The Factors Influencing the Plateau Characteristics of Self-Quenching Geiger-Mueller Counters

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Investigations of factors influencing the starting potential and plateau slope of self-quenching G-M counters show: I. Impurities such as air or oxygen increase the plateau slope and starting potential of argon-alcohol counters. II. Operation of the counter causes changes in plateau characteristics as follows: (a) Immediately after use, the plateau slope increases slightly. (b) With continued use the plateau slope becomes steeper. (c) For moderate use the counter recovers when inactive, but never to the original characteristics. (d) With further use, the counter loses its plateau and does not recover. (e) The pressure in an argon-alcohol counter was observed to increase as a function of the total number of counts recorded. III. For argon-alcohol counters the useful life in these experiments was found to be about 10^9–10^{10} counts; for argon-methane, 10^7–10^8 counts. The observed changes of characteristics are presumably due to the decomposition of the organic vapor by the discharge.

INTRODUCTION

WHEN a G-M counter of the self-quenching type is connected to the usual external recording circuit, the relationship between the counting rate for a given source as a function of the counter voltage is given by the familiar "plateau" curve. The plateau characteristics are the starting potential and the slope of the plateau. The starting potential is the minimum voltage at which the pulse is of such size as to be able to actuate the external apparatus, and its value will depend on the nature and pressure of the gases used and upon the sensitivity of the amplifier. The slope of the plateau is defined as the percentage increase of counts per volt of counter potential. A perfect counter would be one for which the plateau slope is zero, i.e., the counting rate is independent of the voltage. A flat plateau is not possible because of the occurrence of spurious counts which originate inside the counter, often as a result of the discharge itself.

The problem here discussed is to determine what factors influence the plateau characteristics. Two conditions are desirable: (a) the starting potential should be as low as possible without affecting the efficiency of the counter, and (b) the plateau should be as nearly flat as possible over a considerable voltage range, so that the counting rate be independent of voltage fluctuations.

Many articles have appeared on the problem of the preparation of alcohol-argon counters and on the effect of the preparation on the plateau characteristics.[1–6] Care in preparation reduces the spurious counts and, therefore, the plateau slope. Experiments on the temperature coeffi-

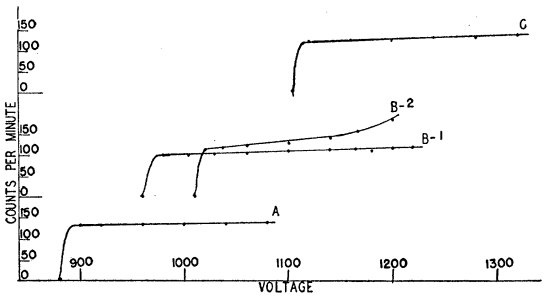

FIG. 1. Comparison of plateau characteristics of alcohol-argon counter having a total pressure of 10 cm of Hg with different mixtures. (*A*) 5 percent alcohol vapor: starting potential 840 volts, slope less than 0.01 percent per volt. (*B*-1) 10 percent alcohol vapor: starting potential 940 volts, slope 0.025 percent per volt. (*B*-2) 10 percent alcohol vapor and 2 percent air; starting potential 1000 volts, slope 0.15 percent per volt. (*C*) 20 percent alcohol vapor: starting potential 1080 volts, slope 0.05 percent per volt.

[1] S. C. Curren and V. Petrzilka, Proc. Camb. Phil. Soc. **35**, 309 (1939).
[2] H. V. Neher, J. Strong's *Modern Physical Laboratory Practice* (Prentice-Hall, Inc., New York, 1941), p. 268.
[3] F. R. Shonka, Phys. Rev. **55**, 24 (1939).
[4] G. L. Locher, Phys. Rev. **55**, 675 (1941).
[5] C. H. Collie and D. Roaf, Proc. Phys. Soc. **52**, 186 (1941).
[6] F. J. Davis and L. F. Curtiss, J. Research Nat. Bur. Stand. **29**, 405 (1942).

cient[7] show that in the case where the quenching constituent is a vapor, temperature has a decided effect on the plateau characteristics because of the temperature dependence of the vapor pressure.

APPARATUS

The conventional multivibrator, scale-of-8, and mechanical recorder circuit were used for recording the counting rate during plateau runs. A two-megohm leakage resistance was used in all tests described below. Counters were of the thin-walled glass type[8] of 15-cm length, 2-cm diameter with 4-mil tungsten center wire. The silvered portion extended for a length of 8 cm with the thin-walled section 6 cm long. The glass thickness varied considerably but was of the order of 10^{-2} cm. A circular manifold held 6 counters parallel and equi-spaced at a distance of 7 cm from the center, where a radium source was placed during the continuous operations. The source was of such strength as to give about 100 counts per second. All tests were made on 6 counters, and in most cases the counters were evacuated and refilled a second time and the tests repeated.

OBSERVATIONS

Test of Dependence of Flatness of Plateau on Argon Purity

A manifold of 6 counters was filled with a mixture of 5 percent alcohol vapor and 95 percent argon, which latter gas was 99.8 percent pure, to a total pressure of 10 cm of Hg. The counters were found to require 24 hours for complete mixing and diffusion of gas and vapor. The state of complete equilibrium was verified by checking the starting potential at intervals of several hours; the starting potential being a function of the percentage of vapor present. A beta-source weaker than the one mentioned above was placed 20 cm from the counters in all plateau runs; this yielded about 150 to 200 counts per minute depending on the individual wall thickness. The counting rate was observed for an over-voltage of 200 volts, in steps of 20 volts. At each voltage step a sufficient number of counts was recorded

TABLE I. Dependence of plateau slope and starting potential on argon purity and alcohol content.

Percent alcohol vapor	Argon purity	Starting pot. volts	Plat. slope percent/v
5	99.8	840	0.01
10	99.8	960	.02
10	98.0	1000	.15
10	90.0	1125	.25
20	99.8	1080	.05
20	90.0	1220	.35

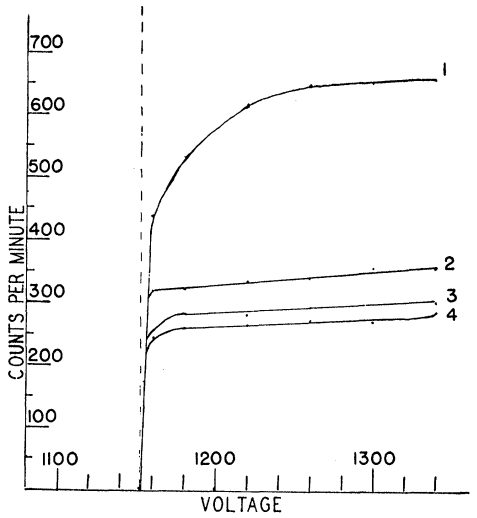

FIG. 2. Recovery of the plateau of an alcohol-argon counter after an exposure of 125 hours (5×10^7 counts). Immediately after exposure and one day later the counting rate was too high for the mechanical recorder and is represented by the broken vertical line. Curves 1–4 represent the plateau after a rest period of 2, 3, 4, and 5 days, respectively.

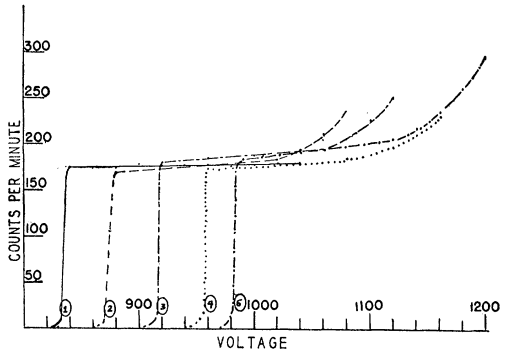

FIG. 3. Plateau characteristics of alcohol-argon counter after exposure at a rate of the order of 4×10^5 counts per hour (1) before exposure, (2) one day after 34-hr. exposure, (3) one day after 63-hr. exposure, (4) one day after 97-hr. exposure, and (5) four days after 169-hr. exposure.

[7] S. A. Korff, W. Spatz, and N. Hilberry, Rev. Sci. Inst. **13**, 127 (1942).

[8] Silvered thin-walled type made by Eck and Krebs of New York.

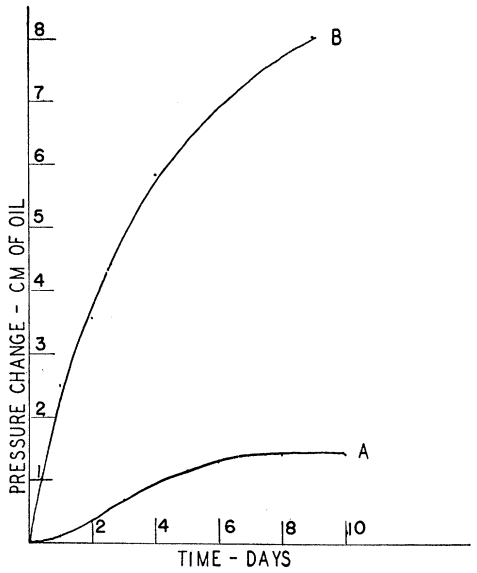

FIG. 4. Pressure change for alcohol-argon (*B*) and methane-argon (*A*) with exposure time. Differential manometer attached to the manifold contained Apiezon *B* oil. Counting rate was of the order of 10^7 per day.

so as to have the statistical fluctuations (based on the square root of the total count) below 3 percent. The plateau slope in all cases was less than 0.01 percent per volt.

The process was repeated with mixtures of 10 percent and 20 percent alcohol vapor at the same total pressure. The effect of contamination, produced by adding 2 percent and 10 percent of air, was examined. Typical curves in Fig. 1 show the effect of increased alcohol vapor and of air contamination. Table I gives a comparison for these mixtures. In all cases 6 counters were used so as not to be influenced by the possible peculiarities of any one counter.

Aging Tests

The manifold was filled with a mixture of 5 percent alcohol and 95 percent argon to a total pressure of 10 cm of Hg, and the characteristic curves taken. A radium source was placed at the center of the manifold and the counters connected to the high potential with an over-voltage of 60 volts. The pulse size was noted for this over-voltage, which was maintained by checking the pulse size every 6 hours. The counters were operated for a period of 24 hours, and then the plateau was measured by using the same beta-source and geometry. Immediately upon removal, the counters exhibited counting rates much higher than those previously recorded for the given source and geometry, but, after 24 hours during which the counters were inactive, they again showed plateau characteristics. With each successive period of use the slope of the plateau changed very little, but the starting potential increased steadily. When the counters had been subjected to prolonged use (over 100 hours), it was noted, as before, that immediately after removal the counters had no plateau, and it took several days for them to recover. This recovery effect in the case of 125 hours use is shown in Fig. 2. The change in starting potential is shown in Fig. 3, in which plateaus were determined after progressively greater use. In each case the counter was allowed to recover fully before the plateau was measured.

Pressure Variation

It was observed in a preliminary test that the pressure in the counter increased with use. The above aging process was repeated for a mixture of 20 percent alcohol and the same total pressure. In this case a differential oil manometer was attached to the manifold to measure the pressure variation as a function of the counter's use. The results are shown in Fig. 4.

The question was raised as to whether the change in the starting potential could be ascribed to a change of both composition and total pressure of the quenching vapor in the counter. This was experimentally determined by observing the starting potential-pressure curves under the following conditions. The manifold was initially filled with a 20 percent alcohol mixture at a total pressure of 10 cm of Hg and the starting potential change was noted for pressure change when: (1) the total pressure of the mixture increased, (2) the manifold with the original mixture was subjected to use at the high counting rate, and (3) the total pressure was changed by the addition of hydrogen to the original mixture. Figure 5 shows the results of these tests.

There was no measurable pressure change during the recovery period when the counter was not in use.

Increasing Photosensitivity with Use

It was noticed that alcohol-argon counters became photosensitive to visible light after long use.

Tests on Methane Counters

Experiments on methane-argon counters have not been carried out as extensively as those for the alcohol-argon type. Previous experiments[7] on temperature coefficients show that over the range of temperature studied (−22°C to 85°C), the methane-argon counters showed no appreciable change in plateau characteristics. In general, methane-argon counters have a higher starting potential and greater plateau slope, and preliminary tests did not show the recovery effects observed in the case of the alcohol-argon counters. Figure 4 shows the comparison of pressure increase between the two types of counters under identical operating conditions. It is estimated that an alcohol-argon counter undergoes about 10^9 to 10^{10} discharges before losing all plateau characteristics. Methane-argon counters, on the other hand, have a useful life of approximately 10^7 to 10^8 discharges, but this estimate is based on incomplete data at present.

DISCUSSION

When the alcohol-argon counter is operated continuously at the high rate described, the above-mentioned phenomena were observed as length of time of use progressed. We shall see how these effects can be explained on the basis of the breakdown of the alcohol molecule by the action of the avalanche discharge.

As indicated earlier, the avalanche discharge in the counter is believed to be initiated close to the center wire. The electrons in the avalanche have sufficient energy to ionize the argon and alcohol molecules in this region. Probably the discharge also causes the alcohol molecules to disintegrate into ions of lower atomic mass. Cummings and Bleakney[9] subjected ethyl alcohol to electron impacts and measured the mass and relative abundance of the ions formed. The essential part of their results is shown in Table II. However, their experimental procedure was to separate the various ions immediately after bombardment, and as many of the resultant ions are unstable, one cannot assume that all the types of ions are present in the case of the similar electron impacts occurring in the discharge process of the Geiger counter. A similar disintegration of methane has been observed by L. M. Yaddanapalli.[10] In this case hydrogen and heavy hydrocarbons are also the products of the disintegration process.

The relative abundance of alcohol in the disintegration products is only 4 percent, the rest being of lower atomic number. In this way the alcohol present in the counter is gradually decreasing with increased operation time of the counter, and it is to be noted that two of the final products are oxygen and hydrogen, both

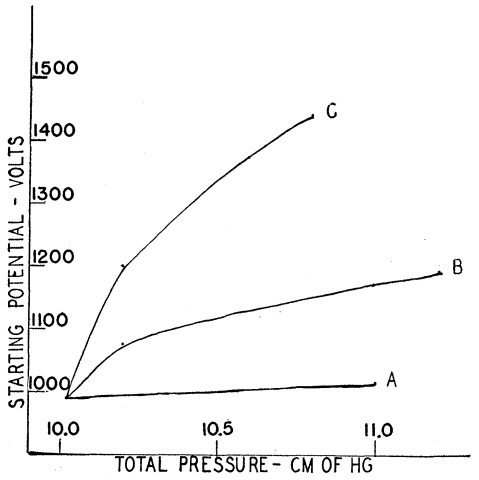

FIG. 5. Variation of starting potential of an alcohol-argon counter originally at a total pressure of 10 cm of Hg using a 20 percent alcohol mixture under the following conditions: (*A*) increase of total pressure of original mixture; (*B*) increase of total pressure resulting from exposure of original mixture; (*C*) addition of hydrogen to the original mixture.

TABLE II. Table of decomposition products of alcohol vapor in a discharge, from the data of Cummings and Bleakney.

Mass	Ion	Relative abundance	Mass	Ion	Relative abundance
46	C_2H_5OH	100	26	C_2H_2	110
45	C_2H_4OH	242	17	OH	3.9
31	CH_2OH	986	16	CH_4 & O	9.6
29	HCO	293	12	C	18.7
28	C_2H_4	390	2	H_2	30.4
27	C_2H_3	272	1	H	4.7

[9] C. S. Cummings and W. Bleakney, Phys. Rev. **58**, 787 (1940).

[10] L. M. Yaddanapalli, J. Phys. Chem. **10**, 249 (1942).

of which increase with operation time. This would seriously affect the slope of the plateau inasmuch as the percentage of organic vapor is less and the percentage of non-quenching gases is greater. The decomposition would be a gradual one.

Increase of Pressure and of Starting Potential

If the alcohol molecule breaks up to form other constituents, the total pressure will increase in accordance with the law of partial pressures. Also, for a given number of alcohol molecules originally available, one would expect that the pressure increase would not be constant with time of operation, but would tend to reach a steady value. This is observed in Fig. 4 for both alcohol and methane; as the length of time of operation increases, the total pressure tends to approach a constant value which would be that for complete disintegration. The fact that the change in pressure for methane is considerably less than for alcohol is explainable on the basis of the observations of Yaddanapalli[10] that heavy polymers are formed in the disintegration of methane thus reducing the number of atoms in the gaseous state.

The resultant increase in the pressure of the gases in the counter automatically brings about an increase of the starting potential. However, the starting potential is also a function of the type of quenching gas. If the increase of pressure is due to merely an increase of alcohol (which is unlikely on the disintegration hypothesis), the starting potential-pressure relationship is far below that actually observed in Fig. 5. On the other hand, if hydrogen is assumed to be the only disintegration product, the starting potential-pressure relationship is above that observed. It is well known that, in general, hydrogen is a "high starting potential" gas, and if 1 cm of hydrogen is added to a given mixture of alcohol-argon, the resultant increase in starting potential is greater than that for an addition of 1 cm of alcohol. The actual observed relationship between starting potential and pressure probably arises from a mixture of organic gases, as suggested in Table II. Just what the mixture composition is and how it changes with time is not known at present, but it evidently does change with time of operation.

Increase in Plateau Slope and Recovery

A change in the plateau slope takes place after the first aging run of 24 hours but does not change much with subsequent use, except for the fact that, as the counter is used more and more, the time required for the plateau to recover also increases. Abrupt change of the plateau does not seem to occur until the time of use is of such length as to destroy the self-quenching completely. This will presumably occur when the percentage of hydrogen present has increased, with the decrease of organic constituent, to the point where the amount of organic constituents is insufficient for complete quenching.

Effect of Percentage of Alcohol on the Plateau Slope

Tests on the correlation between plateau slope and percentage of alcohol indicate that, with increase of alcohol, an increase in the plateau slope results. If the alcohol is contaminated, as by the absorption of air, we would have a possible source of spurious counts due to negative ions whose quantity is dependent on alcohol content. Such a possibility is indeed present if the alcohol is not degassed after transfer to the mixing system. This had not been done in the present experiment. Calculations based on the known solubility of air in alcohol show that the contamination, for the percentage of alcohol used, would be of the order of several tenths (0.7 percent for the 20 percent alcohol mixture) of one percent. The large increase in plateau slope produced by admitting a 2 percent contamination of air has been found to be 0.15 percent per volt, and thus for the estimated 0.7 percent contamination in the case of the 20 percent alcohol mixture one would expect to find a slope of 0.05 percent per volt, as is observed. The exact agreement may, of course, be fortuitous.

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