DEPENDENCE OF DEAD TIMES OF GEIGER–MÜLLER TUBES UPON BROMINE CONCENTRATION

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1. Introduction

The aggressivity of the halogen ionized in the course of operation renders the production of halogen-quenched GM tubes more difficult than the production of tubes with organic quenching gases. In spite of this they deserve our attention as in many applications especially in two of them we could hardly do without halogen-filled counting tubes. With portable radiation meters it is an advantage by no means negligible that the supply voltage of such tubes is lower by about 1000 V, and instead of a few tenths of a volt the signal supplied may rise to twenty or thirty volts. In addition, in a properly manufactured tube the composition of the gas is not liable to change and thus the results will be reliable even after several years of service.

Halogen-quenched counters find another application in automatic devices working with radioactive radiation in which the tubes are subjected to permanent radiation for long intervals of time. If the service life of such tubes were restricted to the counting of 10^8 impulses for which organic-vapour quenched tubes are guaranteed (with the electric parameters varying also within this limit), the necessity of frequent maintenance and readjustment of the automatic devices would be most awkward. Another important advantage of halogen-filled counters is their much higher signal voltage as mentioned above. For automatic devices dead times are of great importance, too. When the dead time is too long, the counted number of impulses will be smaller than the effective number which distorts the linear characteristics at high activities. The above considerations have advocated the examination of the dead times of halogen-quenched GM tubes.

For the measurement of dead times GM tubes of typical size have been manufactured and soldered to an exhaustion fork by a special method.

2. Production of tubes

The glass used for the production of tubes containing halogen quenching gas must not enter into reaction either with the halogen or with its ions, not even after a longer service life. Only hard glass sorts containing hardly any

alkali are suitable for this purpose. In our experience bromine-filled tubes made of Rasotherm glass have shown no changes, not even after several years of use. For the work discussed here only Rasotherm glass was used. The radius of the typical tubes was 10 mm, their working length 100 mm; working length meaning the distance between the small screening tubes applied at the two ends of the anode.

GM tubes need inlet electrodes. Their perfect sealing was obtained by soldering them into the glass. The coefficient of thermal expansion of Rasotherm glass is fairly near to that of tungsten, and tungsten electrodes (even of diameters much larger than needed for a GM tube) may be soldered in Rasotherm glass almost without any risk. In order to obtain a proper sealing the production of a chemical bond, not only a mechanical contact between filament and glass is needed. To obtain a chemical bond the tungsten filaments to be used as electrodes were carefully cleaned: first washed with 20 per cent hot NaOH solution then with diluted hydrochloric acid and with plenty of distilled water of at least 10^5 ohm resistance. The cleaning process ended by rinsing with alcohol. The cleaned tungsten wire was oxidized in the oxidizing part of the flame of a jet burner and the glass was melted together with the tungsten oxide thus produced. In this operation first a minute quantity of glass, called bead, is melted and soldered to the filament, and electrodes thus beaded are soldered to the vacuum body after close scrutiny. A properly soldered electrode displays a yellowish red colour characteristic of a glass and tungsten combination without the presence of bubbles.

The electrodes of the tubes should preferably not be of the same size. So as to obtain a high field strength about the anode the radius of the latter was selected to be smaller by about two orders of magnitude than that of the cathode. As tungsten has stood the test even with Br ions, tungsten filament of 0.1 mm diameter was used as an anode. To produce a cathode of large surface is by no means simple. Attempts to weld tungsten to tungsten i.e. a tungsten plate to a tungsten electrode filament had failed. The application of the usual Ni intermediation was out of question as Ni reacts with Br.

A coating of SnO_2 directly applied to the glass has proved suitable. Several methods of production SnO_2 coatings are known, not all of them are reproducible as regards electric conductivity (semiconductivity). Coatings for the cathodes of the GM tubes discussed here were made by the process forming the object of our Patent No. 145472, through the thermal decomposition of evaporated alcoholic $SnCl_2$ complexes by the surfaces to be coated. The resistance of the coatings was below 50 kohm. The two bottoms of the vacuum body containing the electrode inlets and the vacuum stub had to be made of non-coated glass, as an SnO_2 layer melted into glass is liable to develop micropores. Glass tubes partially coated were produced in an oven of special design used for the decomposition of the tin complex.

3. Vacuum-technical treatment and filling

Preliminary vacuum to the filling pump represented diagrammatically in Fig. 1 is supplied by a two-stage rotary vacuum pump type BP—500. The equipment made entirely of Rasotherm glass includes a mercury diffusion pump, a Torricelli vacuum gauge (5), a Pirani vacuum gauge (6) and a chiller (7). It has a test stub closed by tap (1) and a filling part closed by tap (2), connected through the bottom orifice of the vacuum tap (3) to a baking fork and through the two side orifices of vacuum tap (4) to two neon gas ampoules of about 1 litre capacity each. The baking section of the fork is marked by a dotted line in the Figure.

The tubes were evacuated through the test stub at tap (1) by means of a rubber vacuum tube sealed with silicone grease. The evacuated tube was sealed by soldering and "parted off" by a hand blowtorch. The vacuum in the tubes parted off was tested daily with a Tesla inductor.

After a 28 day vacuum test five tubes found faultless were soldered to the stub near the tap (3) of the pump in a horizontal plane through junctions of 8 mm inside diameter without neck contractions. Unusually wide junction and the absence of any parting-off neck result in rapid setting of equilibrium at the chilling of bromine. This consideration gave us to renounce to the advantage of a later easier parting off. The fork clamped to a Bunsen stand was soldered to the stub of the vacuum stand in such a way as to permit the stove of the vacuum stand moving on rails to be pulled over the fork. Outside the section of the fork entering the stove, a vertical descent pipe contained an ampoule of bromine of special purity.

The fork soldered to the vacuum stand and evacuated was tested at each soldering point with a Tesla apparatus, then allowed to stand under vacuum for at least 24 hours. The arrangement was re-tested with a Tesla unit until all leakages, to the very smallest ones, were traced and removed. Then the "stove" was pulled on and the section of the fork beyond the dotted line heated under permanent evacuation to 450 °C in 2 hours and kept at this temperature for another three hours. Then it was slowly cooled and checked for traces of gas. As the setting of equilibrium takes some time it is advisable to repeat this test the following day. Should there be any air left, the search for leakage is repeated. If we are lucky the set may be stoved again. This time it is enough to reach the temperature point specified after which cooling may start immediately. The next step after cooling is filling with a noble gas. This is checked with the pressure gauge (9) of the filling pump.

The Geiger—Müller tubes soldered to the fork were filled uniformly with spectrally pure neon gas of 200 mm Hg pressure. The gas ampoule, a product of the Hoechst factory, had a breaking seal. The tube equipped with a solenoid, and the breaking seal protected by a steel sleeve, the ampoule was fixed to

the filling pump at the point shown in Fig. 1. The seal must not be broken before complete evacuation. All the five tubes developed glow discharges starting at 200 V. Having checked this the fork was parted off from the vacuum stand and the seal of the bromine ampoule broken by its own weight.

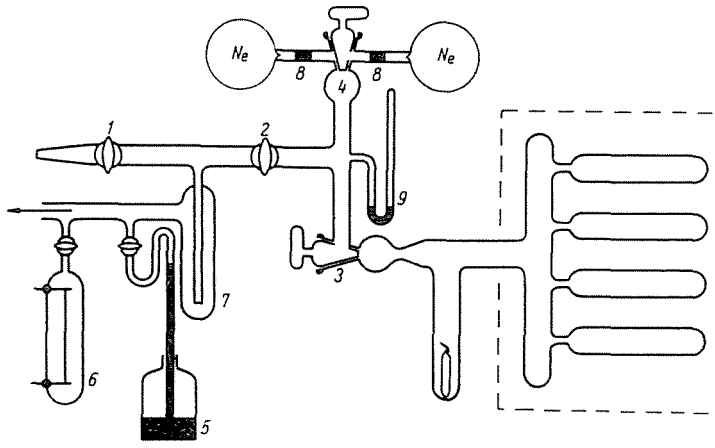

Fig. 1

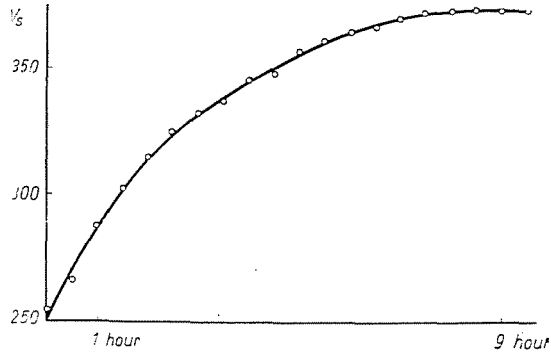

Fig. 2

Bromide vapour was kept in the tubes for several days and meanwhile the tubes were heated to 100 °C for a few hours.

To adjust the bromine concentration in the tubes the extension of the fork containing the bromine ampoule is usually put into solid CO_2 wetted with alcohol, and in the scheme represented in Figure 5 the starting voltage and the signal shape are examined. The starting voltage of a few hundred volts corresponding to fully reproducible signals of 0.3 V amplitude, differs but by a few tenths of a volt from the voltage at which the lowest signals appear in the oscilloscope.

In our sets of measurement this process was modified inasmuch as the alcohol was cooled to the required temperature by adding solid CO_2 and the temperature of the coolant was kept constant to the nearest 1/4 °C. To reach such a temperature (—50 °C) from —78 °C is represented in Fig. 2, and from room temperature in Fig. 3, Fig. 4 shows the starting voltages (V_s) of the tubes as a function of the temperature (bromine vapour tension).

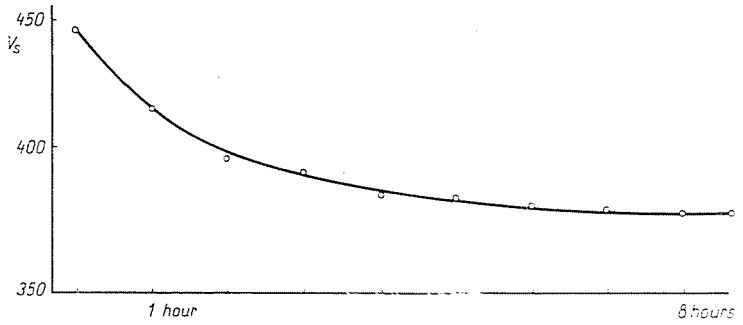

Fig. 3

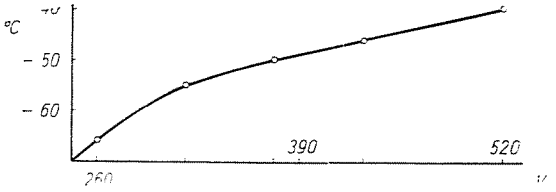

Fig. 4

4. Measurement of dead time

Dead time may be measured under any condition by increasing the irradiation to a known extent through an additional radiation source and calculating the change in the counting speed from $n_0 = \frac{n}{1 - n\tau}$. Another technique, a triggered oscilloscope, gives a more direct image. By a third method, published and used by CURRAN and RAE [4], the output of the counter is fed into two channels, one of which is delayed. The impulses are mixed at the output of the channels and coincidences are counted. Coincidence is observable only if the delay is greater than the dead time. This method is affected by quite a number of error sources and is not nearly as precise as the first two. Particularly the oscilloscope method is most illustrative and by photos taken permit measurements of high accuracy. In our work we have adopted this method.

The schematic diagram of the equipment used for the measurement of dead time is shown in Fig. 5. The Geiger—Müller tube is supplied from an adjustable high-voltage source (1). The working resistance of the Geiger—Müller tube is 1 Mohm. The impulse taken from the filament passes through a capacitor either to scaler (2) or to oscilloscope (3). The impulse is coupled to the oscilloscope through a 1000 pF capacitor. The EMG oscilloscope of 10 Mc/s band width is a synchronoscope designed for using in TV techniques permitting the examination of triggered impulses, too. Along the schematic diagram represented in the Figure either the number of impulses is determined with the scaler or the shape of the impulses is examined with triggered saw-tooth generator start. Our oscilloscope has also a marker signal on the impulse which

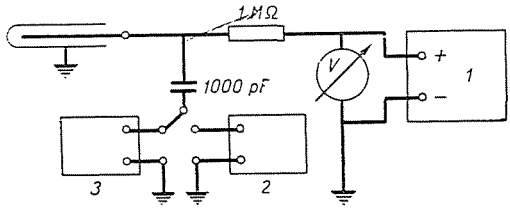

Fig. 5

permits the direct reading of the duration of the impulse. The marker signal is of 1, 10 or 100 microseconds. In our work the 10 μs marker was made use of (as the most suitable for the examination of the duration of the impulses). In measurements of this type the interval between the beginning of the impulse and the start of the saw-tooth generator is of the greatest importance, because if this interval is comparable with the duration of the impulse it may be the origin of a significant error. The shorter the time lag to the starting of the image as compared to the starting of the impulse the better the accuracy of the time measurement. In this respect our oscilloscope has stood the test as the interval between the start of the impulse and the start of the triggered saw-tooth generator is less than 0.1 μs, whereas the length of the signals to be examined is 100 to 200 μs. Accordingly, the error in our counting is less than 0.1 per cent.

5. Measurements

In Chapter 3 the method of adjustment of the bromine vapour pressure was discussed in detail. To test this it seemed necessary to adjust vapour pressure in overcooled and overheated conditions to find out the variation of the starting voltage of a Geiger—Müller tube under such an influence. The measurement results are shown in Fig. 4. The starting voltage of an overcooled Geiger—Müller tube is lower than that specified for standard conditions and

as a previously overcooled tube is warmed up to the temperature which gives the required pressure the starting voltage increases (Fig. 2) until the bromine pressure corresponding to equilibrium temperature is reached. When an overheated tube is cooled first the starting voltage is much higher than specified and from this value it continuously decreases to the specified value (Fig. 3).

The Figures show that the curves of cooling from overheating and warming from overcooling are in fair coincidence, provided sufficient time is left after overheating and overcooling. This permits us to coordinate the bromine tensions to the temperatures measured for the given apparatus.

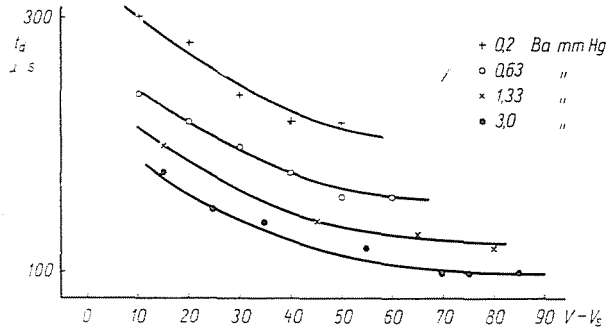

Fig. 6

Naturally the curves were taken with the coolant at the specified temperature. The first aim of these tests was to find out the extent to which dead time depends on bromine vapour tension. For this reason the dead times were examined at various bromine vapours, pressures in the plateau range. Four measurement results have been represented for bromine vapour tensions of 0.2, 0.63, 1.33 and 3 mm Hg in Fig. 6.

A comparison of the dead times belonging to the same pressure shows that with the starting voltage increasing dead time decreases first rapidly then at a pace gradually slowing down and finally, the dead time does not vary any longer with the starting voltage and becomes constant. This result is in a qualitative agreement with the calculations of WILKINSON [3] for tubes when working with organic quenching vapours. Though Wilkinson's calculations give qualitative results only the dependence on the operating voltage of our curves obtained by experimental work and that of the dead times in Wilkinson's calculations are of the same character. In accordance with this, at the starting section of the plateau, i.e., in the close vicinity of the starting voltage, dead times are relatively long, then they rapidly decrease and finally, they become constant as was pointed out above. Considering that the plateau of halogen-quenched tubes is shorter the change of the dead time with the voltage compared with the starting voltage ($V - V_s$) can be followed through

to a shorter section only. Here the decrease of the dead time is more than 30 per cent which, as regards the accuracy of measurements, is by no means negligible, not even in level meters. The Figure clearly shows how the plateau is lengthened with increasing bromine vapour content, since the maximum of $(V - V_s)$ represented indicates the length of the plateau.

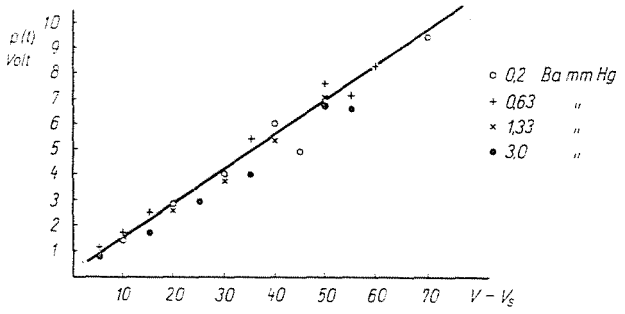

Fig. 7

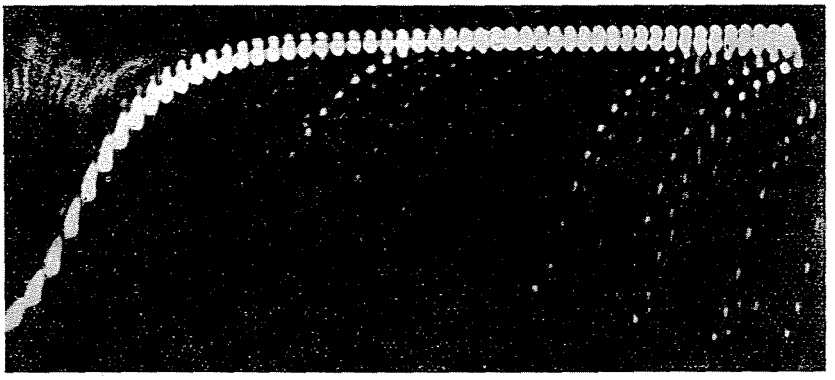

Fig. 8

For the interpretation of the results further experiments were needed. The results may be explained by the phenomenon of space charge. To take space charge into consideration the overall charge produced on the filament at various bromine vapour tensions had to be determined. Space charge for each bromine concentration and overall charge corresponding to this were examined by the inspection of the overall charge produced on the filament at 50 V above the starting voltage in a single discharge. Namely, Fig. 6 proves that at 50 V above starting voltage the dependence of dead time on voltage ceases to exist at all bromine vapour tensions. The oscilloscope used for dead time measurement was also suitable for the measurement of the impulse amplitudes as it had a voltage impulse calibrated for amplitude.

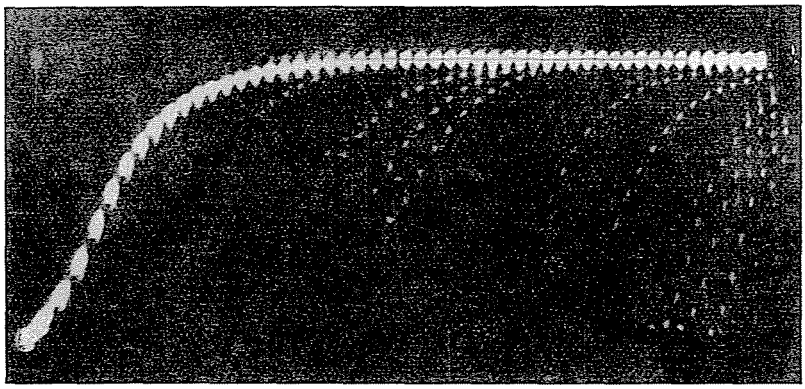

Fig. 9

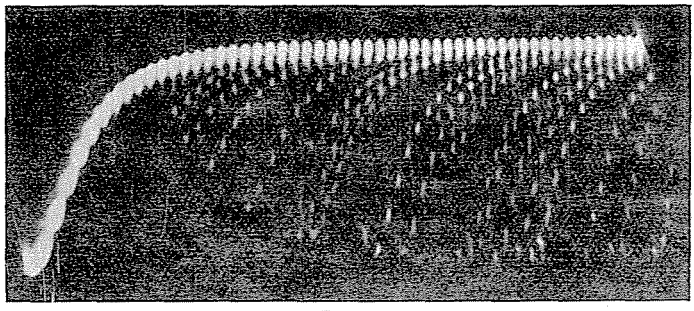

Fig. 10

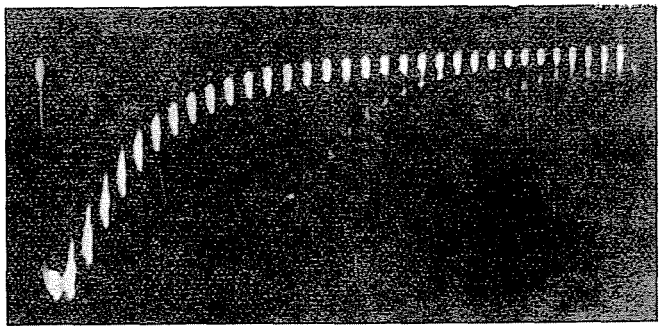

Fig. 11

For an additional checkup the calibrating impulse of the oscilloscope was again calibrated. The amplitude of the impulse $p(t)$ was measured in the plateau at the end of the voltage, separately for each of the four bromine concentrations. The results are shown in Fig. 7. As a function of overvoltage: amplitude shows a nearly linear relationship with overvoltage. It is remarkable that the amplitude of the impulse does not vary with the bromine concen-

tration, as the relationships of the amplitudes found for the four concentrations coincide within an error margin. The amplitude corresponding to 50 V overvoltage may be read off from this Figure. To determine the absolute value of the overall charge produced on the filament the area under the impulse had to be summed up. Four photos, Figs 8, 9, 10, 11 illustrate the shape of the impulse amplitude and the dead time measurement method for four bromine vapour pressures.

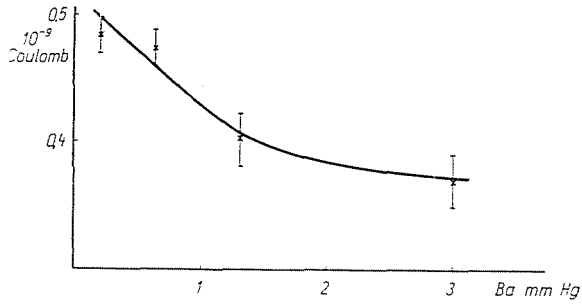

Fig. 12

In addition to the impulse amplitude also its characteristic being known the amplitude of the impulse at any section of the impulse is known, too, and so may be integrated. Owing to the marker signals of 10 μs also the times of the impulses are known and the overall charge produced may be calculated from these data. Figure 12 represents the overall charge found at 50 V above starting voltage as a function of bromine vapour tension. The curve permits to draw the following conclusions: as bromine concentration increases the absolute value of the charge represented by the ion cloud around the filament decreases. This means that at a low bromine vapour tension the overall charge produced in a single impulse is considerably higher, by about 30 per cent, than at higher bromine vapour concentrations. During the operation of a Geiger—Müller tube the large quantity of ions produced in the vicinity of the filament anyhow gives rise to a field charge.

The relationship between charge and bromine concentration is further proved by the fact that as bromine concentration decreases the overall charge increases at a quickening pace and, in accordance with this, the field charge also increases.

6. Conclusions

Our measurements have shown that the increase of dead time with the decrease of bromine vapour tension comes from field charge increasing at a rapid pace. Though field charge greatly influences the motion of ions from the

anode filament towards the cathode, on the other hand, with higher field charges also the absolute ion concentration is higher. Accordingly, as positive charges are present in the field, the screening effect increases. The increased screening effect lowers the field produced by the voltage fed to the filament to such an extent which corresponds to the magnitude of the charge. The experiments have shown that this phenomenon *may be* and *is* the origin of the increase of dead time with decreasing bromine concentration. The increase of the field charge has another consequence, too. With a higher field charge the critical radius at which the counter can count new signals also increases, though the mobility of the ions decreases also in the space extending to the critical radius, as it is the effective field that decreases. This is another reason for which dead times increase.

From the above results it is proposed that halogen-quenched tubes should be used at voltages of 40—50 V above starting voltage and the bromine tension in the tubes should be selected from 1 to 3 mm Hg whenever to keep the dead time at a low level is important.

Summary

Examinations of bromine-quenched Geiger—Müller tubes with an oscilloscope have revealed a relationship between the concentration of bromine and the dead times of the tubes. Measurements have shown that with increasing bromine concentration the dead time of the tube decreases. A theoretical interpretation of this phenomenon is given in the paper.

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