes charged

to a moderate potential. The current flow in the chamber due to the ionization also charges the electrometer.

These detecting devices have been put together in many ways (6). The conducting fiber quartz electroscopes are probably the most widely used. The small, pocket variations of this electroscope are widely used for personnel protection.

These detectors are sensitive; however they have disadvantages. Small quantities of radiation are detected only by long readings. Since they are sensitive to the quantity of ionization caused by the radiation, they show preferential sensitivity to some types of radiation. Readings are tedious to make, and this with the slowness and humidity problems make other methods easier to use. Electroscopes are most widely used under conditions that require measurements with limited laboratory facilities.

Cloud Chambers

The cloud chamber discovered by C. T. R. Wilson in 1911 has also been used to record the presence of radioactive materials. It has undergone many modifications through the years (8). The operation of this device is based upon the fact that the ions formed in the path of a radioactive ray or particle are excellent nuclei for the deposition of liquid droplets from a supersaturated vapor. The radioactive material thus creates a fog track in the supersaturated area. This can be photographed. The track

provides an indication of the path of the particle or ray.

The maintenance of gas in a supersaturated condition for continuous indications has proved to be a problem. Continuous readings are obtained by the maintenance of wide temperature differentials between two parts of the chamber. A vapor evaporated in the warmer end will, upon cooling, become supersaturated at some point in the chamber; thereby maintaining a sensitive area continuously between the two temperature extremes (9). Otherwise adiabatic expansion is used to cause the necessary supersaturation.

The use of this method as a quantitative detector would be rather difficult. It is best used in the detection of single rays. It has been used widely with various accelerating devices for nuclear transformations (10). Additional data can be obtained if the chamber is surrounded by an electromagnetic field so that the deflection of the particles can be seen (11).

The cloud chambers method is limited in its use because it is time consuming, and does not readily yield quantitative data. It has not been used to any extent in the evaluation of radioactive tracers. The main present use is in cosmic ray baloons.

Spinthariscopes

The activation of a fluorescent screen by radiation was discovered by Crookes in 1903 (12). If the type of radiation is kept constant, the light given off by the phosphor

is an indication of the activity of the sample. The individual scintillations can be counted in the case of the more energetic, easily absorbed radiation by observing a fluorescent screen through a microscope or lens. This device was used by Rutherford in the detection of the first artificial transmutation of atoms by alpha particle bombardment (13).

An observer must count each scintillation; hence, a reading is rather time consuming. The number of counts that can be accurately detected per unit of time limits the maximum amount of activity that can be detected. Spintharioscopes are at a serious disadvantage when compared with the other instruments now available. They are probably best used for qualitative, and semi-quantitative determinations under difficult conditions, such as those found in field assay work. For this function they are ideal because no batteries or other equipment are required. They also may be used to detect large quantities of contamination on laboratory equipment.

ELECTRICAL COUNTERS

Scintillation counters

The methods mentioned so far have been of the visual type; those to follow are of the electronic type. The detected radiation gives short voltage transients that are recorded by electrical instruments. The relationship between the visual and the electronic methods can be shown by the relationship of the scintillation counters to the spinthariscope. For a long time, the human eye was the only detector sensitive enough to detect the weak light from a bombarded phosphor. The eye is much more sensitive than a gas or high vacuum phototube (14). It might be possible to detect large amounts of radiation by the use of a gas phototube, but it would be very insensitive to small amounts of radiation. The discovery of the multiplier phototube (15) which can amplify (by factors of at least 10^5) the photo electrons caused by the incident light opened up the scintillation counters to electrical recording.

The variations that can be made in the nature of the phosphor used and the forms that it can take are many. A thin film of silver-activated zinc sulfide has proven to be satisfactory (16). The duration of scintillation by this material is rather long (17), and, if faster recording is desired, organic materials such as anthracene may be used. Materials such as anthracene allow counts to be recorded in less than one tenth of a microsecond (18), probably the fastest rate that is possible with any widely used detector.

In this method problems have arisen that make the system troublesome. In some cases the noise voltages that are started by stray electrons in the phototube can be as

large as the signal caused by the detection of radioactivity. This has been minimized by refrigeration of the phototube (19). When anthracene or other low emitters of light are used, pure materials in excellent crystal forms must be used to permit the maximum light to be incident on the phototube. Probably one of the most serious troubles is the hysteresis shown by the phototube at high count rates, resulting in lower sensitivity of the phototube (20).

The method is relatively new, and much work has been done to try to improve the technique. As better phosphors become available, and as newer and better phototubes are developed, the method may displace some of the techniques now used.

Crystal counters

Fluorescence is only one of the reactions of matter to bombardment by radiation. Van Heerden reported in 1945 that the conductivity of some crystals, when placed between electrodes maintained at a high potential differential and subsequently struck with radiation, was increased.

In crystal counters, the electrons that are liberated by the radiation are attracted toward the positive electrode, and, if not trapped in the crystal before collection at the electrode, will yield small voltage transients (21). The positive ions left are somewhat of a problem since they are fixed in the crystal lattice, and thus can exist in an unchanged state for some length of time.

Some materials that have proven to be satisfactory for crystal counting are diamonds, sulfur, cadmium sulfide, and silver halides. Due to the high potentials applied, however, silver halides and some of the other materials are

decomposed by electrolytic action if not refrigerated to liquid air temperatures.

At present, the counting system using crystals is little used. The low temperatures required is one disadvantage. The polarization of the crystal is a disadvantage in two ways. It causes many crystals to have a limited life in counting, and it also limits the maximum recorded count rate to a small fraction as compared to the permissable recorded count rates of other methods (22). If more work will produce a crystal that has fewer of the disadvantages found at present, the method may gain in favor. This has not as yet been accomplished (23).

Ionization chambers

The next three detectors to be described are similar in one respect. The majority in use take the form of a fine wire installed coaxially to a cylindrical outer electrode. The center wire is usually positive and is called the anode; the external conducting cylinder is negative and is called the cathode. The operation of the devices depends to a great extent upon the terminal voltage applied to the two electrodes. However, the mode of operation is different in each case.

An ionization chamber has moderate terminal potential. The passage of radiation through the gas in the chamber causes ion pairs to be formed. These consist of an electron and a positive ion. The voltage impressed upon the chamber

is such as to permit collection of the electrons formed by the anode. This, however, does not accelerate them enough to cause further electrons to be liberated by collision with other gas molecules (24). The electrons, due to their low mass, are quickly collected at the anode. The positive ions move slowly and thus do not contribute appreciably to the current flow. In this procedure, the current flows are not very large under ideal circumstances (25). Alpha particles, because of their high specific ionization, are easiest to detect by this method.

The individual counts that are recorded by this detector can be registered if they are amplified sufficiently. To get a voltage high enough to be amplified, the electrons must flow through a large resistor. The time required to charge and discharge the capacitance encountered with any chamber and auxillary circuitry through this resistor is so long that the maximum count rate with this instrument is seriously limited; one thousand counts per second is the fastest that can be accurately registered (26). The voltage transient from an energetic radioactive particle is very small, being less than 10^{-3} volt. Therefore, the amplification of the initial voltage to a size that will allow reliable triggering of stable circuits is a complex problem. These amplification circuits are not only slow, but also unstable due to noise interference. Therefore, individual counting techniques with ionization chambers are difficult to accomplish.

Chambers are widely used in monitor instruments that do not require the accuracy possible with the individual count method. An integrating device is used to totalize the current flow caused by several ionizing events (27). In this case, extremely sensitive electrometer vacuum tubes are used to amplify the currents flowing in the chamber, and semiquantitative data can be obtained by relatively simple equipment. Ionization chambers are of use because they not only detect the presence of radiation, but also permit the approximate evaluation of its effect on personnel (28).

Proportional counters

If the voltage between the electrodes of the counter is increased, the electrons in their travel toward the positive pole will acquire higher and higher velocities. Soon a point will be reached where the kinetic energy of the particle is high enough to yield two or more electrons of lower energy on collision with a gas molecule. The cascading of this phenomenon will result in a much larger flow of electrons at the anode than was started by the original ionization. The current has thus been amplified; the result being the same as that encountered in the gas phototube. The only difference between the two is the source of the original ionization. This is called the proportional region (29).

This method has been widely investigated recently.

It is capable of relatively fast counting rates (30); a count being recorded in less than one microsecond. It also permits the separation of radiation having different ionizing powers by a circuit that discards all pulses below a minimum size. It satisfactorily operates with the gas in the detector at atmospheric pressure; therefore, the sample to be assayed can be placed directly into the chamber of the detector (31).

In analyzing some samples, the range of the size of the pulses that are generated may be quite large. In many cases the range is so large that amplification of the transients becomes a problem, causing large pulses to overdrive the later stages of the amplifier. It has been estimated by other workers that a ratio of 10^5 to 10^6 in the size of transients detected would be needed for accurate counting. This range has not been obtained, but in a report by Baker (32), much more reliable results are obtained with amplifiers that will compress voltages that vary by a factor of 10^4 to a range that can be utilized by the subsequent circuitry. The use of gas at atmospheric pressures also causes the required terminal voltage to be relatively high, a factor that presents problems from the leakage standpoint and necessitates extreme precautions to prevent errors due to this cause (33).

As the problems of the amplification and other electrical troubles are surmounted, proportional counters will probably become more widely used. It is the fastest

of the counters based on gas ionization with subsequent electrical recording. It still is not so easily handled as the Geiger region, and therefore has not been so widely used. The flow type counters seem to be the universal type of proportional counters now in use; because of the flushing action of the gas they retain their characteristics well over a period of extended use.

Geiger tube detectors

The amount of amplification due to gas ionization increases as the terminal voltage of the detector is raised. Over a range in terminal voltage the ratio between the number of electrons caused by two events of different ionizing power remains constant. As the terminal potential increases farther, the quantity of electrons collected by the center wire tends to approach the same value--those of limited ionizing power being amplified more by the electron multiplication process than those of higher ionizing power. Finally, a voltage is reached that yields the same voltage transient no matter what the cause of the original ionization. This is the Geiger threshold voltage.

The operation of the tube at this voltage is somewhat more complicated than in the two previous cases (34 - 39). The ionization of the tube is not limited to the area of the original ionization, but the whole tube can be ionized if no barrier is placed on the central wire (40). If a simple gas is used in the tube, such as air or argon, once

a discharge has ionized the tube, the tube will stay in a state of ionization as long as a high potential is applied. As in the case of the proportional counter, which also has the highly concentrated field close to the wire, most of the electron multiplying occurs in a region near the anode. Photons of such high energy are released that the localized ionization soon spreads completely along the wire by additional electrons released by the photons. The electrons can be readily collected; they are close to the wire and have only slight inertia due to their small mass. This positive ion sheath which is left close to the wire then moves slowly toward the cathode. The sheath effectively shields the anode and reduces the electrostatic field around it to a point below that necessary to register counts. As the sheath is attracted toward the cathode, the field again recovers and becomes large enough to register counts again. The voltage transient produced is smaller than that of a normal count unless the original field has again been established by the complete neutralization of the positive ion sheath at the cathode. The neutralization of the positive ions is the process that makes the discharge self-maintaining. The positive ions have enough energy to cause electrons to be liberated at the cathode. These electrons then become the source of another discharge. Argon, the rare gas usually used in the tube, can exist in metastable forms that have high enough energy to ionize the gas on reversion to a normal state, and, thus, also help in the

maintenance of the discharge (41).

For a long time, the Geiger tube, because of its complexities, was a difficult piece of apparatus to use (42). The first successful quenching employed a high resistance in series with the tube and the high voltage supply (43). The flow of electrons through this resistor lowers the terminal potential of the tube, and if the resistor is high enough (10^{10} ohms or higher), the tube will not continue to discharge. The flow of electrons through this resistor will lower the terminal potential to a point that no longer permits the discharge maintaining factors to operate. The recharging of the detector tube limits the count rate in a manner similar to the ionization chamber. This accomplishes quenching; however, the technique for stopping the discharge has been accomplished by methods that are much faster, and allow a more rapid count rate to be recorded.

The second method used to stop the discharge also reduces the terminal voltage. The original quench circuit was described by Meher and Harper (44). Voltage created by the Geiger tube was used to control the terminal potential by the effect of the voltage on a vacuum tube. In this case, the terminal potential is kept reduced only long enough to stop continuous discharge of the tube. As no extremely large resistors are involved, this method is considerably faster than the resistance quenched tube.

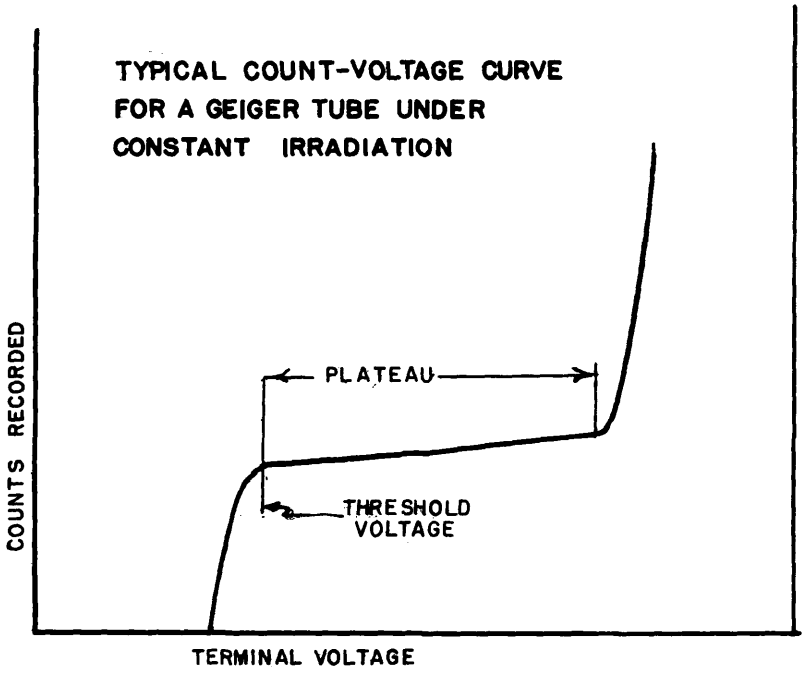
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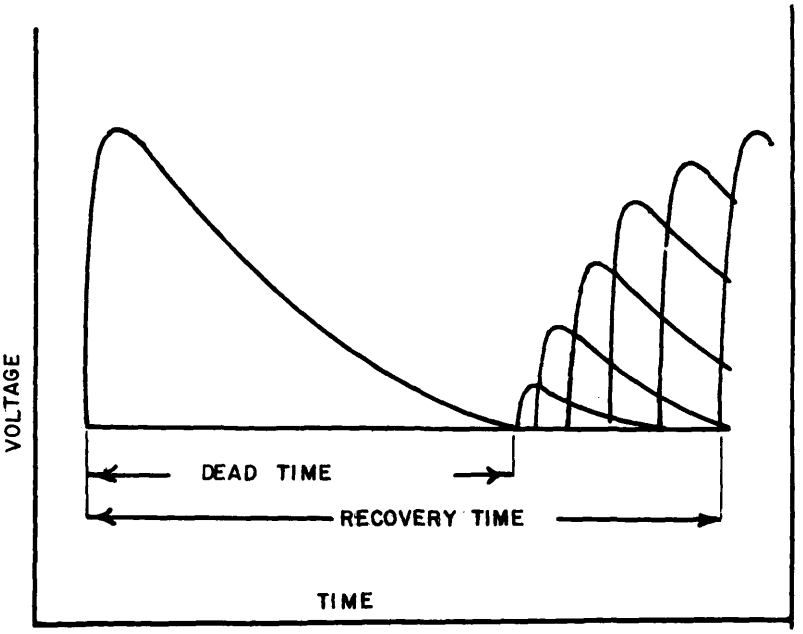
its dimensions and the voltage applied; the proper adjustment of the quench circuit, therefore, will vary from tube to tube (41). This method of quenching the discharge is used only in those cases that make the following method of quenching difficult or impossible.

Trost in 1937 discovered that no external discharge quenching was needed if organic vapors were mixed with the rare gas in the tube. This discovery led to the self-quenched type of Geiger tube which contains a vapor that will cause termination of the discharge. The energy that would create the new electrons to maintain the discharge as well as the energy contained in the metastable ions (42) is dissipated by decomposition of the organic vapor.

The characteristics of an organic gas-quenched tube over a range of terminal potential is shown on Figure 1. No counts are recorded until the minimum potential is reached. The counts at this voltage are mostly in the proportional region, and can be observed to be of different amplitudes on an oscilloscope. Soon the plateau is reached, and the change in number of counts recorded will vary only slightly with increased terminal potential. At the end of this range, which is 200 volts or more in the best tubes, the count rate again increases; and if the voltage is increased too far over the end of the plateau, the tube undergoes continuous discharge, and will be destroyed in a few seconds.

The nature of the voltage pulse produced by a Geiger tube and the method of determination of dead time and





SYNCHROSCOPE DISPLAY OF GEIGER TUBE VOLTAGE TRANSIENTS AT HIGH COUNT RATE

FIGURE 1

recovery time by a single sweep triggered by the incoming transient as described by Stever (45), is shown in the lower half of Figure 1. After discharge has been started by one disintegration, no further disintegrations can be detected until a minimum time has passed. This time is called the dead time. As the positive ions move closer to the cathode, a point will be reached where they no longer neutralize the field sufficiently to prevent the further counts from becoming recorded. If the ion sheath has been completely neutralized, the next ionizing event will cause a pulse equal to the first; the time required for this to occur, after a given pulse is the recovery time. The instruments necessary to obtain such an analysis of the voltage transients will be discussed in a subsequent section.

The dead time and the recovery time depend mainly upon the physical size of the tube used—the shorter the tube, the shorter the distance over which the ions must travel. Tubes filled as described later in the thesis, with a 20 mm diameter, have a dead time of about 100 microseconds and a recovery time of about 200 microseconds.

This self-quenched tube is the most popular detecting device in use at the present time. Minimum amplification is required because the voltage that results from the detecting device is large. Stabilization of the high voltage supply is simple because wide range of terminal potentials causes little change in count rate.

The self-quenched tube, however, has several

disadvantages. The majority of the tubes used are of the organic quench type. The energy dissipated by the quench results in the decomposition of the material and causes the slope of the plateau to increase with use. A count of about 10^8 is obtained with most tubes before the slope of 25% per 100 volts increase in terminal potential is reached (46), the accepted end of usefulness of a Geiger tube. The change in slope is gradual in most cases. Also concurrent with the increase in slope is a shortening of the plateau, and sometimes an increase in sensitivity to light.

The usual tube is filled to only about 100 mm internal pressure. A gas-tight barrier must therefore be interposed between the sample and tube. This limits the operation of the tube to counting radiation that can penetrate thin barriers. Windows of glass and mica can be made thin and gas tight enough to yield tubes that are stable for long periods. Nylon and other plastic films have been used, and can be made in densities less than the 1-2 mg. per sq. cm that seem to be the limit of glass or mica windowed tubes. The permeability of the organic films, on the other hand, do not permit the tubes to be used over a long period without refilling.

EXPERIMENTAL SECTION

INSTRUMENTATION

Synchroscope Amplifier

Theoretical. The most widely used forms of detectors yield small voltage transients when they are struck by radiation. The size and duration of the voltage depends upon the detector used. All voltages are reasonably short, and amplifiers are used to increase their amplitude if necessary. All detectors require some sort of instrument to analyze the rate at which the voltage transients are detected. As has been stated in the discussion of ionization chambers, some devices do not record individual counts, but integrate the count rate. At best, these are only semi-quantitative and are used only where the highest accuracy is not desired (47), and simplicity is required, as in a survey meter. The individual recording of each ionizing event can yield much higher accuracy of radiation counting and is almost universally used for quantitative radiation detection.

Electronic operations on the transients are made in all radiation detecting apparatus. These voltage transients, which may also be called pulses, are similar to those that are encountered in other electronic work; and the terminology used is common to both fields. If the instantaneous voltage developed by any source is plotted as ordinates and time is plotted as abscissas, a graph called wave form is produced. The time required for the transient to be completed is

usually called pulse width or pulse length. The maximum voltage of the transient determines the pulse height (amplitude).

The pulse from a radiation detector may have many electronic operations performed on it before it is registered. Changes in detector pulse length in many cases improves the reliability of a scaler. The pulse may be too small to be used as a trigger for the following circuitry. Voltages of this sort require amplification. If separation of the radiation detected is to be based on the proportionality of the detector output, it is necessary to have a proportional increase in pulse height. Neglecting this separation, the circuit has to record only once for each time that the detector is hit by a radiation. The information can be transmitted by any voltage that will produce a recorded count of one. Thus, the length of a pulse can be widely changed with no change in the accuracy of the recorded count. For examples, the pulse may be shortened to permit short resolution time scalers to function properly; the pulse may be lengthened to permit slow acting electromechanical registers to complete their cycle.

Observation of the changes in these transients as they are utilized in the electronic circuitry is the best method of adjusting circuits for proper operation. The observation of duration and amplitude is usually made by means of the cathode ray tube. In this instrument an electron beam is deflected across a fluorescent screen by

means of two pairs of deflecting electrodes. The electrodes are mounted so that the locus of deflection produced by one pair is at right angles to the locus of deflection produced by the other pair. The deflection of the beam is proportional to the voltage causing the deflection. Consequently, if a voltage that increases linearily with time is impressed upon one pair of electrodes—the pair chosen customarily caused deflection in a horizontal plane—a linear time base will be established. The voltage to be studied is applied simultaneously to the other pair. In this manner, the length and instantaneous voltages existing at any time can be determined. The superimposed tracings of a series of similar voltages taken from an instrument developed in the course of the work are shown in Figure 8.

The cathode ray tube may require voltages many times those available from the circuit being studied. The voltage required to produce a deflection of one inch on the face of a 5BP1-A, a representative cathode ray tube, is about sixty-five volts (48). In most cases the voltage studied is but a small fraction of this potential, so amplification must be applied to the voltage if a visible deflection is to be observed. Some cathode ray tube installations have amplifiers built into the instrument to permit low voltages to be handled. This is the case for most of the oscilloscopes.

Both the synchroscope and the oscilloscope have a circuit that generates a linearily increasing voltage to yield the horizontal time axis. Because both instruments

are designed to detect relatively rapid changes in voltage transients, the phosphorescence of the screen is held to a minimum so that the pattern will not be so lasting that the changes are masked. With rapidly changing transients, observations of the voltage transients are difficult because the screen must be repeatedly retraced to give an observable pattern. The oscilloscope causes traces to be superimposed by allowing the time sweep to be initiated by some portion of the transient being studied. This process of coincident retrace is called synchronization. As the sweep generator is cyclic in nature, synchronization can occur only if the transient being studied is also cyclic in nature. The statistical nature of radioactivity thus makes an oscilloscope relatively useless as an indicating device for the study of radioactivity. The synchroscope, however, does not depend upon cyclical transients, but permits the sweep to be generated only after a transient has been received, and, thus, allows superimposition of random voltages. The model P4 M.I.T. synchroscopes used did not have any amplification, yet must have a minimum of 100 volts to give proper operation.

It became obvious soon after experimental work started that it would be necessary to utilize a synchroscope. An amplifier was constructed to permit the observation of small transients. It was decided that this amplifier must retain as much proportionality as possible, and, as used here, has only the function of amplifying proportionally all the voltages placed on its input terminals. The demands placed on this amplifier were quite extensive. It can be

shown from Fourier analysis of non-sinusoidal waveforms (49), as well as by actual experimental techniques, that to preserve the fast rising portions of any short voltage transient, good high frequency response is required of an amplifier which increases such a voltage. The electrons which are collected by the anode of a Geiger tube, for example, can be collected in less than a microsecond; and thus, fast rising transients are to be found in radiation detection. The usual type of amplifier used in this work is a variation of the resistance coupled amplifier. The amplification possible at high frequencies with a resistance coupled amplifier is limited by the characteristics of the tube and the characteristics of the circuit in which it is used. The most critical parameter is the capacitance that is associated with the tube and the stray capacitance between the wiring. This provides an additional degenerative path for the currents that flow in the coupling circuit when a signal is amplified (50). The shunting effect becomes most pronounced at higher frequencies where the capacitative reactance becomes low. The phase relationships at these high frequencies become poor at the same time. This adds to the distortion of a waveform, since the Fourier analysis shows that not only must the wide frequency range be amplified, but also a linear relationship between the frequency and phase shift must be retained (51).

A second requirement of a test amplifier is that it must show a low shunting effect when placed across any

circuit being studied. The maximum resistance of any amplifier will be shown between the grid and cathode. This resistance cannot be too high as this is the path of the electrons that neutralize the positive ions formed due to electron bombardment of the residual gas in the tube and are attracted to the negative grid. Too high a resistance will allow this current to be so large that the negative grid voltage is neutralized. If the grid is driven positive, it will draw current from the electron stream (as will any anode) and the grid to cathode resistance will drop to a low value.

The grid cathode resistance is also shunted by the input capacitance of the vacuum tube. The shunt capacitance of a triode acting as an amplifier is the highest of the tubes available for this purpose. This is due to the Miller effect (52), which is an apparent multiplication of the input capacitance due to the relationship of the grid and plate voltages. The pentode voltage amplifier does not suffer from apparent change in input capacity because the plate is screened from the grid and has little effect upon its operation.

One of the lowest input capacitances is shown by a cathode follower. This is a device that has a slightly lower voltage output than the voltage input. It can, however, increase the power in the voltage pulse, and this slightly smaller voltage can then be developed across a much smaller resistance. The response of a cathode follower

extends from D. C. to frequencies much higher than those amplified by good high frequency amplifiers (53).

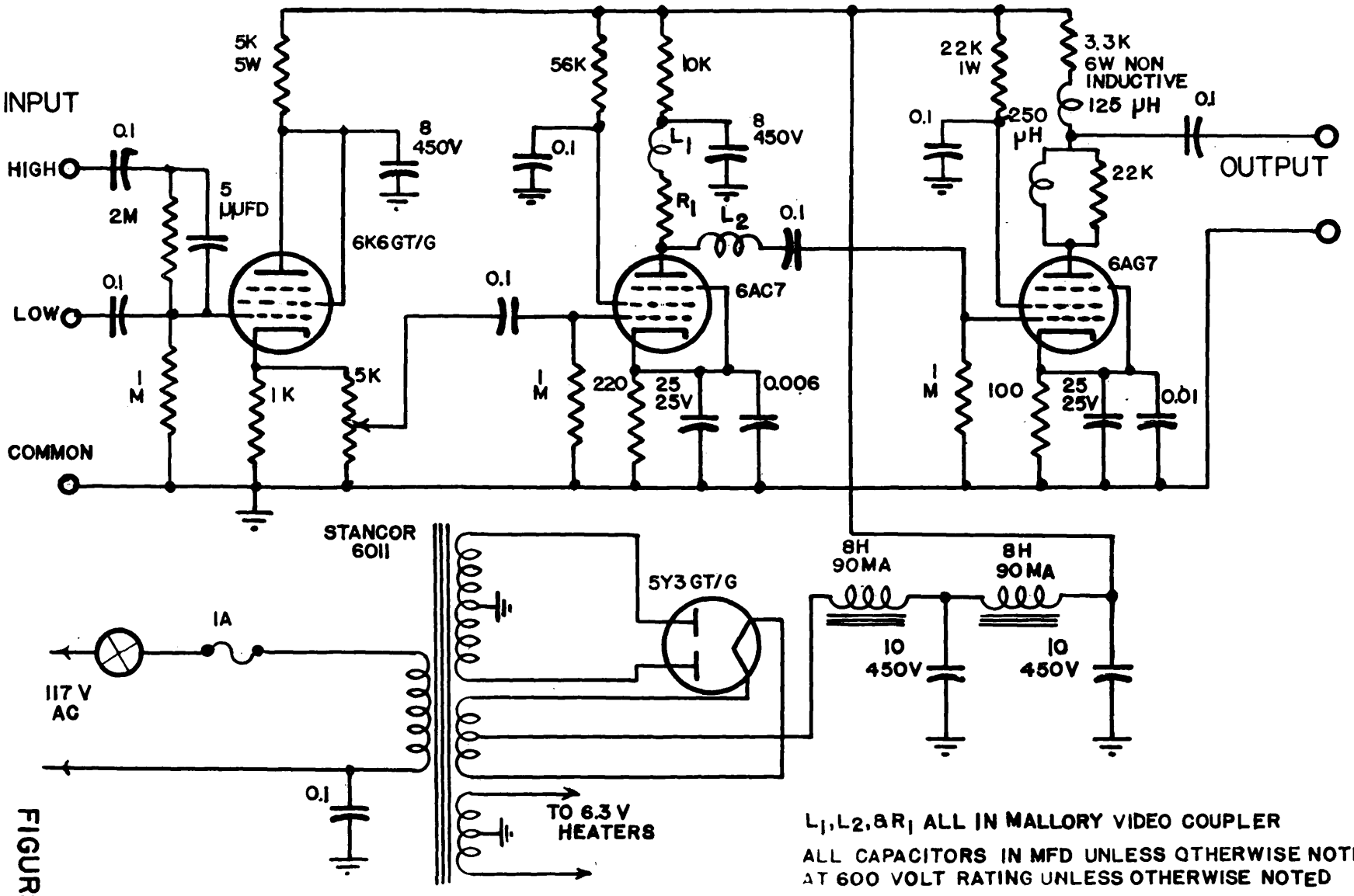
The problem of adjusting the overall amplification of an amplifier becomes a problem when high frequencies are handled. There are two kinds of devices that are used to subdivide the voltage to a voltage more easily seen on the cathode ray screen. They are both voltage dividing resistances—one is fixed; and the other is variable. The fixed voltage divider is the more easily handled. The resistance that is used as a voltage divider usually has the shunt capacitance inherent to either the next tube or the indicating device across only a portion of the two or more resistors. This capacitance, therefore, shunts only a portion of the divider, and at high frequencies, the voltage division is no longer constant. The change in voltage division is a function of the shunt capacitance of the device utilizing the divided voltage, the frequency being subdivided, and the total resistance in the divider network. A fixed voltage divider has a capacitor shunting the portion of the network that is not shunted by the next portion of the circuit. The size of this capacitor is made variable, and is usually adjusted in size until the capacitive voltage division is equal to the resistive voltage division (54). A resistor-capacitor network of this sort is called a frequency compensated voltage divider network.

The problem of the continuously variable voltage divider is not so easily solved as is that of the fixed type. The size of the capacitor required will vary with

each setting of the control. One of the most satisfactory methods used in this case is the use of as small a total resistance in the voltage divider as is possible.

Construction of synchroscope amplifier. The considerations just discussed were all kept in mind when the amplifier was constructed for use with the synchroscope. The circuit developed is shown in Figure 2. The voltage being studied is first impressed upon a cathode follower, the 6K6-GT. As the voltage that the 6K6-GT will handle is somewhat limited, the installation of two inputs was necessary. One allows the whole input voltage to be utilized, and the other which is frequency compensated allows division by the fixed factor of three. The 6K6-GT has a low output impedance, and, therefore, can be loaded by a small voltage divider. Thus, voltage division is possible with a minimum of frequency distortion.

The portion of the voltage to be amplified is fed to a 6AC7 pentode voltage amplifier. This is a high frequency compensated amplifier. Two inductances are placed in the coupling network between this tube and the next. These inductances have the effect of preserving the response at the end of the high frequency range, and preserving the relationship between frequency and phase closer to that which is ideal (55). These inductances and the resistor used as a plate load are all included in a Mallory Videocoupler. The next tube, a 6AG7, is also a resistance coupled amplifier; in this case, a tube capable of a higher output voltage



$L_1, L_2, & R_1$ ALL IN MALLORY VIDEO COUPLER

ALL CAPACITORS IN MFD UNLESS OTHERWISE NOTED AT 600 VOLT RATING UNLESS OTHERWISE NOTED

ALL RESISTORS ½ WATT UNLESS MARKED

$M = 10^6$ $K = 10^3$

FIGURE 2

WIDE RANGE AMPLIFIER

than that possible from a 6AC7. Here again, both shunt and series compensation are used to increase the marginal high frequency gain. The parameters used are those that are recommended by the Radio Corporation of America in the data offered for the 6AG7 (56).

The power for the plates, and the heaters are provided by a power transformer and a conventional full wave rectifier with a choke input filter.

To check the operation of the amplifier at high frequencies, an operational check was made. The source of the signal was a General Radio 605B Standard Signal Generator set at one volt output. The measuring instrument was a 5BP1 cathode ray tube. The low gain channel was used with the variable gain control set to permit one quarter of the maximum gain of 400 to be employed. Table I on the following page shows the frequency in megacycles in relation to the percentage of output.

The lowest frequency that the instrument has to handle is relatively high, and is easily handled without any special circuit compensation.

The apparatus was successfully used in many investigations. The three decibel loss at about 3 megacycles was more than sufficient to reproduce the voltage transients developed by Geiger tubes without noticeable deviation from the published waveforms for this sort of detector. It is probable that it may be somewhat limited in its application in the case of the faster types of counters because

TABLE I . Operational check of the synchroscope amplifier at high frequencies

Frequency Megacycles	Relative Output Percent
0.01	100
0.1	100
0.5	95
1.0	85
1.5	85
2.0	80
2.5	70
3.0	70
3.5	30
4.0	0

amplification used in this sort of equipment is much more demanding.

High voltage supplies

Theoretical. Practically all electronic detecting devices utilize a high voltage in their operation. The voltage that is needed may vary from 400 to 5,000 volts. Any individual detector usually will require only a small fraction of this range in its operation. The range provided for most counting devices used with Geiger tubes is from 500 to about 1,500 volts. This permits almost all conventional Geiger tubes to be used. Once a voltage has been chosen for

the detector, the voltage should remain constant. The reason for this is that many detectors will yield higher counts from a constant source of radiation if the terminal voltage is increased. Constant voltage is maintained by a stabilizing circuit of some sort because unregulated voltage sources generally are not of constant potential. Most regulators are supplied by an unregulated voltage of a higher potential than will be required from the stabilized source at any time. The output voltage is then controlled by the loss in some part of the circuit—the loss in voltage being equal to the unregulated supply minus the regulated terminal potential. This loss is variable so that the supply can be adjusted to meet the demands of the detector used. Once the output voltage has been adjusted, the loss is automatically changed in a manner which tends to keep the terminal potential constant.

Perhaps the most satisfactory source of unregulated voltage, when 60 cycle line current is available, is a half or full wave high vacuum rectifier. This is supplied with alternating voltage from a transformer. The filtering requirements for the pulsating voltage thus generated are simple, because the regulating circuit, in general, is fast enough to smooth variations occuring at the rate of the line supply. This yields a source for regulation that has relatively low internal resistance. While the load, placed on a power supply by a detector, can be neglected, the voltage indicating meters are more rugged if they can

draw a fairly high current. The operation of this type of device is simple and dependable; and the measurement problem encountered is not severe. Some of the high resistance circuits used in other high voltage supplies require extremely sensitive voltmeters.

Stabilization can be accomplished by several methods. Certain gaseous tubes have almost constant voltage across their terminals over a wide range of current flow through the tube. Two types are available. One type has a relatively low constant terminal voltage at a relatively high current drain (57). A large number of these would be required to give a high enough potential. They are, however, used as a source of reference voltage for comparison purposes.

The other type has a terminal potential that is much higher. This tube is designed to operate Geiger tubes and thus does not have the current capacity of the lower voltage devices. The terminal potential is not too constant, and the tube must be designed to operate in the region of the best detector operation. Only one terminal stabilized voltage is thus possible.

All devices that are designed to handle tubes of different characteristics use electronic stabilization. Methods of electronic stabilization have been subdivided into three classes by Hunt and Hickman (58): transconductance bridges, amplification factor bridges, and degenerative amplifiers. The stabilizer is not limited to the use of one circuit alone, but may use several to achieve the desired

results.

The transconductance bridge type circuit maintains a constant output potential by changing the current down by a vacuum tube from a source of relatively high internal resistance. The plate current change is such as to minimize output voltage changes. The stabilization obtained is a function of the transconductance—the change in plate current of the tube for a change in grid voltage at constant plate voltage.

At one output voltage, the regulation will be perfect. At all others it will vary to an extent dictated by the parameters of the circuit.

The amplification factor bridge and the degenerative amplifier both have the voltage drop across a vacuum tube, which is in series with the load and the source of non-regulated voltage. They differ in the manner with which they couple voltage changes to this series tube which minimizes the change.

The amplification factor bridge uses part of the change input potential to vary the grid voltage of the vacuum tube; the increase or decrease thus regulates the plate potential so that it is changed almost opposite to the variation in source potential. This circuit has one voltage where constant output voltage can be obtained; all other voltages are somewhat less stabilized.

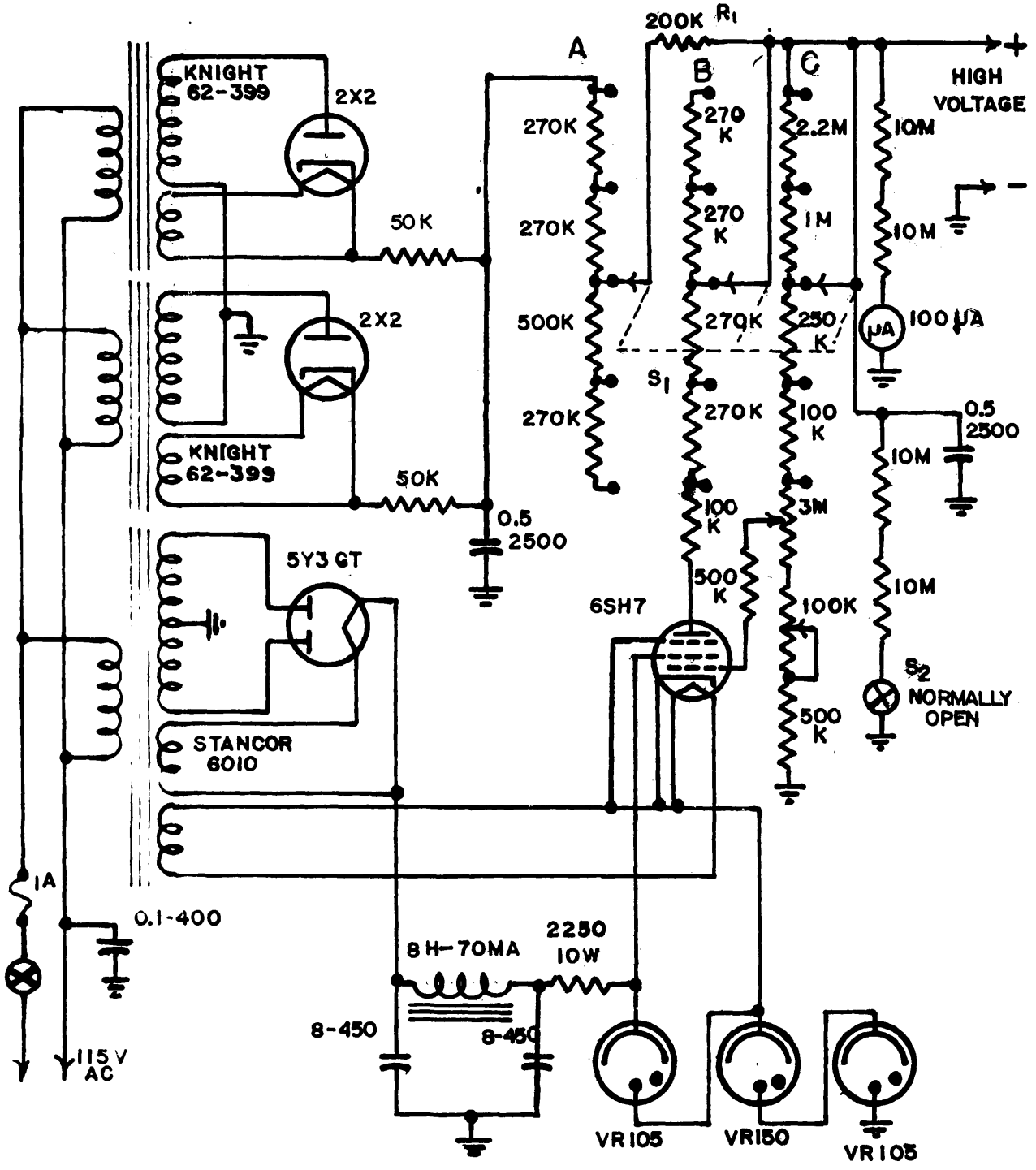
The degenerative amplifier places the output potential, or some fraction of it, between the grid and cathode

of the regulating tube. An increase in output voltage causes the tube to increase its apparent resistance and thus minimized voltage changes.

The single types can be used in conjunction with each other to get better or more flexible voltage regulation. All require one or more stable voltages, either for reference or for other circuit functions.

Experimental high voltage supply Type I. The schematic diagram of the first high voltage supply is shown in Figure 3. The unregulated high voltage is furnished by two 2 x 2 high vacuum rectifiers. The plate and filament voltages are furnished by two Allied Radio 62-399 transformers that supply 1,500 volts R. M. S. for the plate supply and 2.5 volts for the heater. As the drain on the supply is very small, the voltage output will be close to the peak value of the instantaneous voltage of the transformer, or 1.4 times the R. M. S. value. The two transformers are phased so that they conduct alternately, thus reducing the cyclic variations in the output voltage.

The stabilizing circuit used is that of the transconductance bridge type. It requires only one vacuum tube in its operation, the 6SH7. The current drawn by the whole circuit is drawn through the 50,000 ohm resistors that are in series with the cathodes of the tubes; with such resistors of the vertical resistance bank A as are selected by the range switch S_1; and with the 200,000 ohm resistor R_1. The load includes the load of the 100 microampere voltmeter



ALL RESISTORS ½ WATT UNLESS NOTED
K = 10^3, M = 10^6 OHMS

HIGH VOLTAGE SUPPLY

FIGURE 3

and its 20 megohm multiplier; the negligible load of the detector (59); the load of the voltage dividing network C; and the plate load of the vacuum tube, 6SH7. The current drawn by all these causes a drop across the resistors noted and lowers the output voltage to that chosen. To vary the output, the voltage drop is increased or decreased by changing the plate current of the 6SH7. The plate current of a vacuum tube can be changed by changing the voltage at the grid with respect to the cathode. The point of contact of the variable tap on the 3 megohm potentiometer in column C can be changed. This allows that portion of the output voltage applied to the grid of the 6SH7 to be changed. The cathode of the tube is kept at a constant potential by means of the VR 105 and the VR 150, both low voltage stabilizing gas tubes. Most of the current passing through the tubes is drawn from the output of the 5Y3GT rectifier and its filter system. This places no power demand on the high voltage supply.

Voltage stabilization occurs in the following manner. An increase in voltage causes an increase of the grid voltage in a positive direction. This, in turn, increases the plate current of the 6SH7; therefore, minimizing any large changes. Conversely, a lowering of output voltage is resisted by a decrease in plate current of the 6SH7.

A circuit, made up of two 10 megohm resistors and switch S_2, tests the amount of regulation. If the regulator

is working at a stable point, closing the switch will not change the voltage as read on the voltmeter. Resistor bank A allows the voltage drop to be shared by several resistors when low output voltages are required. The resistor column B allows the plate voltage of the 6SH7 to be closer to the recommended value than that which would be apparent if no plate resistance were used. The screen potential for the 6SH7 is also stabilized, since this element also controls the current drawn by the tube, and should not vary if good stabilization is to be achieved.

The full range of from 400 to 1,500 volts cannot be covered by one rotation of the 3 megohm resistor. In order to cover the full range, large voltage changes are made by setting S_1 to the proper position. This simultaneously changes all the resistors in columns A, B, and C to the proper values for the range chosen. Coarse adjustments are made with the 3M potentiometer; fine adjustments are made with the 100K rheostat.

The theoretical considerations of this stabilizing system have been derived by others (60). The complete derivation is given here, because the limitations of the system can be more easily seen. Of the possible relations that can be derived, perhaps the most important one is the rate of change in output voltage with changes in unstabilized voltage. Changes in load are negligible in comparison to the other currents flowing in the circuit.

Definitions and terms used:

R_s = total series voltage dropping resistance

R_b = resistance between grid and positive terminal

R_a = resistance between grid of 6SN7 and minus lead

E_i = unregulated voltage

E_o = regulated voltage

R_l = resistance of voltmeter circuit and load

i_b = plate current of the 6SN7

e_c = the voltage of the grid with respect to the cathode

gm = transconductance, $\left(\frac{\partial I_b}{\partial E_s}\right)_{E_b}$ by definition.

It is assumed that the plate resistors and the plate current drawn by the 6SN7 are such that the tube is kept in a region where the plate current is essentially independent of the plate voltage.

By Ohm's and Kirchoff's laws

$$E_i = E_o + R_s\left[I_b + \frac{E_o}{R_a+R_b} + \frac{E_o}{R_l}\right]$$

and if E_i be increased by ΔE_i

$$\Delta E_i = \Delta E_o + R_s\left[\Delta I_b + \frac{\Delta E_o}{R_a+R_b} + \frac{\Delta E_o}{R_l}\right]$$

then:

$$\Delta E_s = \frac{\Delta E_o R_b}{R_a+R_b}$$

If ΔE_i is not large, gm will remain relatively constant

then: $$\Delta I_b = \frac{gm\ \Delta E_o R_b}{R_a+R_b}$$

and $$\Delta E_i = \Delta E_o + \frac{\Delta E_o R_s R_b gm}{R_a+R_b} + \frac{\Delta E_o R_s}{R_a+R_b} + \frac{\Delta E_o}{R_l}$$

and $$\frac{\Delta E_o}{\Delta E_i} = \frac{R_l(R_a+R_b)}{R_l(R_b R_s gm + R_a + R_s) + R_a + R_b}$$

As can be seen, the larger the value of R_1, the more stable the output voltage will be. This is limited to the maximum sensitivity possible when regged instruments are used as voltmeters. An increase in the value of R_b also increases the stability. If this resistor is made too large, the current drain of the circuit beyond this resistor would require too high a voltage from the unregulated source. The final design must be a compromise between all of the above variables. Since the voltage output is at a maximum when the current of the regulator tube is at a minimum, the regulation at this point will be poorest—the gm of the tube decreases at low plate currents (61).

The operation of the supply, aside from the limited voltage range available from one setting of the range switch, was excellent. Line variations in the neighborhood of plus or minus five volts yielded variations of less than plus or minus two volts in an output voltage of 1,000 volts.

This high voltage supply has given excellent results and has remained relatively trouble free. It was used during the complete course of another investigation using radiophosphorous with no failures of any sort.

Experimental high voltage supply type II. The subsequent voltage supplies designed and used were of slightly different construction. This was necessary in order to eliminate the use of range switches to cover the range between 500 and 1,500 volts. All the voltage supplies of the later design will cover this voltage range with one

setting. The regulation of this method depends upon a direct current amplifier coupled directly to a degenerative amplifier (58). This voltage supply, type II, has been relatively widely used in voltage stabilization circuits for use with Geiger tubes (62 - 64). Although part of a complete circuit, which will be described more fully later, the high voltage supply will be described now in order to allow the two methods, types I and II, to be compared.

The use of a vacuum tube as a voltage dropping device has many advantages. Its range of resistance is almost infinite, thereby eliminating many of the faults of a fixed loss system. The second system devised has a range 450 to 1,500 volts; this change in voltage is accomplished by the rotation of one potentiometer throughout its range. To provide for higher voltages for detectors that require voltages in excess of this value, the third and subsequent systems had two ranges: the one just mentioned, and a range extending from 1,000 to 2,400 volts. The two ranges in this latter case were intentional because this makes setting not so critical in the range of 800 to 1,000 volts—the usual voltage required by conventional Geiger tubes. A representative circuit is shown in Figure 8.

As can be seen in Figure 8, the high voltage is supplied by a half wave rectifier. The transformer used for the low range is the same as that used in the previous circuit, as shown in Figure 3. When the high range is used, the voltage is increased by the introduction of an additional

transformer to provide the necessary increase in unregulated voltage. The 6BG6-G (V4) is the tube used as the variable resistor. The 6BG6 (V5) is the comparison tube.

The voltage reference standard is furnished by the use of two 5651 voltage reference tubes—gaseous type tubes designed for this service. The terminal potential of this type stays more nearly constant than does the terminal potential of the previously used VR types. Two are used to give a higher voltage with which the fraction of the high voltage may be compared.

The meter used in this high voltage supply can be less sensitive, because the current capabilities of this system are much greater than those of the first. A one milliampere meter is used. In addition, in order to expand the scale of the meter in the usable range, a 0.33 milliampere preload was placed on the movement. Zeroing of the meter was accomplished by the use of an external high accuracy meter and multipliers while the voltage was set to a range that was common to both.

The changes in unregulated potential are minimized by the reactions of the circuit. A fraction of the voltage change is compared to the reference voltage by the comparator tube, V_5. The increase in plate current causes an increase in drop across the resistor R in the plate circuit of the tube. The drop in voltage across this resistor is, however, the bias voltage on the resistance tube, V_4. An increase in the bias or negative voltage at its grid with

respect to its cathode, increases its effective resistance. This, in turn, tends to resist the voltage change at the output terminals; and, thus, the change at the terminals is minimized. The reverse reaction minimizes any lowering in voltage.

When the range is changed, the voltage to the regulator is increased, the size of the voltage divider is changed, and the resistance of the voltage output control shunt is changed. The last item is adjusted so that the potentiometer will have a maximum of its useful resistance for controlling the output voltage, and, therefore, will not cram the range into a small amount of its rotation.

The 6BG6-G tubes that are used in the supply are large tubes for the power required. This was necessary because smaller tubes arc between their electrodes when operated under high plate potentials. This phenomenon was not recorded in the older literature (65). This article, however, was concerned with the tubes that were manufactured at that time. The destruction of an old type 57 pentode indicated that the spacing of the elements of the tube, especially the leads to the tube base, were much more widely separated than those of a more recent 6SH7. The 6BG6-G, however, has been designed to handle voltage transients of over 5 killovolts and is, therefore, suitable for handling the voltages that are used in the high voltage system. It may be possible that one of the smaller tubes will be satisfactory in the place of the 6BG6-G tubes used.

The 6BG6-G has proven satisfactory up to two and one half killovolts, and has proven to be more satisfactory in supplies of one and a half killovolts than smaller types.

The cathodes of both V_4 and V_5 are at relatively high positive potentials when compared with the common negative terminal of both rectified voltage sources. This common negative terminal is the place where one terminal of the 6.3 volt heater windings is customarily connected. Separate small filament transformers, therefore, are used to provide the potentials necessary to heat each tube. The cathode is connected to one side of the heater winding in order to eliminate any voltage differential between heater and cathode. The insulation of these transformers must be capable of withstanding relatively high voltage differentials. The Stancor type P-6134 has proven to be satisfactory at potentials up to 2.5 killovolts.

As in the case of the first type of voltage regulator the theoretical voltage regulation of type II was also derived. This derivation is included to show the assumptions required, and the circuit considerations that must be met to give good voltage regulation.

Definitions and terms used:

R_1 = load resistance neglected here because the voltmeter load is part of the voltage divider

R_a = resistance between positive terminal and tap on R_{15}

R_b = resistance between negative terminal and tap on R_{15}

g_m = transconductance, $\left(\frac{\partial I_b}{\partial E_s}\right)_{E_b}$

I_b = plate current of the tube

E_b = voltage at the plate with respect to the cathode

E_g = voltage at the grid of a tube with respect to the cathode

μ = amplification factor of a tube,

E_i = unregulated voltage

E_o = regulated voltage

Numbered subscripts refer to the individual components on Figure 7.

As before, the most important characteristic is the change in output voltage that occurs when the unregulated voltage changes.

By Ohm's and Kirchoff's law:

in the circuit,

$$E_i = E_o + E_{b_4}$$

and, on changing E_i by ΔE_i,

$$\Delta E_i = \Delta E_{b_4} + \Delta E_o.$$

Since ΔE_o will not change greatly, the effect of ΔI_{b_4} on μ_4 can be neglected.

Then:

$$\Delta E_i = \mu_4 \Delta E_{g_4} + \Delta E_o$$

$$\Delta E_{g_4} = \Delta I_{b_5} R_8$$

assuming no change in gm_5 for change in the input voltage,

$$\Delta I_{b_5} = gm_5 \Delta E_o R_b / R_a + R_b$$

substituting,

$$\frac{\Delta E_o}{\Delta E_i} = \frac{R_a + R_b}{R_b(\mu_4 gm_5 R_8 + 1) + R_a}$$

As can be seen, the larger R_b and the larger R_8 are, the better will be the voltage stability. The size of R_b, however, determines the size of the reference voltage, which has practical limits. R_8 has a maximum size as it is the

path of the grid current and this resistor cannot be made too large without losing the bias voltage. One megohm is the maximum for most vacuum tubes that can be placed between the grid and cathode.

The regulation afforded depends upon both the transconductance of V_5, and the amplification factor of V_4. Both tubes are connected as triodes to simplify their voltage requirements. The μ of a triode stays constant over a wide range of plate currents and will not vary too much in this case (66). The gm is a function of plate current, and will decrease as the plate current of the tube decreases. The value of the tube characteristics cannot be determined from the data usually published for the tubes, because the range in which they work is beyond that generally used in their operation.

The overall voltage regulation was determined experimentally. The voltage supplied to the primaries of the transformers was varied. The value of the voltage was determined by an A. C. voltmeter. The line voltage should vary the unregulated supply directly in proportion to its changes. The output from the stabilized supply was determined by a precision microammeter and a precision multiplier. The terminal voltage was chosen at 1,000 volts. At this terminal voltage, the change in output voltage was 0.03% per volt change in line voltage for a range between 105 and 125 volts.

This circuit gave excellent results. The substitution of 6B06-G tubes for the small tubes in the original

model stopped internal tube arcing. The voltage supplies were used extensively during the work described in the later portion of the thesis and no troubles of any sort developed.

Amplifier and Wynn-Williams circuit

Amplifier. The voltage transient furnished by the detecting device is generated by the electron flow through a resistor in series with the detector. This voltage must be capable of causing the subsequent circuitry to record one count. The voltage generated by a Geiger tube may vary between 0.1 volts and well over 10 volts, depending upon the Geiger tube voltage. The smaller voltages must be amplified. However, the smaller voltages that are generated by other detectors may require even more amplification. Amplification is required because the register circuit cannot be made sensitive to extremely small voltages. If the register is very sensitive, small spurious voltages that are present in the A. C. lines would cause spurious counts to be recorded (67). If the circuit is sensitive, small changes in circuit components may cause self triggering; and again, faulty counts will be recorded.

Scalers invariably amplify the detector voltage to one large enough to trigger a stable recorder assembly. The amplifiers used should have good response at high frequencies. The low frequency response in most cases is neglected, or may even be supressed. This supression of low frequency response minimizes the effect of low frequency

disturbance from the 60 cycle line current.

Shaping of the voltage transient may also occur in the amplifying circuits. If a voltage has too long a duration for the best operation of the next circuit, the voltage is transferred between amplifier stages by a short time constant coupling circuit. This is a resistor-capacitor combination that yields a small product when the value of the individual components are multiplied together (68). If such a circuit has a long duration square wave applied to its terminals in comparison to the time constant, the sharp rising wavefront is passed little attenuated. The capacitor charges rapidly—the charge time being the reciprocal of the discharge curve shown in Figure 4. Therefore, the current flow, and the voltage across the resistor decreases rapidly in an exponental manner. When the square wave decays, the voltage rapidly increases in a polarity of the opposite direction; and once again drops to zero as the charge in the condenser is dissipated. This action can also be explained on the basis that the high frequency components in the leading and trailing edges of the waveform are easily transmitted by the coupling circuit. This circuit is called a differentiating circuit and is used widely to create sharp transients. The rise time of the sharp pulse cannot be shorter than the square wave fed into the circuit.

This very convenient circuit takes the form of the circuit used to couple the voltage from the plate of one tube to the grid of the next. The capacitor is also used to

block the D. C. at the output plate from the next grid. This grid normally operates at a different potential. The resistor, the element across which this sharp transient is formed, is the normal grid resistor of the following tube. If the voltage is to be transferred with as little change as possible, the time constant should be made large in comparison to the longest pulse to be amplified.

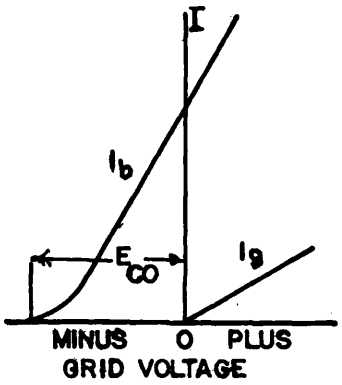
A second method of changing the shape of a pulse is the multivibrator. The circuit is an old one, having been used since its invention in 1919 by Abraham and Black. It has two different forms, periodic, and non-periodic. The periodic forms are susceptible to slight changes in natural frequency. This is due to a synchronizing trigger. However, these periodic forms are not capable of synchronization over the wide rates obtained when counting radioactive sources; therefore, they have little use.

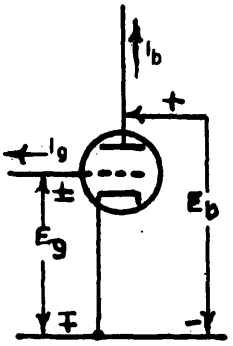
The non-periodic variety are different and are useful. The "one shot" or captive multivibrator, a circuit widely used in subsequent circuits, is described here. It is of the non-periodic type. Such a multivibrator is a circuit containing at least two vacuum tubes having only one stable state (69). It, however, may be driven into an unstable state by a voltage transient. Once it has completed a cycle whose length is determined by the parameters of the circuit, it again assumes the stable state until it is triggered by another transient. The output voltage depends little upon the size of the triggering pulse.

One form of this circuit is shown in Figure 4. The initial situation described will be valid for only the stable state. Tube V_1 does not conduct. It has a negative grid voltage created by the flow of current passing through R_7 which stops the flow of the V_1 plate current. The size of R_7 is chosen so that the plate current of V_2 will create a large enough voltage drop across R_7 to keep the effective grid voltage of V_1 below that which will allow current to flow. The effect of grid voltage on plate current can be seen on the grid voltage-plate current curve in the upper left hand side of Figure 4.

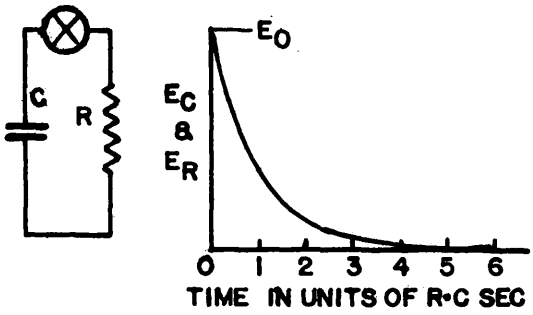
Tube V_2, on the other hand, does not have any negative grid bias. E_{g_2} will be slightly positive due to the effect of R_3, which is connected to the plate voltage supply. The grid potential cannot be more than slightly positive because at a positive potential with respect to the cathode, the grid can draw current from the electron stream in the tube. The grid circuit then becomes a low resistance device. The current drain through R_3 causes a voltage drop almost equal to E_{bb} minus the cathode potential. As V_1 draws no current, E_{b_1} will be equal to E_{bb}. E_{b_2} will not be equal to E_{bb} because V_2 is drawing current through R_2. The charge on C_3 will be determined by the voltage E_{bb} minus E_{g_1}. C_2 will be charged to a voltage E_{b_2}.

The introduction of a voltage transient into the grid of V_1 changes the situation existing at both tubes, providing the voltage is large enough to raise the E_{g_1}



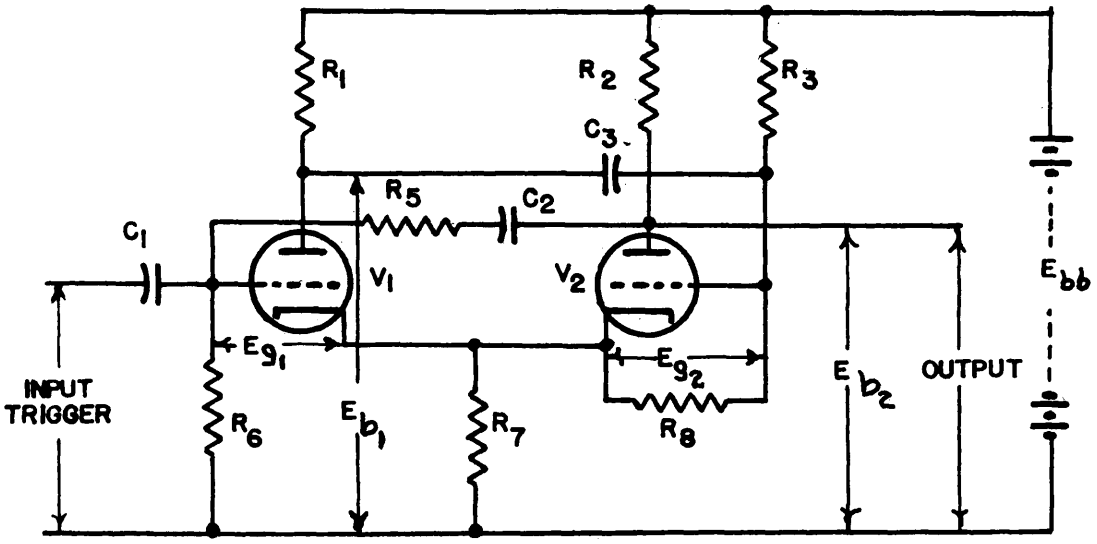


GRID VOLTAGE (E_g) - PLATE CURRENT (I_b), AND GRID CURRENT (I_g) AT CONSTANT E_b FOR A TRIODE.

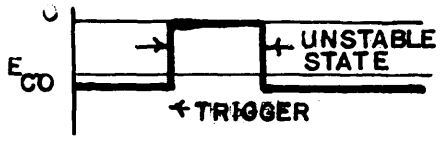


$$E_C = E_R = E_O e^{-\frac{t}{RC}}$$

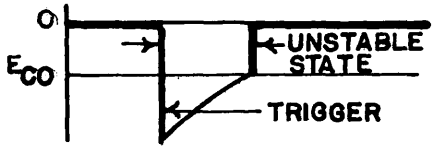
VOLTAGE - TIME RELATIONSHIP OF A CHARGED CAPACITOR WHEN DISCHARGED THROUGH A RESISTOR



"ONE SHOT" MULTIVIBRATOR



GRID VOLTAGE V_1



GRID VOLTAGE V_2

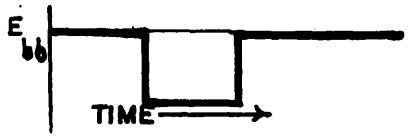


PLATE VOLTAGE V_1

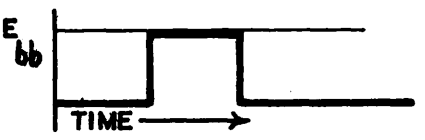


PLATE VOLTAGE V_2

VOLTAGE CHANGES DURING ONE CYCLE

FIGURE 4

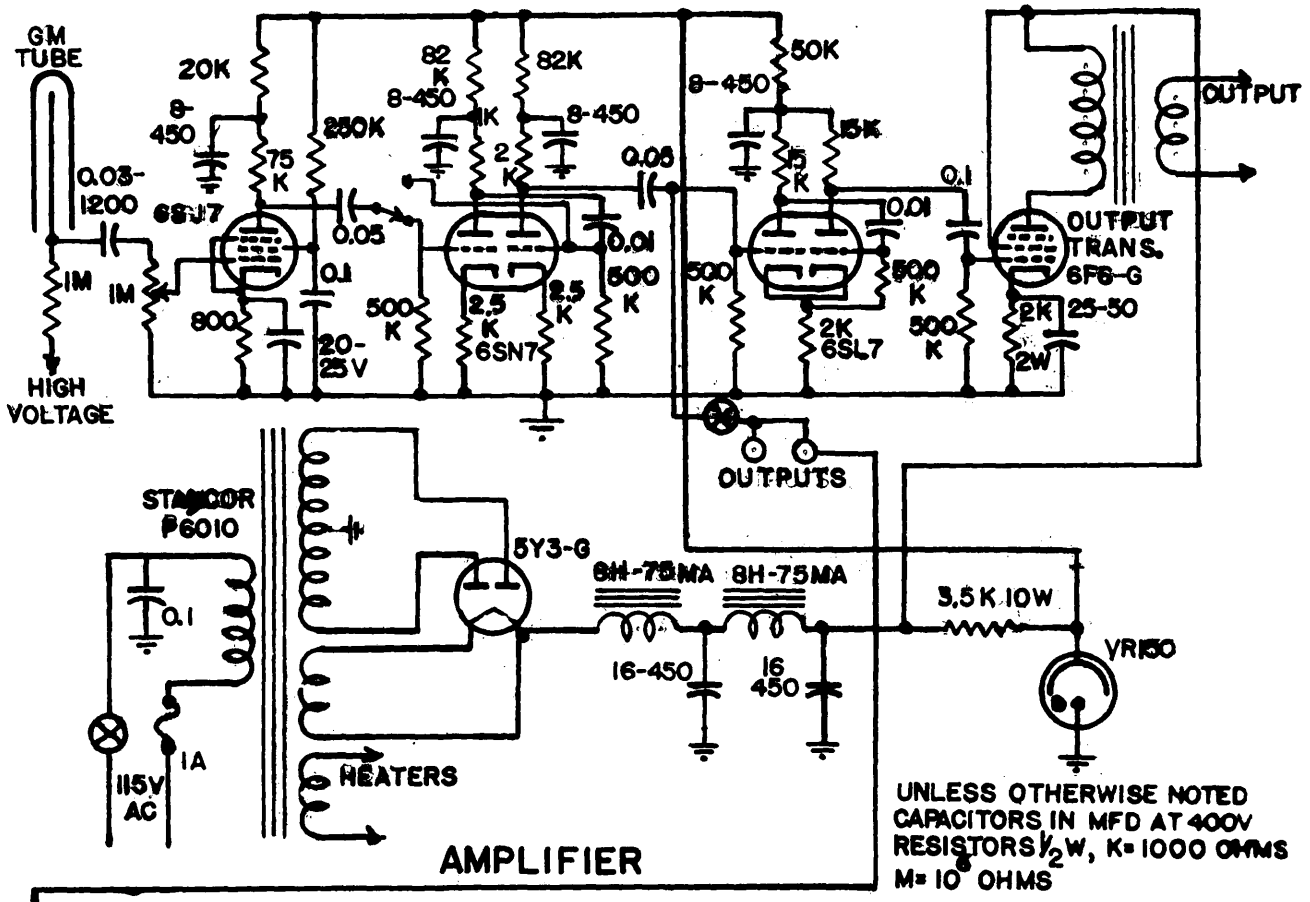
above E_{co_1}. As soon as V_1 starts to draw plate current, E_{b_1} decreases. This causes C_3 to discharge through R_3 and R_8. The current flow through R_8 drives the grid in a negative direction, and this lowers the current drawn by V_2 and raises E_{b_2}. C_2 then charges as it is connected to a source of higher potential than it was during the quiescent state. As I_{b_1} increases, I_{b_2} decreases. V_1 is soon conducting strongly, and V_2 is non-conducting. The speed with which this transition occurs is dependent upon the frequency response of the tube—the better the high frequency response, the faster the transition. The discharge of C_3 will continue as long as E_{g_2} is less than E_{co_2}. The time of the discharge is a function of both the condenser size, C_3, and the resistance through which it discharges. The relationship of voltage and discharge time is shown in the upper right hand corner of the diagram. Condenser-resistor combinations are flexible enough to allow this sort of circuit to complete a cycle in less than one microsecond or over many seconds.

Once E_{g_2} reaches the cutoff potential, the changes which occur when the triggering transient hits the grid of V_1 are reversed. V_1 quickly becomes non-conducting, and V_2 goes back to the stable conducting state. The voltage changes that occur in one cycle are shown below the schematic diagram for the circuit. The rate of triggering is limited to a rate that will not allow the unstable state to be present for more than 0.7 of the elapsed time (70). This

relationship is called duty cycle. The voltage that is delivered to the next circuit depends upon the circuit and not on the transient that caused the cycle to be initiated. The length of the output pulse depends upon the circuit, and not on the pulse causing it. The trigger transient should be shorter than the output pulse, because if too long, it may cause two or more cycles. As connected, the circuit cannot be triggered by negative transients. The voltages that can be generated are shown in Figure 8, C being an example of the short nature of the output voltage available. F is the amplified output of a multivibrator.

The length of the pulse can be calculated (71). The length can also be estimated from the characteristics of the tubes used, and the discharge of the R-C circuit. This latter technique was used in most of the multivibrators employed in the following circuits. Small component changes were made as required to give the desired results.

The first amplifier made operated with the high voltage supply, type I, and the scale-of-eight scaling circuit. The schematic diagram is shown in the top portion of Figure 5. The detector signal voltage is generated across the 1M G. M. tube series resistor and coupled to the 1M voltage divider by the 0.03 mfd capacitor. The amplified signal from the plate of the 6SJ7 then can be placed into either the right or left hand grid of the 6SN7 dual triode. The grid chosen is dictated by the polarity requirements of the following scaling circuit. The voltage to trigger the


GM TUBE
HIGH VOLTAGE
6SJ7
6SN7
6SL7
6F6-G
OUTPUT
OUTPUT TRANS.
OUTPUTS
STANCOR P6010
5Y3-G
8H-75MA
8H-75MA
3.5K 10W
VR150
16-450
16 450
115V AC
1A
HEATERS
AMPLIFIER
UNLESS OTHERWISE NOTED
CAPACITORS IN MFD AT 400V
RESISTORS 1/2W, K=1000 OHMS
M=10^6 OHMS

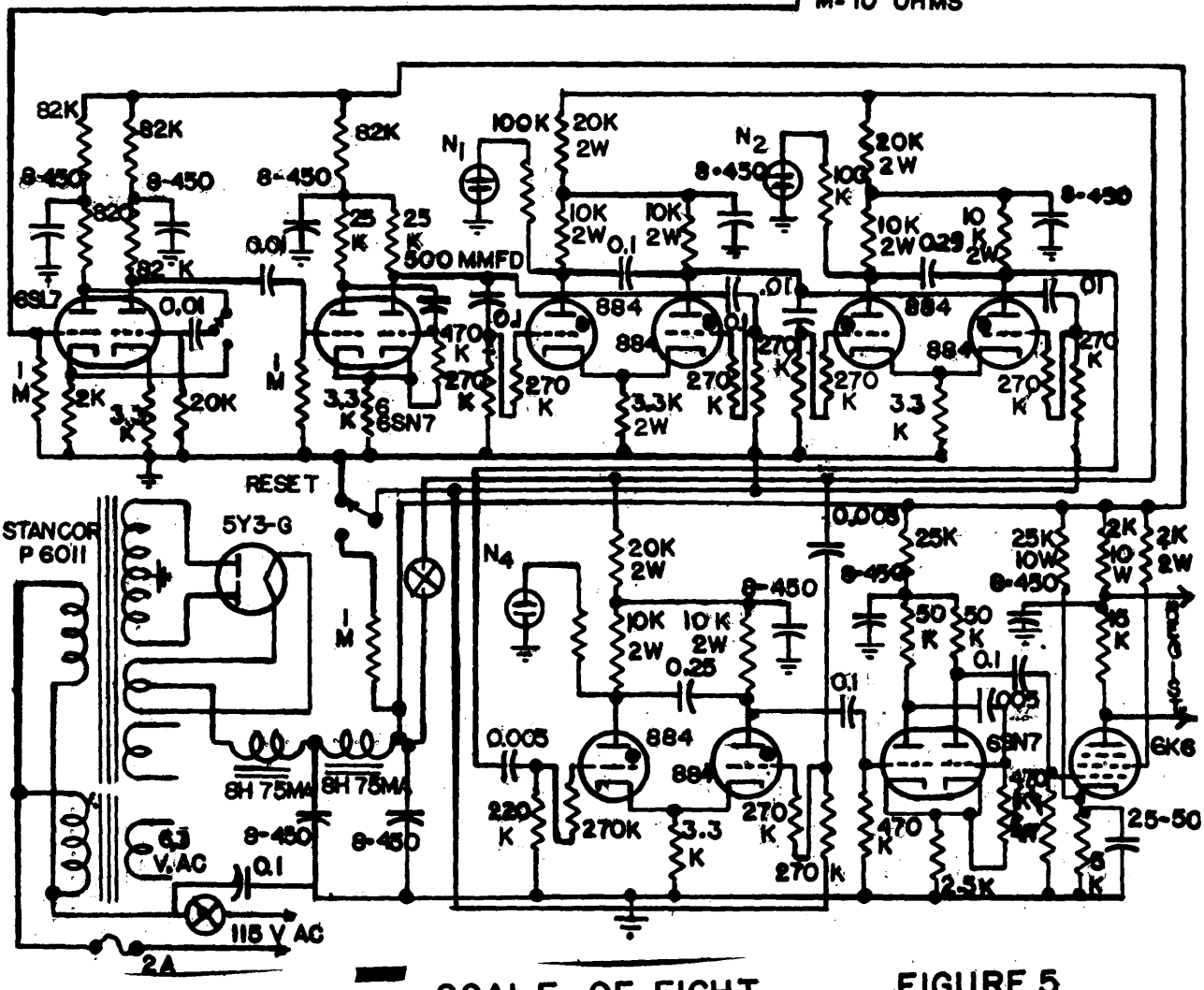

RESET
STANCOR P 6011
5Y3-G
8H 75MA
8H 75MA
6.3 V. AC
115 V AC
2A
884
6SN7
6K6
SCALE OF EIGHT

FIGURE 5

scaling circuit is taken from the right hand plate of the 6SN7. A switch in this connection, allows the triggers to be interrupted without disturbing the operation of the rest of the circuit.

In the case of positive output triggers, a 6SL7 dual triode multivibrator and a 6F6-G power output tube allow a neon light and a speaker to be used as audible and visual count indicators. The multivibrator lengthens the short pulse that is supplied to the scaling circuit to one that is more easily seen and heard.

The A. C. voltages required are obtained from a small radio power transformer energized by the 60 cycle 115 volt line. The D. C. voltages are obtained from the 5Y3-GT full wave rectifier and a choke input smoothing system. The voltages supplied to the voltage amplifiers, the 6SJ7, 6SN7 and 6SL7 multivibrator, are stabilized by the VR150.

The apparatus was assembled as a self sufficient unit. It proved to be satisfactory for the purpose for which it was designed; that is to drive the scaling circuit shown below in Figure 5, and to give audible and visual indications of count rate. Its reliability was indicated by its complete satisfactory performance on a separate research problem. The convenience of completed scalers, developed later in the research, limited its use to initial problems.

Binary Scalers. The voltage transient produced by any detector may be amplified and shaped to a point where it will reliably actuate an electromechanical register. All detectors can detect radioactive disintegrations faster than the fastest register can record them; and, if no buffer circuit is placed between the register and the detector, only very slow rates will be recorded with any accuracy. For example, the Cyclotron Specialties Register No. 401-A can respond to only about 60 evenly spaced counts per second, and even less can be recorded by less expensive types. The uneven spacing of the statistical radioactive counts will lower the rate even farther of such a device. To permit fast counts to be accurately recorded, electronic methods have been devised to divide the count by a known factor, and, thus, impose only a fraction of the detector's rate on the electromechanical register.

Considerable demands are placed upon the electronic circuit which makes this division. It must be capable of functioning over a very wide range of rates. The minimum rate may be quite low—counts as low as one or two per minute being encountered. Extremely active samples, on the other hand, may give rates of many thousand counts per minute. The circuit must give accurate division throughout this range. The circuit must be stable; that is it must not add spurious counts by independent operations of its own. It should not require too wide a variety of component sizes. Last, and related to the item just mentioned, the

circuit must be able to operate reliably after the tubes and other parts have aged and changed somewhat in their characteristics.

The scaling circuits used in counters, in general, can trace their origin to the trigger circuit of Eccles and Jordan. Other types of division circuits are known, and are widely used for division of cyclic voltages (72). Adaptations of this type of circuit have been suggested for the division of radioactive counts (73). These adaptations have not achieved any popularity for this purpose, however, probably due to the inability of such devices to respond accurately to the wide rates possible when counting radioactive samples.

The Eccles-Jordan circuit and most of its modifications provide division by two, and are called binary circuits. If more than one circuit is used, division will be accomplished by powers of two. The circuit requires two or more vacuum tubes in its construction. The use of more than two tubes only increases the reliability of the assembly. In the binary circuit, two tubes are connected in a manner that requires one to assume a condition that differs widely from the state of the other one. A single voltage trigger will reverse the relationship of the two tubes. A second trigger again reverses the relationship to that existing before the first trigger. If the next circuit is made sensitive to only one of the two transitions, division by two has been accomplished. Cascading circuits permit division by a factor of 2^n.

The original Eccles-Jordan circuit was not stable, and Wynn-Williams (74) found that gaseous tubes were much more reliable than high vacuum tubes. The separation between the two states is much easier in this case, because the gas tube is either ionized, and, therefore, conducting strongly, or non-ionized, and in the latter case, non-conductive. The gas ionization and deionization is not an instantaneous process and, thus, limits the maximum count rate possible. The Wynn-Williams circuit has other disadvantages, as well, but, in general, is more stable and easily constructed than many of the high vacuum binary circuits (75).

Many circuits based on high vacuum tubes have been proposed (76 - 82). Their reliability in most cases was questionable (83). Perhaps the most popular binary circuit now used is an adaptation and simplification of a circuit of Stevenson and Getting (84) by Higinbotham (64). The signal causing the reversal of the stage is fed only to the tube requiring the trigger; the other tube is kept isolated until reversal is almost complete. The Higinbotham circuit requires only one dual triode and one dual diode instead of the four pentodes required by the original circuit.

The maximum rate at which such a circuit will respond can be made quite fast (82). As in the case of the multivibrator, the role of transition from one state to another depends upon the high frequency response of the tubes used in the circuit. Pentode amplifiers are used in the very fast circuits, and the compensation methods described in

the section on the synchroscope amplifier may be used.

Certain disadvantages are inherent to binary circuits. Some circuits are critical in that they require precise matching of the components (64). The probability of circuit failure of the Higenbotham type is so high that at least one manufacturer made the whole circuit removable for ease of replacements. The most troublesome feature is the creation of division factors that are not easily integrated into a decimal numbering system. If counts are recorded that are not exact multiples of the scaling factor, the additional counts in the scaling circuit are inconvenient to totalize and to add to the divided counts recorded by the electromechanical register.

Preliminary experiments with high vacuum scaling pairs indicated that they were rather critical in their operation. Experiments with the Wynn-Williams scaling circuit, on the other hand, gave immediate success. They were, therefore, used in the first scaler despite their limitations. The design developed is shown on the lower half of Figure 5. The input triggers from the amplifier are first placed on the left hand triode of the 6SL7 which is a phase splitter. This triode is another place where the pulse can be changed to either positive or negative. The voltage fed to the right hand side of the tube is slightly shaped by the 0.01 mfd and 20 K coupling circuit. The signal is somewhat attenuated by this process, and is amplified by the right hand side of the tube. To present a

constant voltage to the first scaling circuit, the sharpened voltage transient is then used to trigger a multivibrator, the 6SN7. The multivibrator produces a positive transient of about 50 microseconds and uniform height to trigger the first pair of Wynn-Williams tubes which are two 884's to the right of the 6SN7.

One of the two 884 tubes comprising the first Wynn-Williams circuit will conduct. The conducting tube creates a cathode bias by virtue of its plate current flow through the 3.3K resistor that is common to both cathodes. The bias is high enough to prevent ionization and conduction of the other tube of the pair. Under this static condition, the plate potential of the conducting tube will be very low, resulting in a difference between cathode and plate potential of only 10-15 volts. The other plate will be at the effective plate voltage supply since the tube is not drawing any current through its plate resistor. Therefore, the 0.1 mfd capacitor which is connected between these two plates of different potential will acquire a charge.

A trigger from the detector is now fed into both grids. The grid of the conducting tube has little effect upon the plate current of this tube. The positive voltage, however, can cause the non-conducting tube to conduct. This starts plate current flow through the plate resistor which lowers the plate voltage. Since this changes the potential gradient to which the 0.1 mfd capacitor is connected, the capacitor discharges through the plate resistor of the

initially conducting tube. This discharge lowers the plate voltage of this tube to such a point that it no longer conducts. The discharge current decreases exponentially, and soon the plate potential is again high enough to cause conduction in the initially conducting tube. However, the plate current of the newly conducting tube creates a bias that prevents ionization until a second trigger again reverses the situation.

The plate of the right hand tube of the first pair becomes positive and negative when it is triggered by two detector transients. The second pair which is coupled to one plate of the first pair will be triggered by a negative and positive pulse. The second pair is not triggered by the negative transient and is in this manner only triggered once for two input voltage pulses.

The third pair is triggered by the change in one plate of the second pair, and yields a positive and negative voltage change at its plate. This occurs once for each eight pulses generated at the detector.

The voltage that is used to actuate the Cyclotron Specialties Co. Register is generated by the 6SN7 multivibrator and amplified by the 6K6-G power amplifier. The insensitiveness of the multivibrator to negative pulses completes the selective use of the voltages occurring in one cycle of the last scaling stage. A multivibrator is used to provide a pulse of the optimum length—about 10,000 microseconds—for the register.

The count that is in the scaling circuit at the time that the count is stopped is determined from the neon lights that are placed in the plates of one tube of each scaling pair. They are arranged so that they will light when a pair has completed half of its cycle, and thus has 1, 2 or 4 counts in addition to that indicated on the register. As the pairs always start from a given state, the shift in state in each pair will cause one plate to go quite positive when half of the cycle is complete. This voltage change is large enough to cause the neon light to fire, indicating the occurrence of the half cycle. The standard zero state is established by connection of the grid resistors of those tubes that are to conduct at zero count to a positive source. All lights will be extinguished at this point. The sum of the subscripts on the neon lights in Figure 5 is added to the register count when a count is stopped.

The power supplied to the circuit is furnished by transformers and a high vacuum rectifier. The plate supply to the 884 tubes is delayed to allow the cathodes to reach operating temperature before plate potential is supplied. The power demand for the 884 scaling pairs is high; the heater power is large; and the plate current is high because the tubes drop to such a low effective resistance while conducting.

The circuit has proven to be fairly reliable under conditions of relatively low count rate. As had been mentioned, it was satisfactorily used in a separate research problem.

Counters with n-scale rings

Theoretical. The binary circuit has the disadvantage of providing scaling factors that are not easily integrated with the decimal system used on most electromechanical registers. Scaling factors of powers of ten simplify the arithmetic of count recording considerably, and thus, much effort has been directed towards the development of scales-of-ten. Lifschutz, in an article on vacuum tube scaling circuits, pointed out that the binary circuit was capable of operation at scaling factors other than powers of two (85). A scale-of-ten can be altered in many ways to yield a scale-of-ten (86). This scheme was adapted by Potter to a scale-of-sixteen using four binary circuits as the base for commercial decade counters (87) and this system has been subsequently used by others (88). At a count of eight plus two, all four circuits are internally reset to zero, so that the scaling factor is reduced to ten. Scaling factors other than ten can be obtained in this manner (89) but ten is the most useful. If only one dual triode is used per binary stage, the circuit is very economical of parts and power.

The altered scale-of-sixteen has several disadvantages. The neon lights which indicate the residual count in a scale cause problems. If direct indication of the binary stages is used, additions are necessary. If direct single light indication is used, a resistor matrix is required which causes direct indication of the operation

of the scaling tubes to be lost. Scaler operation can be checked by observing lights at slow count rates. This can be done best, however, only when direct connection is possible. Another disadvantage is that the maximum count rate may be decreased. In one such scale, on which full information is available, the count rate was reduced from 125,000 counts per second to 30,000 counts per second by the feed back circuits needed to adapt a scale-of-sixteen to a scale-of-ten (90). These circuits are made by many manufacturers as completely replaceable units; this should not be necessary in a completely satisfactory unit.

Ring circuits or combinations of ring circuits and binary circuits have been used for decimal division of fast transients. The ring circuit can be considered an expansion of the binary circuit. Two tubes react in the ring in a manner similar to that of the two tubes in a binary circuit. By this sort of a reaction to a transient, a state, possessed by only one tube in the ring at a time, becomes rotated around the ring. Transients equal to the effective number of states in the ring are required to cause complete rotation of the ring to any arbitrarily chosen initial state. The electrical change that occurs at this tube is large when the tube enters the unusual state. Since the tube enters this state only once per ring rotation, the subsequent circuit will be triggered only once per revolution.

This method was first used by Wynn-Williams with gas filled tubes, but did not prove satisfactory (91).

Most ring type high vacuum tube scaling circuits have been limited to a maximum of five, because of the additional demand encountered when each subsequent tube is added to the ring (92). Consequently, to obtain a scale-of-ten, binary circuits are usually incorporated in various ways. Lewis described a method that used a scale-of-two before a ring of five (91). Other methods have been described using the scale-of-two with ring circuits to get a decade scale (93 - 98).

A circuit that combines this type ring and binary scaler has been described by Sharpless (92). Each digit in the decade is assigned a binary circuit. The orientation of the binary scale is the distinguishing change in the ring.

This type ring has several advantages. It is expansible and can be made in any size up to at least twenty. It is not much more demanding of parts and tubes than others that have been described (93) and it can operate under a wide range of conditions. It also has the recommendation of successful use in the Eniac computer, which is a device that demands high accuracy in division. In addition, the pair per unit of the scale also permits direct observation of each portion of the scale by the indicating neon lights. Since this circuit was found to be easy to construct, and very satisfactory in operation, it will be more fully described in the experimental section.

Construction. Although the Wynn-Williams scaler was satisfactory for counts within its range, the speed that a

high vacuum tube circuit possesses make these more desirable in a scaler. As laboratory experiments with binary circuits had not been too satisfactory, work was started on a completely contained scale-of-eight which assembled the first power supply, the amplifier, and the Wynn-Williams circuit on one chassis.

The publication of Eniac circuits at this time, with its claims of extreme reliability when made with parts of normal tolerance, seemed to indicate that this circuit might be better than those that have been previously tested. A test ring of five was made and found to be quite stable and to function accurately.

The test ring indicated that best results were obtained when the ring was driven with a pulse of one microsecond duration. The driving circuit published for use with the computer circuit seemed to be somewhat complicated and too demanding for use in an assembly of as limited services as are available in a scaler. Therefore, simplifications were made in this circuit. The new driving circuit uses a multivibrator that generates a constant voltage pulse of short duration. As this triggering voltage is placed into a ring at a point of relatively low impedance (A.C. resistance), a power amplifier is used to amplify the power contained in the voltage output of the multivibrator. The plate supply voltage furnished to the scaler and the driving circuit could be varied from between 75 and 210 volts without changing the scaling factor or its reliability.

The range is probably larger, but this represents the voltage that was conveniently available at the time. The normal commercial tolerance (plus or minus 10%) of parts did not seem to change the operation of the circuit.

Although the data described was obtained with a constant source as generated by a pulse generator, this fine stability was maintained when the scaler was triggered by Geiger tube transients which were enlarged by an amplifier designed for use with the scaler.

As a result of the satisfactory operation of the test ring of five, two rings of five were incorporated into the assembly originally designed to integrate the three individual parts of the original "scaler". At that time, the convenience of a scale-of-ten was recognized; the special limitations of the partially completed assembly would not allow this factor of ten to be used. As this circuit is essentially the same as the more widely used scales-of-ten, it will not be discussed in detail. It differs from the more widely used scale-of-ten in two ways; in size of the scale cathode resistors which are a function of the scaling factor; and in the fact that it has only a single range high voltage supply. A scale-of-one hundred using two rings of ten based on the Eniac ring circuit proved much easier to use.

Because the operation of the decade scale is best explained with its other parts, the whole scaler will be described. The complete circuit used is shown in Figure 6.

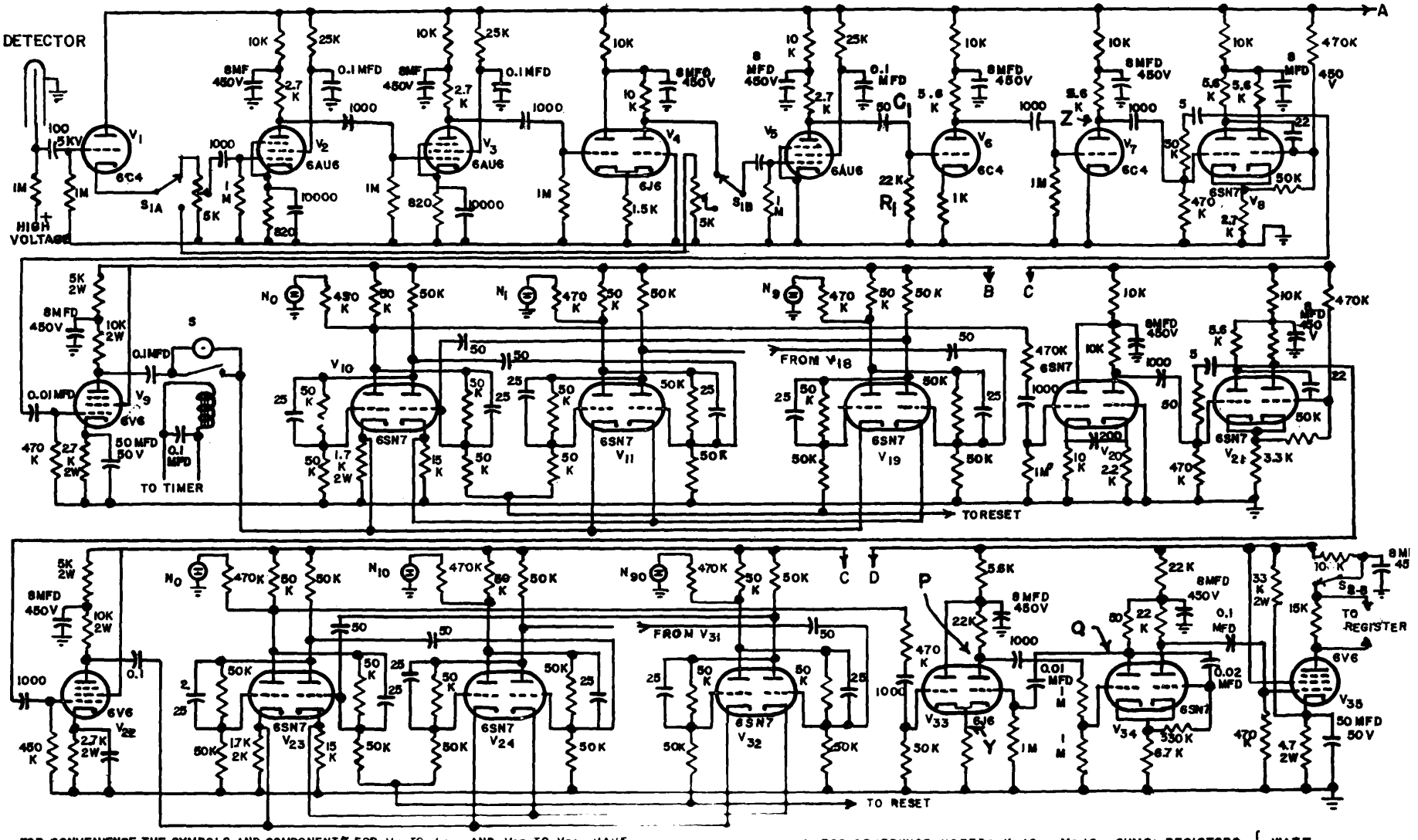
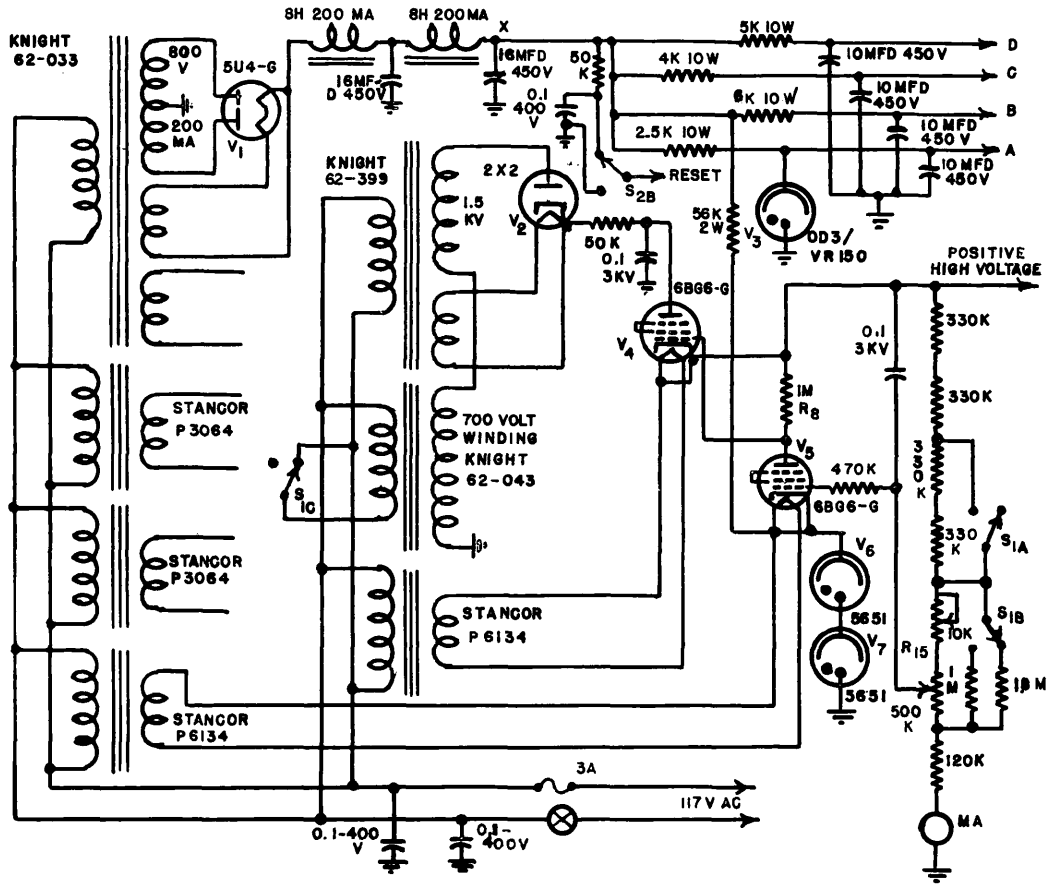


FIGURE 6

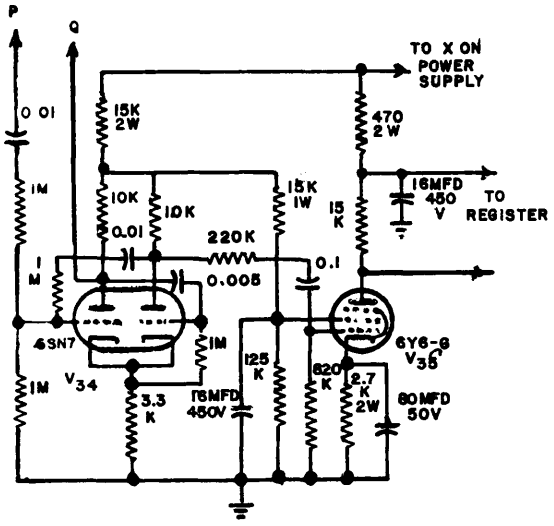
FOR CONVENIENCE THE SYMBOLS AND COMPONENTS FOR V_{12} TO V_{18}, AND V_{25} TO V_{31} HAVE BEEN OMITTED. THEY ARE CONNECTED IN THE RING AS ARE V_{11} AND V_{30}

UNLESS OTHERWISE NOTED: K=10 , M=10 OHMS; RESISTORS $\frac{1}{2}$ WATT
CAPACITORS IN MMD AT 400 V.

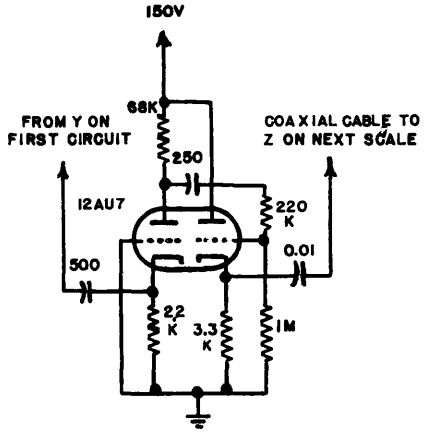
SCALE-OF-100



POWER SUPPLY SCALE-OF-100

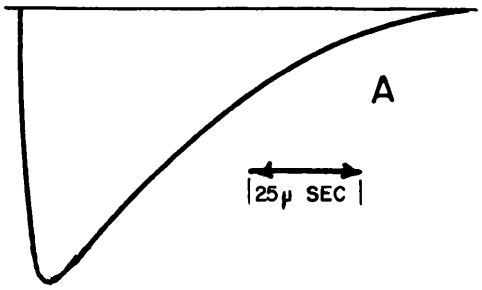


VEEDER ROOT REGISTER DRIVING CIRCUIT

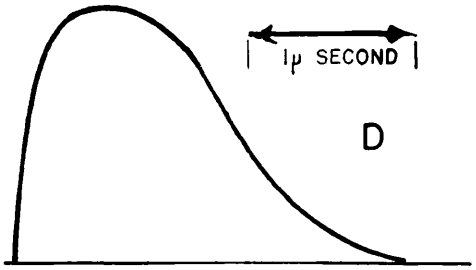


CIRCUIT COUPLING TWO SCALES-OF-100

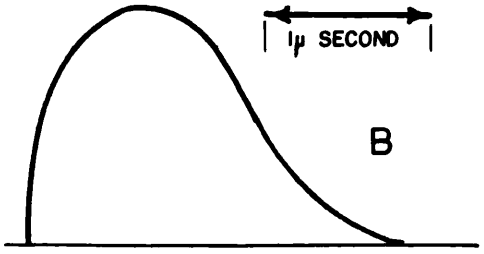
FIGURE 7



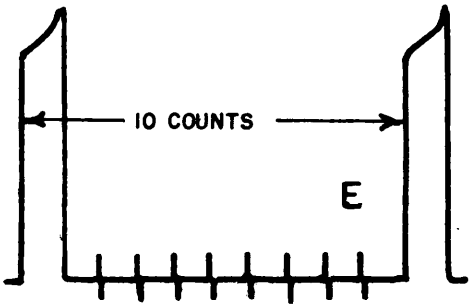
GEIGER TUBE PULSE AT THE CATHODE OF V_1



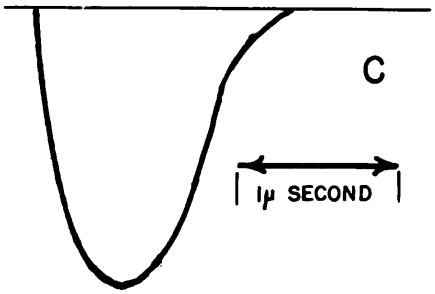
TRIGGER TO SECOND DECADE, I.E. THE SIGNAL AT THE PLATE OF V_{20}



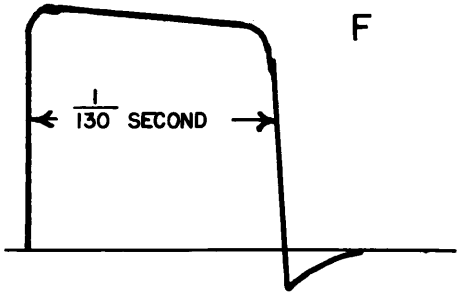
SHAPED PULSE USED TO TRIGGER FIRST DECADE DRIVING CIRCUIT, VOLTAGE AT THE PLATE OF V_7



OUTPUT OF A DECADE WHEN TRIGGERED AT A CONSTANT RATE (1000 PER SEC.)



DECADE DRIVING PULSE, THE VOLTAGE AT THE PLATE OF V_9



VOLTAGE DRIVING CYCLOTRON SPECIALTIES CO. REGISTER

PULSE SHAPES IN A SCALE-OF-100

FIGURE 8

It consists of two amplifiers, two rings of ten, and a high voltage supply. The work on the test ring had indicated that a sharp pulse was necessary to obtain positive operation. Comparison of the pulse requirements with the long duration of a Geiger tube pulse, as shown in Figure 8 indicates that it is necessary to shape the Geiger tube pulse into a form more suitable for the operation of the scaling circuits. The only rapid voltage change shown is that of the initial voltage change, which occurs during the rapid collection of the light and mobile electrons formed in the discharge. The capacitance that is shown by long connecting cables will load the Geiger tube in such a manner that the sharp rise will not be accurately reproduced and will take much longer to reach its maximum voltage. Thus, to permit the pulse to be more accurately transmitted to the amplifiers, and yet, permit the use of reasonably long cables, a cathode follower is placed at the detecting device. The cathode resistance, which is mounted on the main chasis, is also used as the gain control that allows the amplification accorded the pulse to be varied. As the cathode resistor can be relatively small, less distortion, due to frequency discrimination, is also encountered.

The signal, depending upon size, can be placed into the amplifier in one of two places. If the signal is small, it can be amplified by three amplifier tubes in addition to those that are in the circuit at all times. The three tubes are V_2, V_3 and V_4. Tubes V_2 and V_3 are conventional

resistance coupled amplifier tubes with linearity of response aided by the choice of small resistors for the plate circuits of both tubes. The third amplifier used, V_4, is a cathode coupled grounded grid amplifier (99). Here, the signal is placed into the cathode circuit instead of the grid, and at the plate where the amplified signal is recovered, no change in polarity occurs. The low resistance of the cathode circuit will load down V_3, the last pentode amplifier, if the signal for the grounded grid amplifier is taken directly from this plate. The signal is, therefore, buffered by a cathode follower in the left hand side of V_4 and both functions are handled by a single envelope. The output from this amplifier is of the same polarity as that furnished by the cathode follower V_1, and can be substituted for it when additional amplification is required.

All signals, either large or small, are amplified by V_5, a conventional pentode amplifier. Since the signal is negative, the tube can be effectively operated at zero bias to prevent undershoot (trailing pulses of opposite polarity caused by the reaction of the circuit to transient waveforms). If the signal is large, some chopping off of the top of the waveform occurs, but this does not seem to materially change the operation of the subsequent circuits.

The signal is coupled to the next circuit by means of the only short time constant circuit used in the amplifier train, R_1 and C_1. The change that occurs can be seen by comparing the Geiger tube pulse duration of section A in

Figure 8 with the pulse B on the same figure that is shaped from it. This circuit lowers the amplitude of the pulse that is obtained, and it must again be amplified by V_6 and V_7. Two tubes are used since it is desired to have a positive pulse for further triggering.

The output from V_7 is then used to trigger a one shot multivibrator V_8. This tube forms the voltage that drives the scaling ring. V_8 must be driven by a positive pulse, this being the reason why so much care is given to the polarity of the signal when amplified by the tubes of previous sections. This multivibrator, when triggered, will produce a pulse slightly over one microsecond in length and of relatively constant amplitude. The multivibrator thus serves as a buffer between the varying voltage furnished by the amplifier and the ring, which is best triggered by a constant voltage. The multivibrator has also another function when used with a proportional voltage yielding detector; it will not allow any trigger voltage to be furnished to the ring unless the voltage driving the cathode follower is larger than that required to cause the left hand triode to conduct. Larger signals do not cause the output to increase appreciably, thus giving the ring a chance to operate under almost constant driving conditions.

The voltage obtained from the plate of the right hand side of V_8, however, does not have two necessary characteristics. A trigger must be of negative polarity; and the driving pulse which must be placed into a relatively low resistance

should contain relatively large power. Amplification of the signal from V_8 by V_9, a power pentode, solves both problems. The output from V_9 is used to trigger the ring. This also has proven to be the most satisfactory place to interrupt the signals which start or stop the recording of disintegrations; therefore, this signal is transferred through the count switch. A timer, if available, can automatically start and stop the count rate by energizing the relay paralleling this switch.

The rings used are a decade version of those described by Sharpless. The first ring consists of tubes V_{10} and V_{19}. When not triggered, ten of the twenty triodes comprising the ring will conduct, one triode in each envelope As the circuit is shown connected, nine of the left hand and one of the right hand triodes will conduct. This latter tube is called the "odd" tube for the sake of convenience.

The left hand plates that are conducting will cause the plate voltage existing at these plates to be too low to maintain the discharge of a 0.04 watt neon bulb. The interpolation lights are, therefore, connected to these plates. The odd tube contains the non-conducting left hand triode, and therefore, it will be at a potential high enough to cause the neon bulb to light. Thus, the operation of the ring which depends upon the odd state location is followed. When operation is stopped, these lights also indicate any residual count not yet recorded by an electro-mechanical register.

Conduction by only one triode in each envelope is due to the differences in grid voltage of the two triodes as a consequence of the connection of each plate to the opposite grid by the resistor network. Conduction by one side increases the negative voltage at the grid of the other side enough to cause this side to cease conduction.

An attempt by two right hand sides to conduct is suppressed in the cathode circuits. All the right hand cathodes and all the left hand cathodes are connected in two separate circuits. The resistance through which the plate current of the left hand sides must flow is only one ninth that of the right hand sides. An attempt for two right hand sides to conduct will thus increase the negative voltage on the right hand sides enough to suppress this condition.

The injection of a single pulse into the left hand cathode circuit from the pulse driving circuit causes the odd tube to rotate one tube in the ring. As an example, if V_{10} were the odd tube, one pulse would cause V_{11} to be the odd tube. This change may be explained by considering the following reactions. The negative pulse will drive all left hand grids in a positive direction by cancelling cathode bias. This will have little effect upon any of the tubes, except the odd tube, because all but the odd tube have this side conducting and, therefore, must have their grids at almost zero potential with respect to their cathodes. Any attempt to drive these grids farther positive will drive them positive with respect to their cathodes. Since the

driving voltage comes from a source of high internal resistance, most of this positive voltage will be lost in the source, due to grid conduction. The left hand grid of the odd tube, however, will not be so limited because this grid is at a much lower voltage and can be used to drive the left hand triode conducting. This, in turn, drives the right hand side of this odd tube to a state of non-conduction by a transference of the large negative change at the left hand plate, almost unchanged, to the right hand grid by the 25 micromicro farad capacitor connecting these two elements. The large positive increase at the plate of the right hand side of the tube that is leaving the odd state, is transmitted to the right hand grid of the next tube in the ring, causing it to become the new odd tube. A short ring driving pulse is required since it is necessary to remove the negative signal from the cathode of the new odd tube before the positive voltage is removed from the right hand grid.

As it is more convenient for computation purposes for the odd tubes to be in a fixed position in the rings, all the grid resistors of the triode sections that are required to conduct at zero count are connected momentarily to source of positive voltage. This establishes the zero setting of the rings. Once this has been established, all these grid resistors are returned to their normal ground connections. A capacitor is placed at the end of the resistor used to limit the current flow during this operation of

resetting. This allows much higher initial current flow than that of the steady state in order to assure positive resetting. Another portion of this switch removes the plate voltage from V_{35}, since resetting with more than nine residual counts in the scales will cause the register to increase one in count.

The information that the ring has completed a cycle is obtained from the right hand plate of the zeroth tube. The voltage variations that will take place at the plate are shown in section E of Figure 8. This portion of Figure 8 records the variations that occur for more than ten drivings of the ring. The large positive change that occurs when the tube goes into the odd state is used to trigger the subsequent circuits. This is the largest positive voltage that occurs during one complete cycle of the ring.

The device that extracts this information must not load the ring. The loading of a simple oscilloscope will cause a good circuit to stop operating. Unbalance is limited by the use of series isolating resistors in this lead, and also by the use of a cathode follower—the left hand side of V_{20}.

The duration of the square wave that is generated at the plate of V_{10} may be quite long, especially at slow count rates. To be applicable as a trigger for a second decade, it must be sharpened. This takes place in the coupling circuit between the left and right hand cathodes of V_{20}. The right hand side is the grounded grid amplifier.

This short time constant circuit permits only a short, sharp pulse to be transmitted to the next ring driving circuit.

The bias on V_{21}, the multivibrator, in the second decade driving circuit, must be large enough to reject all but the large positive pulse that occurs when the first ring has completed its cycle. Although the negative pulse that follows is large, it cannot cause the multivibrator to cycle, and it follows after V_{21} has completed its cycle.

Aside from this relatively critical adjustment, the second decade driving circuit and the second ring are duplicates of the first.

The information from the second decade is extracted from the zeroth tube in the ten's ring by a cathode follower in the same manner that information from the first ring is extracted. The output from the cathode follower, the left hand side of V_{33}, serves as a trigger for V_{34}, a multivibrator generating a voltage transient. When this is amplified by V_{35}, a power amplifier, the electro-mechanical register advances once.

The operating characteristics of the registers used made certain changes in the output of the cathode follower necessary. The registers used have a certain cycle that must be achieved to permit the maximum possible count rate of the register to be utilized. Electromechanical registers usually have an armature that is drawn to an electromagnet; this motion advances the recording mechanism. The current

for the energization is supplied, in this case, by V_{35}. Once the armature has been attracted, it must be allowed to return to the rest position by de-energizing the magnet. The time that the coil must not be energized in this resetting phase is about equal to that required during the energizing phase. Any shortening of this cycle will result in inaccuracy. Of course, the de-energized time may be any length of time longer than the minimum. This cycle seems to hold for both Cyclotron Specialties Co. type 401A and Veeder-Root form B-124806 registers.

If the multivibrator creates the proper voltages, this voltage will exist half of the time between register impulses at the maximum count rate. In the output from the cathode follower, the positive trigger is followed by a negative trigger by only one tenth the time between register impulses. At slow rates, the multivibrator will have completed its cycle before this negative pulse will occur and thus, the minus pulse cannot interfere. At fast count rates, the negative pulse will occur before the multivibrator will have finished its optimum cycle, and while the negative pulse cannot cause the multivibrator to start a cycle, it can cause it to finish a cycle not as yet complete. If the cycle of the multivibrator is materially shortened, the register will not have had enough time to complete its energization. At a rate that would be the maximum for the register, the energizing pulse would be shortened to one tenth instead of one half of the elapsed time between

register impulses. Under these considerations the register will not operate accurately. Thus, if the multivibrator is to be triggered by the decade positive pulse, the negative pulse must be removed.

A gating circuit was devised to remove the negative pulse. The signal output of the cathode follower is not used directly as the trigger, but is passed by the common cathode to the right hand triode of V_{33}. The positive trigger from this plate is used as the trigger for the multivibrator. The multivibrator, V_{34}, returns a negative signal to the grid of the right hand side of V_{33}, rendering this tube insensitive to any further signals until the tube V_{34} has completed its cycle. As this negative voltage lasts for only half the duration of the time between register impulses at the maximum count rate, no recordable counts are lost. The gating circuit was not included in the original model. Later, when incorporated into the circuit, it increased the maximum count rate by a factor of at least three.

As the maximum count rate that will be handled by the two registers varies by a factor of six and the rated power demand varies by a factor of sixty. A circuit, with no changes in the size of the components that will permit both the count rates and the power demand to be used at their maximum rate, is not easily designed. A circuit, utilizing one register, was designed as a result of the wide differences. The circuit that is shown in Figure 6 is that used

with a Cyclotron Specialties register 401A. This register has a maximum rate of about 60 evenly spaced counts per second. V_{34}, therefore, generates a pulse slightly less than 1/120 second. As only about 0.1 watt is required to operate the register, V_{35} does not have to be of large size. A 6V6-GT is used to limit the variety of tubes that are used in the scaler.

If a Veeder-Root counter is used, the pulse must be lengthened to about 0.05 second, and a larger tube used, because the Veeder-Root counter requires six to eight watts. The circuit evolved by Mr. William Propts for use with this register is shown in Figure 7. It uses a 6Y6-G to furnish this power.

The power supply for the complete scaler is shown in Figure 7. This furnishes heater, plate, and polarizing high voltages. The low plate voltages are supplied by a full wave rectifier and choke input filter. The plate voltages for the amplifier tubes are stabilized by a OD3/VR 150 to assist in maintaining constant amplification. The first multivibrator, V_8, also draws its plate voltage from this source to keep its triggering threshold constant. All the rest of the plate voltages are dropped through series resisters to 150 volts. The scaling rings will operate at a voltage range that extends from 75 to at least 210 volts. Voltages above 150 volts unnecessarily increase the plate current drawn; while plate voltages much less than 150 will cause the neon lights to operate faultily.

The high voltage supply is of the type that has been described under the section on voltage supplies, and will not be further described at this time.

The operation of the scalers, especially the Geiger tubes, has been quite satisfactory. The gain control had no effect on the number of counts registered after it was advanced beyond that point required to cause the rings to be driven. This permits a range of at least six to one in most cases. When the polarizing voltages of the detector were raised, resulting in an increase in the voltage, no large changes in count rate were observed.

The high gain amplifier, utilizing the three extra stages of amplification, have not been thoroughly tested with a radioactive detector. Their sensitivity is that which has been offered by other instruments of the same type, a voltage of less than 0.005 volt causing the scales to be accurately energized. At any setting of the gain control in this high gain amplifier, it will satisfactorily handle a voltage that is at least 100 times as large as that that will just cause it to operate. Voltages too large will cause the scaling operation to be very erratic, due to the overload that is placed on the amplifying system.

If these instruments are used for these detectors, it would be wise to substitute or place in the circuit one of the non-linear amplifiers developed by other investigators that compress the voltage range to a size more easily handled.

The maximum count rate of scalers employing both

Veeder-Root and Cyclotron Specialties Co. registers was determined using evenly spaced pulses of about one microsecond in duration. The pulses were shaped from sine waves that were generated by an audio frequency signal generator. This frequency was continuously monitored by either Lissajou figures or circular sweeps that employed the 60 cycle line frequency as a reference standard.

Using this method of calibration, the maximum count rate of a scale-of-one hundred using a Cyclotron Specialties register is in excess of 3×10^5 counts per minute. With a Veeder-Root counter, the maximum rate is in excess of 54,000 counts per minute. Neither of these rates utilizes the maximum count rate that can be obtained with either a Geiger tube of 100 microseconds resolving time, or the scaling rings of about 5 microseconds.

The statistical rate of a radioactive count will lower the maximum count rate appreciably. Since the scale-of-one hundred using a Cyclotron Specialties register is much faster than the scale-of-one hundred using a Veeder-Root counter, the first will not lose counts at the point where the slower one will. Therefore, the output of a Geiger tube, using radioactive sources to cause discharge, was used to simultaneously test both scales-of-one hundred. A cathode follower was used to distribute the same pulses to both scales.

Using the fast circuit as a standard, the scale-of-one hundred driving a Veeder-Root counter started to lose

counts at about 45,000 counts per minute—losing approximately 1% by 47,000 counts per minute. A similar test could not be made on the fast counters, because no greater scaling factor than 100 and only the Cyclotron Specialties register was available.

The maximum statistical count rate at a scale-of-one hundred using a Cyclotron Specialties register was determined in a slightly different manner. The alterations in a 100 scale using such a register follow: Another tube, furnishing power from a separate power supply was driven by the left hand side of V_{33} in a manner similar to the right hand side of V_{20}. The circuit was the same except for a 500 mmfd coupling capacitor between cathodes. The output from the right hand triode is buffered by a cathode follower to produce low impedance level transmission. This means that the second decade in this circuit not only drives the register, but also drives the equivalent of a crossover network between decade rings. The output of the cathode follower, shaped for further decade use, is then placed into the first decade of another scale using a Veeder-Root counter. Proper adjustment of the bias on the second scale-of-one hundred allows the Veeder-Root counter to have an effective scaling factor of 10,000. The rate at which the Cyclotron Specialties register missed counts was, therefore, easy to detect. Using a 100 microsecond dead time Geiger tube, the loss of counts started at 140,000 counts per minute, increasing to 1% at about 240,000 counts per minute, and to 2% at about 300,000 counts per minute.

GAS PHASE COUNTING

Discussion

The decade scalers, just described, worked very well with commercial Geiger tubes. The 100 mm internal pressure found in most tubes requires walls with enough mechanical strength to withstand atmospheric pressure. With gamma rays, this does not materially decrease the count. However, most artificial radioactive materials are beta emitters, and thus yield much less penetrating radiation. If the material to be counted is outside the tube, thin windows or walls are necessary to permit the radiation to enter. Thin concave bubbles in glass have long been employed to permit electrons freer access through container walls (100). This method is satisfactory only with some materials; the low energy beta form H^3, for example, will not pass through the thinnest practical wall. Window counters are inefficient; at best only 3% of S^{35} or C^{14} radiation is counted by a commercial thin end window tube (101).

The weak radioactive emitters would be much more easily counted if they did not have tube walls to penetrate. Several early papers incorporated H^3 as water vapor into Geiger tubes (102 - 103). No operational data on such tubes is given. Powell and Reid found that gaseous H^3 could be used with butane; however, the limit of H^3 concentration was low (104). Other workers have used mixtures of H^3, ethyl alcohol, and argon; although, in this case, a quenching circuit was necessary (105).

Due to the interest in C^{14}, work has also been done on gas phase counting with this material. Brown and Miller used carbon dioxide as the C^{14} carrier (106). Carbon dioxide alone could not be used because it forms negative ions by two electron capture at the cathode, and thus, maintains discharge. Adding carbon disulfide to the mixture permitted this difficulty to be overcome, although a Neher-Pickering circuit was needed to achieve successful operation. This technique has been further developed by Mann and Parkinson who point out several refinements they found necessary (107).

The introduction of solid radioactive materials into the sensitive volume of a counter working in the proportional region is possible (108). Continuous flow of a gas—usually methane—through the chamber at atmospheric pressure is used (30). Here, however, the self-absorption of the radioactivity not emitted close to the surface of the particles of the radioactive containing material causes loss of counts. Gas phase proportional counting, therefore, has been proposed for such materials (109).

No gas phase Geiger tube technique has been proposed for S^{35}. Consideration of the known gas phase methods shows that none is completely satisfactory. Most scalers are designed to operate only with self-quenched Geiger tubes and do not have quenching circuits. A system that would operate as a self-quenched gas phase Geiger tube would thus have wider application. The proportional techniques have several disadvantages. In addition to the problems that

were discussed under the section on proportional counters, they also require voltages usually in excess of those provided by ordinary high voltage supplies. These difficulties were considered when the sulfur gas phase problem was considered, and the research was concentrated on methods that minimized these disadvantages.

Exploratory work

The first technique that had to be evaluated concerned the preparation of Geiger tubes by known techniques which contained normal fillings that would give consistent results. This proved to be a time consuming piece of work. Tubes that would count were readily prepared. However, their initial characteristics differed widely, and the operation of the tube on repeated filling was far from consistent. The latter difficulty, if not corrected, was a problem that would limit the usefulness of the gas phase work, since few counts are of enough value to compensate for the use of a new Geiger tube for each count made.

The location of the source of trouble was made more difficult by the many variables in the operations used to make a Geiger tube. Broadly, the variables may be classified as the vacuum system that is used to evacuate the tube before filling, the gas mixture that is placed into the tube, and the material and shape of the tube assembly.

Filling systems. The vacuum system used to fill the tubes underwent many changes. The first system used ordinary

glass stopcocks and a Hyvac rotary oil pump to provide the exhaust. Although several tubes were obtained with this system that would count for a brief period, the system had so many leaks and was so limited in ultimate vacuum that it was soon abandoned.

The second system was much more elaborate than the first. The high vacuum was produced by a three stage D. P. I. GF-25W oil diffusion pump, which was then backed by a Cenco Megavac oil pump. The Hyvac pump was used only to remove the organic vapors from the system to prevent contamination of the high vacuum producing system. The apparatus was all glass, with pyrex stopcocks of the mercury seal type. The vacuum obtained with this system was high—approximately 10^{-6} mm of Hg.

The first tubes prepared on this system operated and, due to good sealing techniques, would maintain their characteristics for a long period of time, if they were sealed off. Almost all were faulty in that their count rate did not remain constant over any range of terminal potential. They were obviously of poor quality since one of the reasons why the Geiger region is used so extensively is due to the lack of sensitivity to terminal voltage variations when the tube is properly made. To illustrate the character of the results that were obtained at this time, the least increase in count that could be determined for a tube for a change in 100 volts is given in the following table. The values were chosen at random from the large

amount of data recorded. The slope that is claimed for commercial Geiger tubes is usually less than 5% for organic quench type.

Table II. Variable behavior of early Geiger tubes.

Run number	% slope	Run number	% slope
5	no plateau	55	7
7	10	67	20
17	10	73	11
35	16	75	66
45	9		

As the slope resulting from data obtained from Geiger tubes prepared on the second system was not satisfactory, the method and materials used in the manufacture were changed to see if the cause of the trouble could be located. The vacuum system was left to the last since this second system gave a very good vacuum.

Geiger tube components. The tubes that were filled underwent many changes. One of the tube components that underwent the most change was the cathode. If the work function of the cathode is too low, the positive ions may be able to generate additional electrons when neutralized at the cathode during the time when the tube is again becoming fully sensitive to additional ionization. Silver, graphite, stainless steel, copper and brass were each employed as cathodes. The thin nature of the deposit and

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the inability to properly outgas chemically deposited silver made this cathode surface less desirable than the others. Colloidal graphite with the glue binder removed has little adherence to the tube on which it has been sprayed; therefore, this cathode surface was little used. The other cathode materials were given many treatments; they were etched with reagents that would attack the surface; they were internally bored on a lathe to give as smooth a bore as possible with this technique.

Microscopic investigation of several lengths of the tungsten wire that was used for anodes indicated that the wire was relatively smooth. Since this material is almost universally used for anodes in Geiger tubes, no further consideration was given this raw material. The technique of anode glowing was used on almost all tubes. In this procedure, the tube is first evacuated and then an electrical current is allowed to flow through the wire until it is incandescent. This not only outgasses the wire, but also is supposed to burn off any sharp protuberances on the surface (110).

The material of the envelope was glass in almost all cases. The difficulty of outgassing tubes that had organic insulators as well as the slow decomposition of the organic insulators at low pressure on aging, makes glass a much better insulating envelope. The thermal instability of large metal-to-glass seals makes metallic tube bodies much harder to use than those requiring small glass-to-metal wire seals.

Tube treatment and fillings. The tube itself, when assembled, was given many types of pre-treatment. All were thoroughly washed once the glass blowing operations were completed. Then they were treated with one or more substances such as concentrated mineral acids, ammonia, or organic solvents before being thoroughly rinsed. Some were further rinsed with redistilled organic solvents such as benzene, ethyl alcohol, dioxan, and so forth.

The fillings that were used were varied to a great extent. The quenching agents used include methyl alcohol, ethyl alcohol, diethyl ether, xylene, and n-amyl acetate. A dry, commercially prepared, raw material was chosen for purification and then redistilled under conditions that produced dry samples for use as quenching materials. The ether was distilled from sodium; the ethyl alcohol was distilled with anhydrous benzene and only the last fraction used; the xylene and n-amyl acetate both form low boiling azeotropes—any water present would be discarded with the forerun; methanol was distilled from C. P. Methanol using only the middle fraction of the distillation. All samples were distilled through a fractionating column that had approximately 10 theoretical plates—the theoretical plates being determined by total reflux with benzene and toluene.

The commercial tank argon used in the tubes was furnished by two different suppliers. To purify this material by removing any reactive contaminants, the gas in some runs was led over magnesium at 600° C. Other inert

gases besides argon were used, namely, helium and neon. The ratio of rare gas to organic vapor, as well as the total pressure of the gas mixture, was widely varied.

Although the residual air in the system was removed by heating both the tube and the system with the flame of a hand torch, it was thought that residual gas still in the tube might be a cause of trouble; therefore, an oven was built to house the Geiger tube so that it could be exhausted at a temperature of 400° C for fifteen hours or more.

Vacuum system changes. Although all of the items that have been listed above were used in an attempt to increase the reliability of counting, no appreciable change in results was noted. This left only the vacuum system to be changed. The need for a change in the system was confirmed by an experiment run on an unusual tube prepared on the system—a tube that had only a negligible slope. The tube was again evacuated and filled to 100 mm with only argon for seven cycles. At the end of the seven fills, the tube was again filled with argon and ethyl alcohol and the slope determined. The filling operations had increased the slope to 13%.

It was postulated that the tubes might be contaminated by either volatile material from the stopcock lubricant or decomposition products from the oil diffusion pump. Alkaline titration showed that the high boiling ester in the diffusion pump had acquired quite a high acidity; thus, this material had decomposed.

A new system was therefore constructed that eliminated both of these possible contaminants. A two stage mercury diffusion pump was substituted for the oil diffusion pump. Dry ice traps were kept between the vacuum rack and both exhaust systems at all times. The stopcocks were replaced by mercury cutoffs, adaptations of a design of Stock (111). The float valves in the two legs were replaced by atmospheric legs, as it was hard to get valves that did not leak. Fritted glass discs were used to allow partial equalization of the pressure between the two legs; otherwise, high pressures caused violent displacement of the mercury.

The first two tubes prepared on this system were excellent, and had low slopes. At the end of filling and exhausting these tubes seven times, the slopes were 1.7 and 1.2 on the tubes. The intermediate runs did not differ appreciably from the first and last.

The fragile nature of this system made continuous use of mercury valving system impossible. The diaphrams developed mercury leaks after a period of use. The large volumes of mercury needed, and the ease with which it could acquire high velocities led to much glass breakage. Thus, while it was possible to get good tubes on this system, it was necessary to replace the mercury cutoffs because of maintenance reasons.

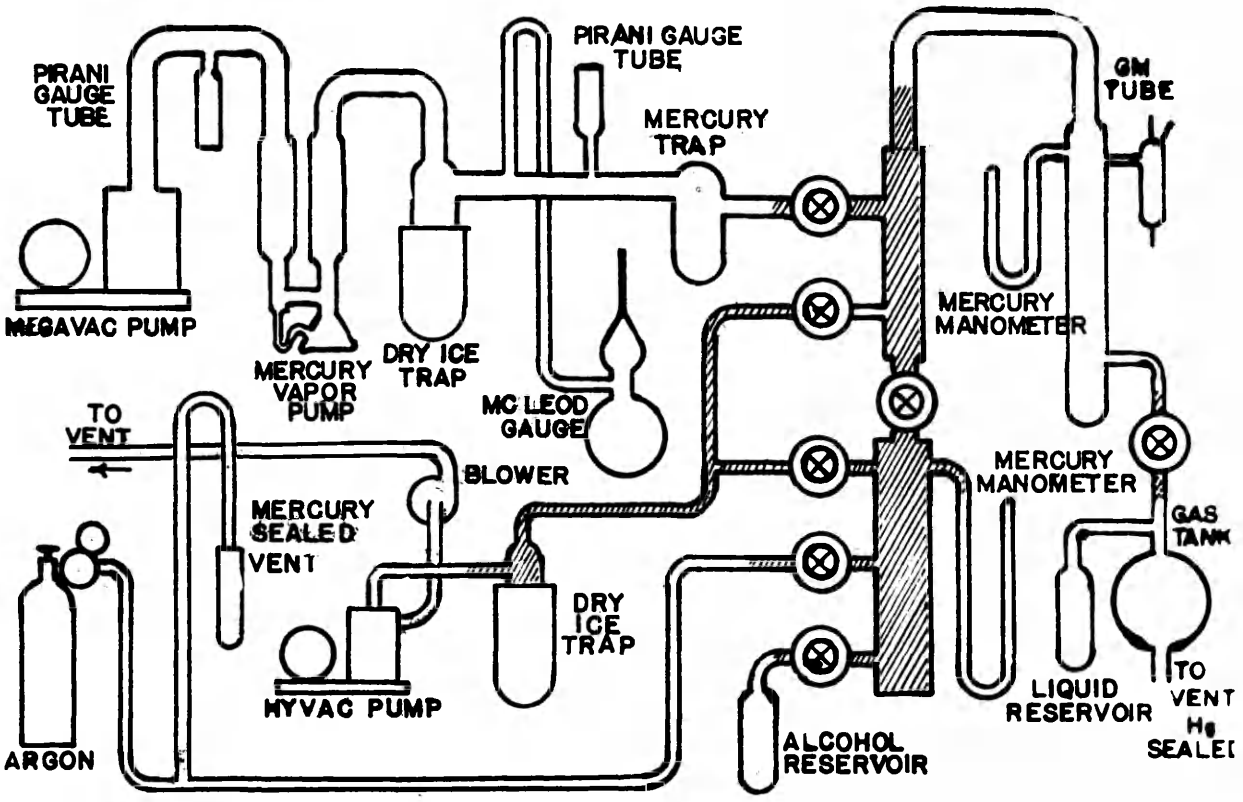
The mercury cutoffs were successfully replaced by brass valves of the type that are used on high pressure gas cylinders. Simple replacement of the stopcocks by these

would have meant that many metal to glass seals would have to be used; so much of the glass tubing was replaced by copper and brass tubing. The copper tubing used to connect the valves to the system was silver soldered directly to the ports of the valve. Silver solder was used to seal all other metal-to-metal joints. Metal-to-glass seals were used only when necessary.

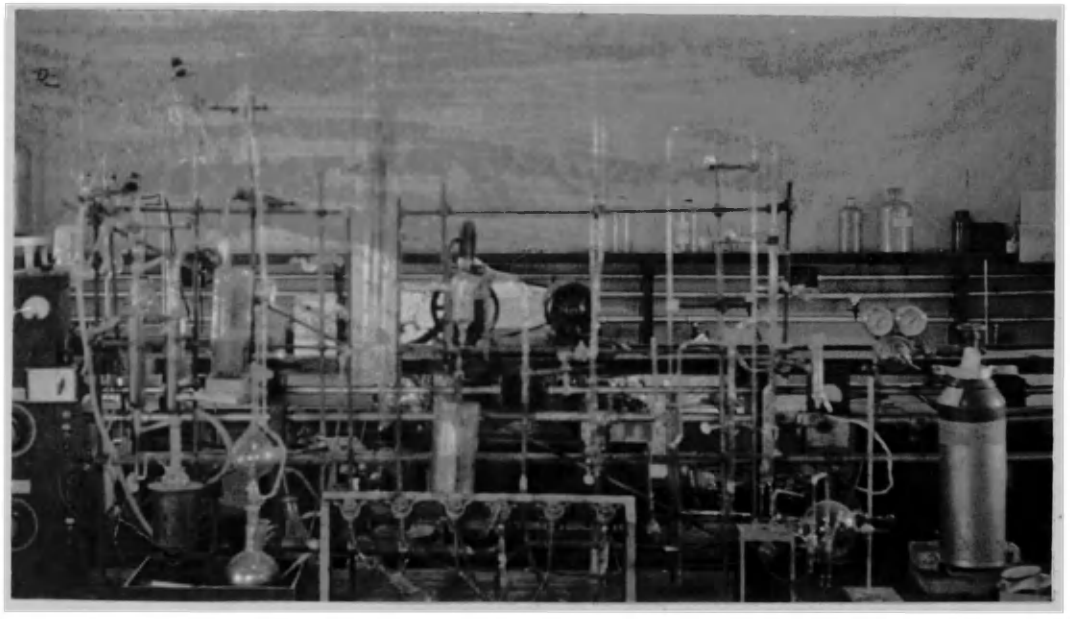
The system was found to function quite satisfactorily in the preparation of Geiger tubes. The average slope per 100 volts obtained from the tubes filled on this system was less than 3% and rarely more than 5%. A diagram of the system is shown in Figure 9 along with a picture of the completed system.

Geiger tube manufacture. The experimental technique that was evolved for making Geiger tubes, which are reasonably simple, requires several steps. A drawing of the tube that was chosen for the gas phase work is shown in Figure 10. Since the radioactivity is placed into the tube, no thin walls are necessary. As many types of experimental mixtures were to be placed into the tube, the tube had to be easy to make because some might be ruined by corrosion or other contamination, and therefore discarded.

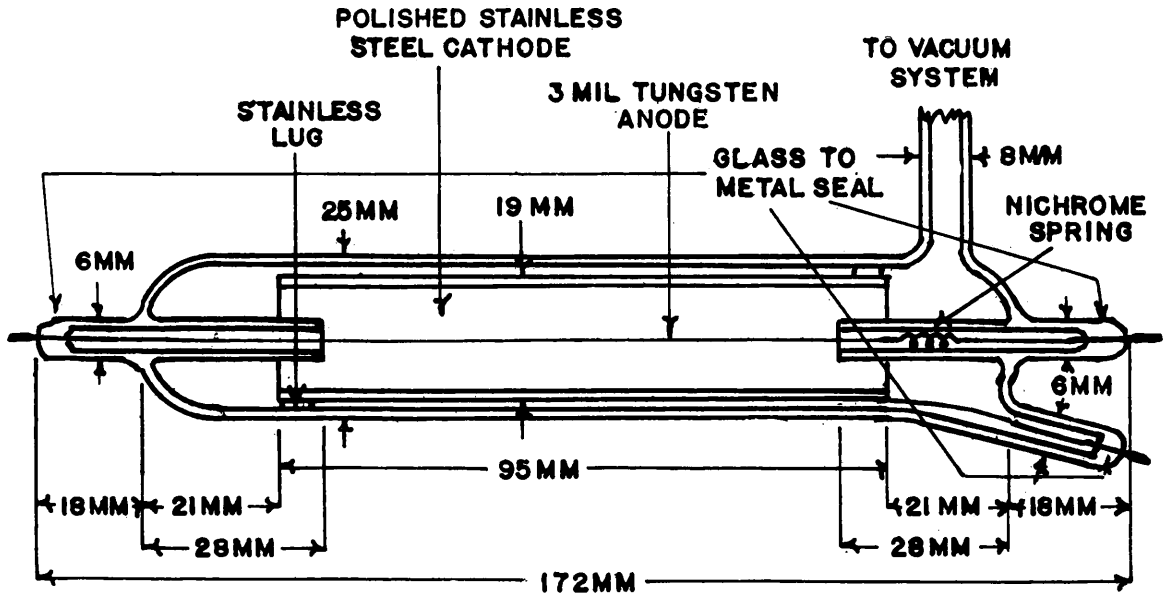
Of the component parts in the Geiger tube, the cathode received the most attention. Etched cathodes used in the beginning of the work gave more reliable results than the non-etched electrodes. Highly polished cathode surfaces gave more reliable results than did the etched cathodes.



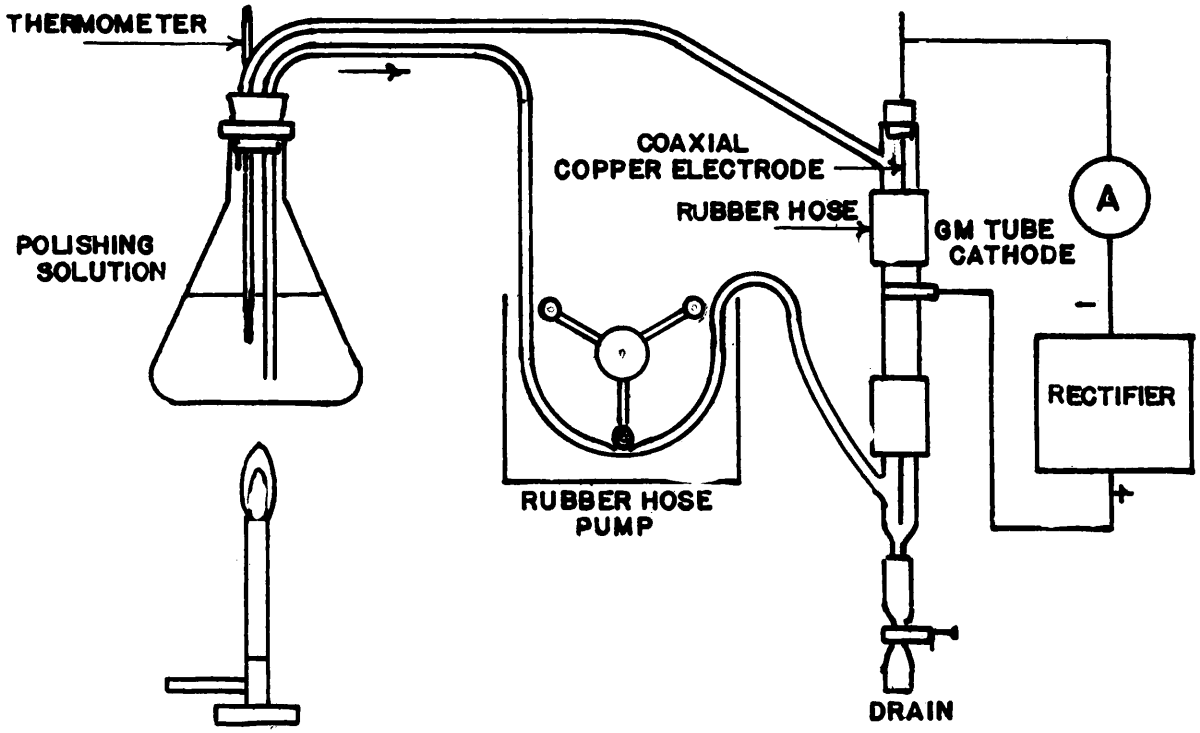
VACUUM SYSTEM



PHOTOGRAPH OF THE VACUUM SYSTEM FIGURE 9



GM TUBE DETAIL



ELECTROPOLISHING APPARATUS

FIGURE 10

This latter fact is probably due to the lack of sharp particles on the surface of the electrode. Stainless steel is quite easily electropolished and it became the standard material for cathodes.

A cathode was prepared in the following manner. A section of stainless steel pipe slightly longer than the finished cathode was cut from the stock. After chucking it in a lathe, a 60 grit aluminum oxide abrasive wheel was used to grind the weld marks from the bore. The bore was then polished with a rubber bonded polishing wheel. After trimming off the excess length, the tube was mounted on a brass expansion mandrel, and the outside machined and polished to a smooth surface. Oil and metal chips were removed by washing in hot detergent solution. The blank was then ready for electropolishing.

The electrolyte employed for the polishing was a mixture of 10% n-butyl alcohol, 60% phosphoric acid, 15% sulfuric acid, and 15% water by volume (112). The cathode was polished by 70° C at 1 S/in^2 anode current density for one half hour. The apparatus used is shown in the bottom half of Figure 10. The pump was used to circulate the electrolyte, otherwise a high concentration of hydrogen bubbles from the cathode built up in the solution and caused poor conductivity. The solution colored rapidly during its use due to the presence of chromium and nickel salts. However, these salts did not seem to hinder the polishing operation. This treatment leaves the stainless

steel in a very passive state which aids its corrosion resistant ability, and in addition, probably makes it a better electrode material.

The cathode was then well washed and dried. Next, six small stainless steel lugs were spot welded on to the outside of the ends of the tube, three on each end. This kept the cathode separated from the glass wall of the envelope, and permitted much more thorough washing of the completed Geiger tube. The cathode was then placed on the brass mandrel and the lugs machined to make a close fit to the bore of the glass tubing that was to be used in the envelope. A beaded tungsten-to-Pyrex seal was then spot welded to the outside of the cathode, which completed the cathode. It was introduced into the tube at the proper point in its construction.

The only anode material that was used was tungsten wire. It has high corrosion resistance, and resists the pitting action of electrical discharges. It was spot welded to the tungsten-to-metal seals and the nichrome spring that is shown in Figure 10.

When the tube body was half made, the cathode was inserted and the other half of the tube body blown. The anode was then inserted with the help of a stiff wire and sealed into the small tubes that surround it at each end of the Geiger tube. The tube was then ready for cleaning. This involved many rinses before being dried under vacuum; two with benzene, one with dioxan, at least two with alcohol,and

finally five with distilled water. If the tube was not to be used immediately, it was filled with distilled water until needed.

Final vacuum system. The vacuum system required very little work after assembly. Much less trouble was experienced with oxide coatings inside the system due to the heat necessary for soldering, if natural gas was allowed to slowly flow through that part that was being silver soldered. The pipes were soldered to a dismanteled valve, the seat ground with 600 mesh carborundum powder, and lapped with rouge after grinding. Even with these precautions, valves sometimes leaked. The finished valves were soldered one by one to the system, the system evacuated, and allowed to stand for at least 16 hours. If any appreciable change could be seen in a mercury manometer connected to the system, the valve was replaced. Later, it was found that acetone, dripped over a suspected leak, caused an increase in the pressure that was recorded by the left hand Pirani guage in Figure 9. Most of the metal-to-glass seals that were used in the system were Housekeeper copper-to-Nonex seals. They were recovered from used klystron vacuum tubes. In most cases the Pyrex tubing was sealed directly to the Nonex of the seal. The silver soldering of these to copper tubing gave some difficulty, because the copper tubing left on the recovered seal was rather short. This short copper tubing of the recovered tubes conduded the heat of

soldering to the metal-to-glass seal. Initially, an attempt was made to try to keep metal-to-glass seals cold by immersing them in water while the soldering was being done. The rapid removal of the heat by this coolant made the preparation of silver soldered joints very difficult. Later, the whole joint was heated, and after soldering, the Housekeeper seal was very slowly cooled to relieve strains in the seal caused by the heating.

The Geiger tube to be filled was sealed to the Pyrex manifold in the right hand side of Figure 9. If the Geiger tube was to be sealed off after filling, the glass tubing connecting the Geiger tube to the system was narrowed down to permit easy sealing.

After all the tubes had been sealed onto the Pyrex manifold, the air was removed by the Kyvac pump. Then the diffusion pump was connected by opening the proper valve, and the tube and vacuum system pumped until the pressure did not rise above 1 micron while the system was hot. The heat was supplied by a hand torch. The anode was "glowed" at a high vacuum by allowing an A. C. voltage (about 1 V) to flow through the wire. The pressure was allowed to again drop below 1 micron if any raise was noted due to "glowing". Then, proper pressure of premixed argon-ethyl alcohol was diffused into the tube from the lower manifold in Figure 9 The slope was determined while the tube was on the rack. Satisfactory tubes were sealed off from the system if this was to be the deposition of the finished tube.

Contaminant trials. Consistent tubes were prepared by the technique using the brass valved system. As this represented a change in several variables, an attempt was made to further isolate the contaminating material from those that were thought to be causing the trouble.

The work in preparing a good vacuum system using brass valves was so time consuming that direct introduction of a possible contaminant into the system was not considered. The testing was done by breaking connections in assemblies that had been sealed completely from the vacuum system.

Two types of apparatus that permit exposure to contaminants were used. The first was a Geiger tube that had two chambers sealed to it, each separated initially from the tube by break off seals. With two seals, a good tube can be re-evacuated, and then, by breaking the proper seal, can be exposed to the material under test as a contaminant while a high vacuum exists. The tube is then brought to the proper pressure of rare gas-organic vapor by breaking the second break off seal.

The second, and more simple apparatus, used only one side arm. A good Geiger tube was used for the experiment. To this tube, a single side arm was attached, and, after filling the tube with a standard 100 mm of 10% alcohol and 90% argon, the seal was broken and the characteristics of the tube determined from time to time. The absence of a high vacuum did not seem to hamper the effect of some of the constituents on good tubes. This is probably due to the long

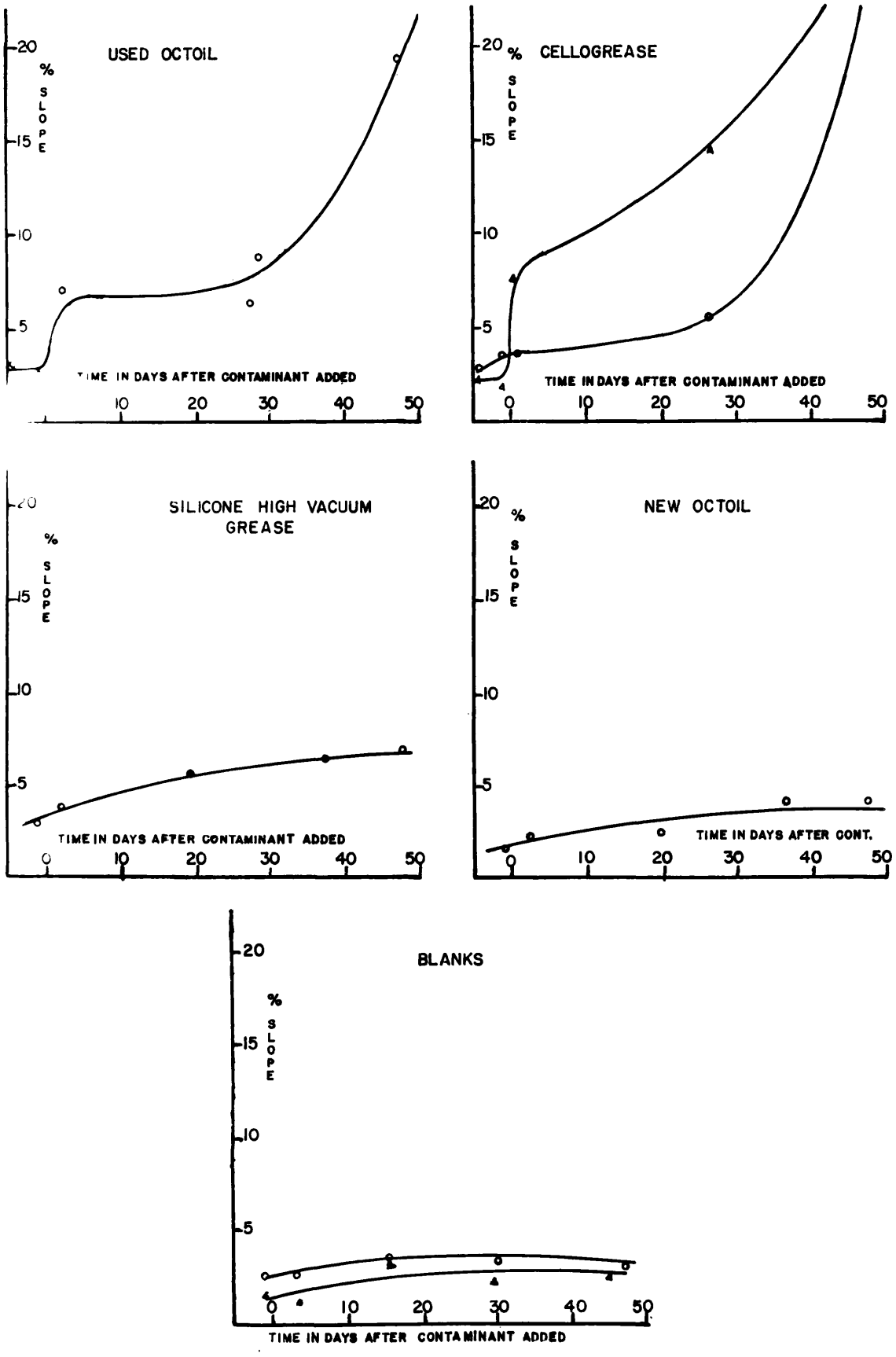
duration of exposure. The single side arms were less complicated and, as a result, less subjected to leaks and other undesirable complications. Single side arm tubes were used for almost all experiments.

The procedure evolved for making test on the possible contaminants is as follows: First a good tube of the sort illustrated in Figure 10 was made, its plateau being checked after completion. The contaminant was sealed into a small bulb at the bottom of a Pyrex tube approximately the same length as the tube. The small bulb in the bottom connects to the larger volume at the top of this 12 mm tube by means of a break off seal similar to that used on rare gas flasks. Above this was placed a hammer made of Pyrex rod several millimeters smaller in diameter than the 12 mm tube bore. The comparatively long side arm is necessary to permit the hammer to acquire enough kinetic energy to break the seal. The small bulb for the contaminant was thoroughly washed, rinsed, and dried in the same manner as the Geiger tubes. A drop or small quantity of the material to be tested was placed in the bulb; the bulb's sidearm sealed onto a dry ice trap that isolated the bulb from the vacuum system; and the bulb thoroughly evacuated. After less than 1 micron of pressure had been maintained for more than 30 minutes, 100 mm of argon were added to the bulb, and the bulb sealed off from the vacuum system. A mixture of alcohol and argon, which would have been less disturbing to the mixture of the Geiger tube when the break off seal was broken, was

not used because condensation of the alcohol in the trap would have contributed another variable.

After the top of the side arm and the hammer were thoroughly washed with solvents and water, the hammer was placed in the side arm and the top of the side arm sealed to the filling tube of a cleaned Geiger tube. The assembly, sealed directly to the vacuum system, was pumped down, again "glowed", and filled with 10 mm of ethyl alcohol vapor and 90 mm of argon. The tube was sealed off from the system after a quick check, and allowed to stand for several days. The starting potential of the tube was determined several times and at least one slope determination was made. If the slope was satisfactory, and appreciable increase in starting potential due to leaks had not developed, the hammer was allowed to break the break off seal. After the break, the slope of the tube was determined from time to time. Assemblies were made with the following materials: used Octoil, Silicone High Vacuum Grease, Cellogrease, and new Octoil. Two blanks were run at the same time. The increase in slope that occured is shown in Figure 11. As can be seen, only used Octoil and Cellogrease gave slopes that increased appreciably with time. No starting potentials of any blank assemblies changed appreciably during the run; thus, the change noticed was probably due to the contaminating effect of the material and not due to air leaks.

To test the effect of exposure of the contaminants on the alcohol partial pressure in the Geiger tube, a



TUBE SLOPES AFTER EXPOSURE TO VARIOUS SUBSTANCES

FIGURE II

separate experiment was conducted. A glass cylinder, equal in volume to a Geiger tube, was sealed to a closed end manometer. A side arm, hammer, and break off tip was sealed to the cylinder. The bulb below the break off tip contained 10 mm of argon in addition to used dioctyl phthalate, the contaminant, that was employed in the contamination trials. Dioctyl phthalate was chosen because it should be the test solvent for ethyl alcohol of the contaminants tested. The cylinder was then evacuated, filled with 10 mm of ethyl alcohol and sealed off from the vacuum rack. No change in alcohol pressure was noticed over a two day period. The cylinder, therefore, was tightly sealed. Next, the break off seal was broken and the pressure recorded periodically over a space of more than one month. Any decrease in pressure due to absorption was less than that visible on a mercury manometer (0.5 mm). This data, therefore, indicates that any changes in the Geiger tube were not caused by loss of alcohol pressure.

The presence of Cellogrease and the used Octoil in the unsatisfactory old system was thus probably the cause of the poor results that were observed over a long period of time. Silicone grease did not seem to contaminate the tube to which it was connected. However, this lubricant, due to its inhomogenious nature, was not satisfactory as a stopcock lubricant.

The tubes that were connected to the new Octoil, and the silicone grease were heated after the period of slope determinations was completed. The sidearms were

heated in the furnace that was designed for the Geiger tubes. The temperatures at which the tubes were heated started at 250° C and finished in both cases at 450° C. The tubes were heated for one hour at the terminal temperature and for an additional hour at each 50° interval between the extremes. A slope was determined after each heating period. The new Octoil did not change the characteristics of the tube even when heated. At the end of the highest temperature heating period, the ester had apparently decomposed, since a solid substance had sublimed from the side arm into the cooler portions of the Geiger tube.

The silicone stopcock grease did not change the slope appreciably until 400° C heating period. At this temperature the slope increased from 5% to 9% and after heating for one hour at 450° C, the slope was raised to 35%. A hard deposit was seen to be present in the bottom of the bulb that had originally held the grease, and a deposit of an oil was noticed in the Geiger tube. The grease had apparently been separated into its volatile and non-volatile fractions.

The failure of the new Octoil to contaminate a tube, even after such drastic heating, is probably due to the lack of air in the system to oxidize the ester.

Experiments with sulfur compounds

Inorganic sulfur compounds. As soon as the technique for making consistent Geiger tubes had been developed, attention was given to the gas phase counting of sulfur. This part of the work was commenced only after the Geiger tube procedure had been perfected, because the variations obtained before would have masked any changes due to the sulfur compound.

The material used to carry the sulfur had to have definite specifications. It must not be corrosive to the stainless steel cathode or the copper and brass of the metal portions of the vacuum system. The sulfur compound must be a gas or a liquid possessing a high vapor pressure. A high vapor pressure is required for two reasons: a high vapor pressure would assure enough sulfur in the tube to yield an appreciable count; and also it was found during the experiment on variations in quenching agents, that materials which were relatively non-volatile, such as n-amyl acetate, were difficult to remove from the vacuum system. The material must not contaminate a Geiger tube when used repeatedly in the same shell. And it should not change the operation of the tube upon aging, because time must be allowed to obtain a count.

Two types of fillings were considered with all sulfur fillings. At first, the material was mixed with the inert gas, argon, and evaluated without any additional quench. If this was not satisfactory, the second filling

employed an organic quench to see whether or not the results could be improved.

The use of a quenching circuit would probably extend the range of materials that could be used in the Geiger region. This circuit is not available on most commercial counters, however, and sketchy experiments in the laboratory indicated that quenching circuits were not easily used. The use of a quenching circuit, therefore, was relegated to the position of last resort—a method to be used if all the compounds tried would not count in the Geiger region without quenching.

It was unlikely that sulfur to be counted would be in the proper chemical form, so only simple compounds were considered so that the gas containing form of the sulfur could be easily prepared. Inorganic compounds were considered first because they are the most easily obtained, especially if quantitative yields are required. Sulfur dioxide, carbon disulfide, hydrogen sulfide, and thionyl chloride which were all easily available for evaluation, passed most of the requirements of the sulfur carrier.

A technique for the introduction of the sulfur compound into the Geiger tubes follows: The Geiger tube used was similar to that shown in Figure 10. It was sealed directly to the upper manifold as is shown on Figure 9, and left there unless further operations on the tube necessitated it being sealed off after filling. The tube was first filled with 90 mm of argon and 10 mm of ethyl alcohol by

the Geiger tube procedure already described. A plateau was determined to make sure that the tube was of good quality.

The tube was then re-evacuated and outgassed by flaming. After this, the sulfur gas was then placed in the manifold by opening the valve above the liquid reservoir until the proper pressure was indicated on the mercury manometer. The proper amount of argon or argon alcohol mixture was then let in from the lower manifold, and at least one half hour allowed for diffusion to take place. The characteristics of the tube were determined while the tube was still sealed onto the vacuum system. As no radioactive materials were used in the screening process, electrons were liberated in the tube by bombarding the enclosed gas with an external radium source.

Sulfur dioxide was the first gas evaluated. As conflicting statements were to be found in the literature (41, 113) as to the effect of this gas on a Geiger tube, it was found necessary to evaluate this substance in the laboratory.

The sulfur dioxide used was prepared by adding 50% sulfuric acid to C. P. potassium metabisulfite. The mixture was gently warmed to complete the reaction. The sulfur dioxide evolved was dried by passing it over anhydrous calcium sulphate, and then condensing it in the gas tank while cooling it with dry ice. After about 100 ml of liquid sulfur dioxide had been collected, the outlet of the tank was

sealed, and the flask allowed to warm up to room temperature slowly. The sulfur dioxide that was in excess was allowed to bubble through the mercury sealed vent, and the final gas pressure was thus limited to about 15 mm of mercury. The large excess of sulfur dioxide when vaporized swept the air from the system. A sample of this gas was completely absorbed in a 10% aqueous solution of sodium hydroxide.

No plateau resulted when 10 mm of SO_2 gas was mixed with 90 mm of argon. Less than five volts caused the tube to go from a state of non-counting to continuous discharge. As smaller quantities of SO_2 would result in the tube having qualities close to that of argon (which will not self-quench) no further work was done on binary mixtures of sulfur dioxide.

The results that were obtained when sulfur dioxide was mixed with argon and ethyl alcohol are shown in the table on the following page. As can be seen from the data in the table, the introduction of even small concentrations of sulfur dioxide to a tube, that otherwise should give good results, causes very poor results to be obtained. The sulfur gas raises the starting potential; however, this raise is not enough to make its supply a problem. The slopes were so poor, however, that the use of SO_2 was temporarily discarded.

Hydrogen sulfide was the next inorganic sulfur gas to be tried. This, the end product of a vigorous reduction

Table III. The influence of sulfur dioxide on Geiger tubes.

Fillings			Tube characteristics	
mm SO_2	mm alcohol	mm argon	starting voltage	best slope/ 100 V
--	10	90	910	3.7
10	10	90	1650	no plateau
5	10	90	1400	67
2	10	90	1350	160
1	10	90	1150	170
--*	10*	90*	930*	1.0*

*Plateau of tube after SO^2 experiments.

of a sulfur compound, is relatively easy to obtain from a sulfide. To prevent its contamination with the air in the generating system, the system was evacuated to less than 1mm pressure before the 50% sulfuric acid was added to FeS to start the gas generation. The gas was collected in the liter storage flask under slight pressure after it was dried by calcium sulphate. A sample of the H_2S used, dissolved completely in a slightly acidic solution of lead acetate. A mixture of 10 mm of H_2S with 90 mm of argon did not have any appreciable range of terminal potential, so its use was also discarded as was the binary sulfur dioxide—argon system.

The mixture of hydrogen sulfide with ethyl alcohol and argon gave the results that are tabulated in the table on the following page. The results show that again small

Table IV. The influence of hydrogen sulfide on Geiger tubes.

Fillings			Tube characteristics	
mm H_2S	mm alcohol	mm argon	starting voltage	best slope/ 100 V
--	10	90	900	1.9
10	10	90	1350	70
5	10	90	1300	23
2	10	90	1260	18
1	10	90	1250	11
--*	10*	90*	930*	4.3*

*Plateau of tube after H_2S experiments.

quantities of the sulfur gas raise the starting potential and the slope of the Geiger tube. Proportionately, the change was not so much as that shown by the sulfur dioxide, but it was enough to warrant the attempt to find a better material to carry the sulfur.

As Miller has found, CS_2 aided in the quenching of a carbon dioxide containing tube. This material was next considered. No attempt is made here to claim CS_2 is an inorganic compound. It is included here because of its simple structure, and for the sake of convenience. The chemistry of carbon disulfide's preparation is not so simple as the two materials already discussed, and it was thus tried after the other two had proven unusable and discarded. As it contains carbon as well as sulfur, and if it would work, this compound might also be used for C^{14} counting.

The carbon disulfide used was a C. P. laboratory sample. It was further purified by distillation. Only the center cut of the material was saved for use. The sample had a boiling range of about 0.1° C and was clear and colorless.

The carbon disulfide was introduced into a good tube with argon. It proved not to be a quenching agent, and, when mixed with ethyl alcohol and argon gave the following results as shown in the table.

Table V. The influence of carbon disulfide on Geiger tubes.

Fillings			Tube characteristics	
mm CS_2	mm alcohol	mm argon	starting voltage	best slope/ 100 V
--	10	90	930	1.9
20	10	70	1700	no plateau
10	10	80	1450	"
5	10	85	1350	"
2	10	87	1200	"
1	10	89	1060	"

Thus, the carbon disulfide itself was not a quenching agent and it interferes with the quenching action of a proven quenching agent. The success of Miller with the carbon disulfide-carbon dioxide mixture, in all probability, was due to the quenching circuit.

Experiments with thionyl chloride indicated that this material would not be satisfactory, due to extreme corrosion

of the mercury and the copper in the system. Other sulfur compounds containing halogen were set aside until other less corrosive materials could be tried.

Organic sulfur compounds. As the investigations described had eliminated most of the simple inorganic materials, consideration was next given to the organic sulfur containing materials. The more simply prepared of these materials, the sulfides and the mercaptans, had chemical similarity to ethers and alcohols—substances that had proven to be satisfactory quenchers. These were thus considered first. The low molecular weight mercaptans do not seem to be as associated as are the alcohols; they boil at a lower temperature and have a higher vapor pressure at equivalent temperature. The higher molecular weight of the sulfides make them boil higher than the corresponding mercaptans—a reversal of the low molecular weight oxygen counterpart behavior. Methyl sulfide has a boiling point close to that of diethyl ether. This relatively high vapor pressure, thus, would make it easy to purge from the vacuum system. The odor of the sulfides, while bad, are not so bad as the odors of the mercaptans, and therefore, were less offensive to handle. For these reasons experimental work was started on sulfides first.

The methyl sulfide used was an inactive sample purchased from the Eastman Kodak Co. and was packaged in a sealed glass bottle. The material was allowed to stand over $CaSO_4$ to dry, but no further purification was given to

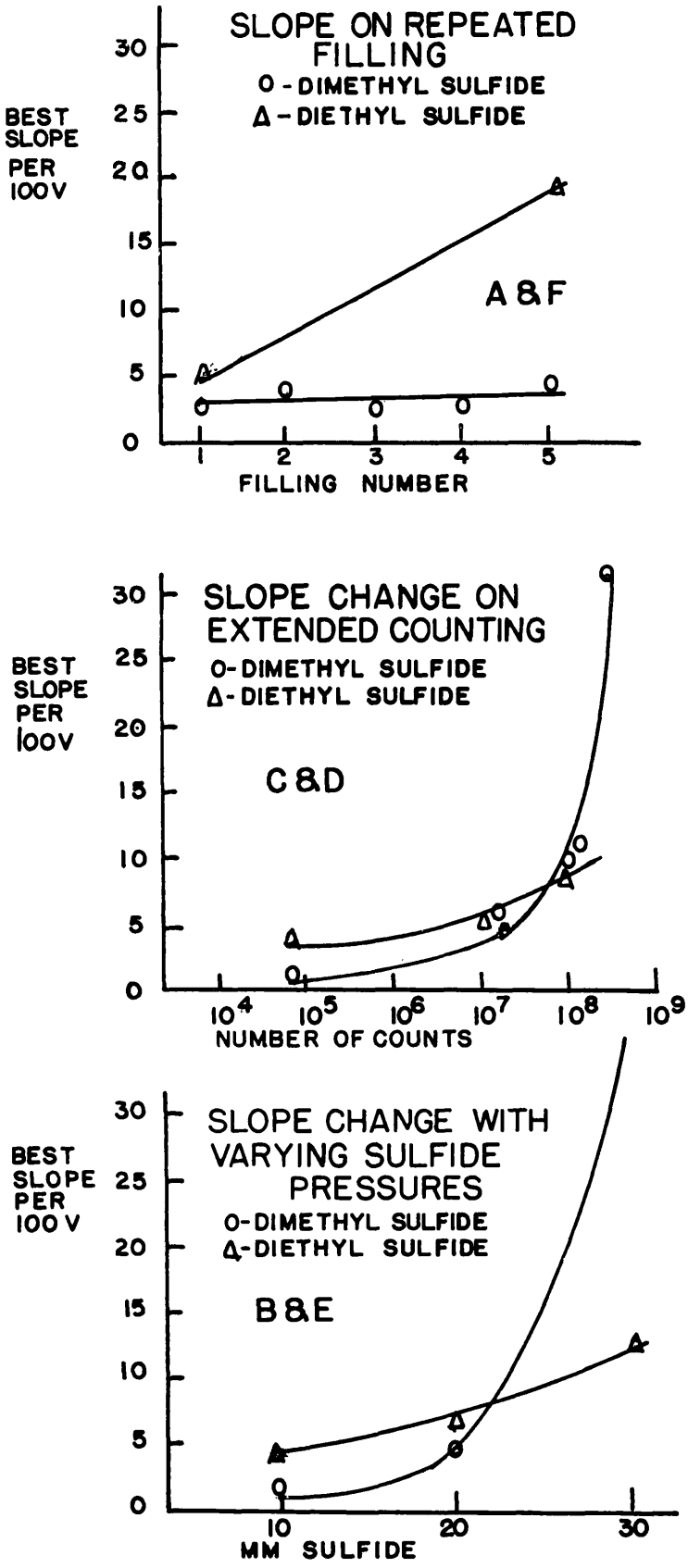
it. It was then placed in a bulb, and sealed onto the vacuum system while cold. Several samples of the gas were evaporated and discarded to remove the air in the system. As the material was evaporated to provide the vapor in the Geiger tube, some purification was obtained by the filling process.

The procedure was the same as that used on the inorganic sulfur gasses. A tube was first filled with argon and alcohol to check its performance. If this filling gave satisfactory results, it was used for further experiments with the untried mixtures after the tube had been pumped again to a high vacuum.

The first sample of dimethyl sulfide used gave excellent results when mixed with argon and irradiated with gamma rays from an outside radium source. Subsequent tubes proved to be equally satisfactory. Consequently, systematic evaluation of this compound was made.

One of the most important factors that had to be considered was the effect of the disulfide on continued use in the same tube. The results that were obtained in five runs are graphed in Section A of Figure 12. The slope did not change at all and the tube was as good at the end of the several runs as it was at the beginning.

The maximum tolerance of the tubes to methyl sulfide vapors was determined, and the results are shown in Section B of Figure 12. The slope of the tube did not exceed 5% until over 20 mm of dimethyl sulfide had been placed in the tube.



ORGANIC SULFIDE TUBE CHARACTERISTICS

FIGURE 12

The use of larger pressures is not satisfactory as they increase the potential required. An increase in terminal potential of about 600 volts higher is required to cause a tube to count at 30 mm pressure than that which is required at 10 mm.

A tube was filled with 10 mm of dimethyl sulfide, and 90 mm of argon and then irradiated with gamma radiation at 100 volts above its starting potential for a long period of time. The slope of the counting rate terminal potential curve was determined from time to time, and is graphed in Section C of Figure 12. The slope was less than 5% for about 10^8 counts. The dead time and recovery time of the dimethyl sulfide filled counters was determined by observing synchroscope displays such as are shown in Figure 1. The dead time was the same as that shown by ethyl alcohol and diethyl ether, 95 microseconds. The average recovery time was equal to that of the ethyl alcohol, 190 microseconds, but larger than that shown by the diethyl ether, 175 microseconds.

The starting potential averaged about 1,000 volts when disulfide was used at 10 mm pressure. This is about the same as that shown by the same tube filled with ethyl alcohol at the same pressure, and under the same conditions.

Methyl sulfide tubes seem to be equal to or better than ethyl alcohol filled tubes, when operated as self quenching Geiger tubes. Thus, methyl sulfide seems to be an excellent sulfur carrier. The plateau is generally longer

than that found with ethyl alcohol. Enough vapor can be placed into the tube to get large counting rates if dilution of the radioactive sample by inactive sulfur is not too large. A dilution factor of 10^6 gives very acceptable counting rates. The tubes do not suffer from change in characteristics on repeated filling; therefore, it would be possible to use the tube again for a different sample. The sulfide vapor is not corrosive to any extent to the brass, copper, Kovar, or mercury used in the metallic parts of the vacuum system. The counts that a tube can record before appreciable change will occur is much in excess of those required to obtain very high accuracy. Although in such tubes, filled with S^{35}, continuous bombardment of the gasses is always going on inside the tube. The very limited number of ion pairs formed when no terminal potential is applied should reduce the decomposition to a negligible extent. Thus, the only decomposition that would change the tube's slope and other properties would be that occuring when the count is made.

Diethyl sulfide was given an evaluation similar to that given to dimethyl sulfide, to ascertain if the properties that were found for the lower molecular weight sulfide would also hold for the new compound. This material possesses a lower vapor pressure than the dimethyl sulfide; however, at room temperatures this is still high enough to give relatively high pressures, pressures much in excess of those normally used in Geiger tubes.

The diethyl sulfide used was also a relatively high purity, inactive sample purchased from the Eastman Kodak Co. It was dried over $CaSO_4$ before being sealed to the system in a glass container. The procedures followed those employed when dimethyl sulfide was evaluated.

The slope change with repeated radiation at 100 volts above starting potential is shown on Curve D of Figure 12. The slope change occuring with different pressures of diethyl sulfide is shown in curve D, and the slope change on repeated filling is shown in curve F.

When filled with 10 mm diethyl sulfide and 90 mm argon, the tubes shown in Figure 10 had a starting potential of about 1 kV, a dead time of about 100 microseconds, and a recovery time of about 175 microseconds. All of these results are very close to those shown by a mixture of dimethyl sulfide in the same proportion with argon.

The only property shown by the graphs for diethyl sulfide that does not equal or better the same properties for dimethyl sulfide is a tendency for the diethyl sulfide to contaminate the Geiger tube. The cause for this was not determined. It probably lies in an impurity in the sulfur compound. Experiments with this material were not made as thoroughly as those run for dimethyl sulfide since it was thought that the contaminants might cause the vacuum system to become contaminated as well. The purity of any sample will depend upon the method of preparation of the sample used for counting, and a method will probably have to be

developed that will yield a relatively pure sample. If this difficulty could be surmounted, diethyl sulfide would be as good or better a sulfur carring quench as dimethyl sulfide.

The work on the preparation and evaluation of the sulfur containing compounds was not carried on any farther due to lack of time. A satisfactory method of preparing the sulfide must be developed, and when this is accomplished, it must be evaluated with radioactive sulfur. Too much trouble with the latter is not anticipated; S^{35} yields chlorine on losing an electron, and chlorine will also quench the discharge of a Geiger tube (114). The quantity of chlorine formed would be very small under any circumstances, unless, sulfur vapor pressures greatly in excess of optimum are used.

The ease of preparation of the inorganic compounds would seem to make them preferable to the organic sulfur carriers, but their incompatibility with known quenching mixtures makes them almost impossible to use. Dimethyl sulfide, on the other hand, possesses all the necessary requirements for a good quench, if it is possible to develop a method of preparation it should be an ideal material.

Table VI. Results of contamination trials.

Contaminant	Tube No.	Time +	Best Slope /100V	Starting voltage	Tube no.	Time +	Best Slope /100V	Starting voltage
Cellogrease	391	5*	2.7	900	392	5*	3.1	870
		4*	2.1	900		4*	4.1	920
		0	8.5	925		0	4.6	960
		3	14.2	950		3	14.0	970
		28	o	950		28	14.2	980
		47	o	950		47	o	960
Used Octoil	390	5*	2.9	925	397	2*	4.3	850
		4*	2.2	925		0*	3.6	870
		0	3.5	950		0	6.0	820
		1	7.0	940		60	16.0	840
		3	7.0	950		78	o	850
		27	5.9	940				
		28	8.8	950				
		47	19.0	950				
Silicone grease	386	2*	--	880				
		1*	3.0	910				
		2	3.8	940				
		18	4.9	930				
		37	6.4	930				
		46	7.0	930				
New Octoil	389	2*	--	900				
		1*	1.5	900				
		2	2.0	900				
		18	2.2	880				
		37	4.2	920				
		48	7.9	900				
Blanks	388	2*	--	900	384	2*	--	880
		1*	2.5	930		1*	1.5	870
		2	2.8	890		2	1.0	930
		17	3.8	910		17	4.0	880
		29	3.2	920		29	2.3	900
		48	2.8	900		48	3.0	900

+ Elapsed time in days.
* Time before introduction of contaminant.
o Would not count over 100 volt range--slope infinite.

Table VII. Results with Dimethyl (M) and Diethyl (E) Sulfide tubes.

mm Sulfide	mm Argon	Filling no.	Total Counts (M)	Total Counts (E)	Starting# Potentials (M)	Starting# Potentials (E)	Best Slope/ 100 V % (M)	Best Slope/ 100 V % (E)
10	90	1*	--	--	--	--	3.0	4.9
10	90	2*	--	--	--	--	4.0	--
10	90	3*	--	--	--	--	1.9	--
10	90	4*	--	--	--	--	1.9	--
10	90	5*	--	--	--	--	2.0	23.0
10	90	--	--	--	1000	1200	2.0	4.2
20	80	--	--	--	1280	1250	3.3	4.5
30	70	--	--	--	1680	1450	68.0	11.0
10	90	1	10^5	10^5	--	--	0.5	3.2
		1	3×10^7	3×10^7	--	--	3.8	5.7
		1	8×10^7	8×10^7	--	--	14.4	5.9
		1	10^8				29.0	

* Same tube shells, refilled
\# In volts

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