THE
PHYSICAL REVIEW

A journal of experimental and theoretical physics established by E. L. Nichols in 1893

SECOND SERIES, VOL. 91, NO. 4 AUGUST 15, 1953

Formative Time Lags of Uniform Field Breakdown in Argon*

G. A. KACHICKAS† AND L. H. FISHER
Department of Physics, College of Engineering, New York University, University Heights, New York, New York
(Received March 10, 1953)

The nature of the uniform field breakdown in argon has been investigated by measuring the formative time lags of breakdown as a function of overvoltage (from about 5 to 100 percent), pressure (150- to 700-mm Hg) and electrode separation (0.3 to 3.0 cm). Formative time lags in argon at a given percent overvoltage are very long compared to the values previously obtained in air and nitrogen. For most values of pressure and electrode separation studied in argon, an overvoltage of 100 percent must be applied before the time lags decrease to the order of 1μsec. The results in argon (like those in air and nitrogen) indicate a Townsend buildup before breakdown. For the range of variables studied in argon, the data cannot be accounted for satisfactorily by buildup as a result of metastable action at the cathode, positive ion bombardment of the cathode, or photoelectric emission from the cathode by photons crossing the gap with a velocity close to c. There is a possibility that the results may be explained by a photoelectric effect at the cathode if delays are introduced due to the diffusion of resonance radiation; this conjecture remains to be verified. At this time the positive identification of the effective secondary mechanism in the breakdown of argon cannot be made. The present results indicate the universality of the Townsend buildup before breakdown, but the buildup may proceed by different mechanisms in different gases.

I. INTRODUCTION

THE present experiments are an extension of earlier work in air and nitrogen.[1] KF concluded that the secondary mechanism active in the spark breakdown of nitrogen in air is photoemission at the cathode, the electromagnetic radiation being transported from the anode to the cathode with essentially the velocity c.

The present work represents a further step in a program to ascertain the experimental facts on which a general theory of breakdown may be based. It seemed desirable to compare the breakdown mechanism of a noble gas with that of nitrogen and air, and argon was chosen for this purpose. These are the first formative time lag studies in argon for relatively high pressures.

II. APPARATUS AND EXPERIMENTAL PROCEDURE

Unless otherwise noted the apparatus and experimental procedure were exactly as described by FB and KF. Values of the first Townsend coefficient α as determined by Kruithof and Penning[2] were used to estimate the initial photoelectric current i_0. Although (for reasons to be discussed later) it is difficult to determine the actual value of i_0 accurately, the value of i_0 was always large enough to prevent statistical lags. About three observations of the time lags were made at each overvoltage studied. This small number of measurements were all that were necessary because of the remarkable reproducibility of the results. About ten different overvoltages were studied for a given pressure (p) and electrode separation (δ); such measurements were taken at various values of p and δ. The dependence of the time lags on i_0 was studied. An extensive statistical study of the time lags was carried out for one particular overvoltage (with $\delta=1$ cm and a pressure near atmospheric).

All pressures have been corrected to 22°C.

III. EXPERIMENTAL RESULTS

In order to calculate the percent overvoltage at which a time lag is being measured it is necessary to know the sparking potential V_s. Measurements of V_s for argon

* Supported by the U. S. Office of Naval Research and the Research Corporation. For a preliminary report of this work see G. A. Kachickas and L. H. Fisher, Phys. Rev. **82**, 569 (1951). Also see L. B. Loeb, Phys. Rev. **81**, 287 (1951).

† Now at North American Aviation, Downey, California.

[1] L. H. Fisher and B. Bederson, Phys. Rev. **81**, 109 (1951); G. A. Kachickas and L. H. Fisher, Phys. Rev. **88**, 878 (1952). These papers will be referred to as FB and KF, respectively.

[2] A. A. Kruithof and F. M. Penning, Physica **3**, 515 (1936).

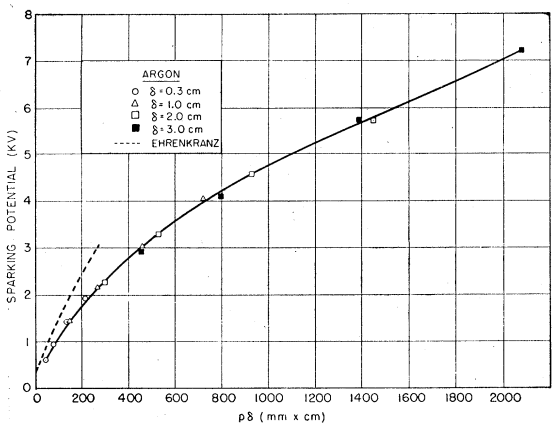

FIG. 1. Sparking potential *vs* $p\delta$ for argon.

were made and are given in Fig. 1. V_s could be measured at any given time to within a few volts. The values of V_s are not affected by the value of i_0 between 0 and approximately 100 electrons/μsec (primary currents greater than this were not used). This independence of V_s on i_0 is in strong contrast to the results in pure nitrogen (KF). Figure 1 also includes the sparking potentials obtained by Ehrenkranz[3] using platinum electrodes in argon. Ehrenkranz's values, which are in good agreement with those of Penning and Addink[4] using iron electrodes, are approximately 40 percent higher than the values obtained in the present work

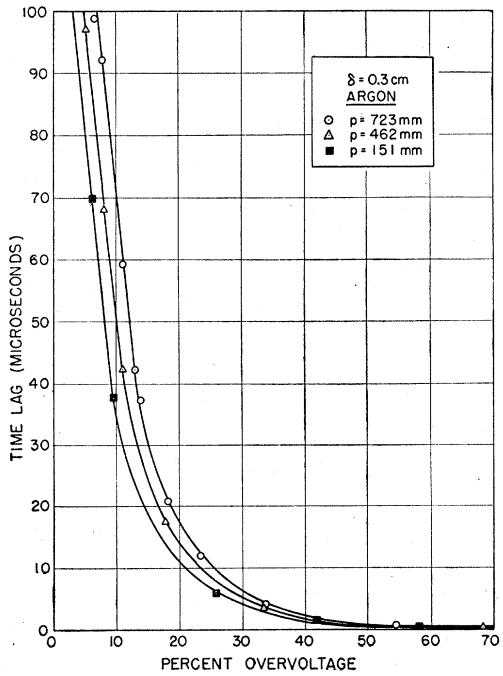

FIG. 2. Formative time lag *vs* percent overvoltage for at $\delta = 0.3$ cm at various pressures.

using brass electrodes. The measurements of Ehrenkranz and Penning and Addink go up only to values of $p\delta$ of about 300 mm×cm, while the present measurements go up to $p\delta = 2000$ mm×cm. Ehrenkranz and Penning and Addink used a calcium arc to purify their gas. Ehrenkranz observed the spectrum of the gas and reported sparking potential measurements for samples which showed no mercury and no bands. Penning and Addink state that the calcium removes all gases except the noble ones to less than 10^{-4} percent although they do not give any purity data on their gas. In the present experiment tank argon guaranteed by the Linde Company to be at least 99.8 percent pure was used; a mass-spectrographic analysis of a sample of the gas in contact with the chamber showed the purity of the

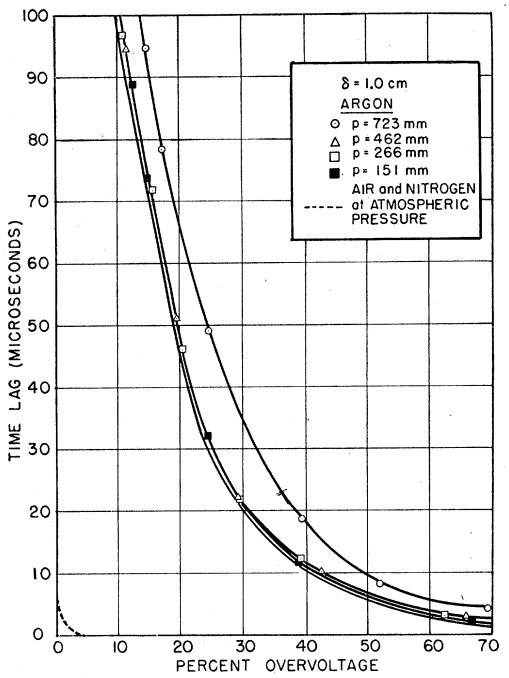

FIG. 3. Formative time lag *vs* percent overvoltage for argon at $\delta = 1.0$ cm at various pressures (also for air and nitrogen at atmospheric pressure).

gas to be 100 percent argon, this result being accurate to 1 part in 10^4.[5] When the argon was contaminated by impurities left by not outgassing the chamber as described by KF, the sparking potential and the time lags changed radically. For example, in one typical case without outgassing, V_s increased by about 50 percent and the formative time lags at 10 percent overvoltage decreased by about 90 percent. These changes are to be compared with the very consistent sparking potentials and formative time lags observed when the chamber was outgassed properly. It is not the primary purpose of this paper to evaluate the relative merits of these various measurements. However,

[3] F. Ehrenkranz, Phys. Rev. **55**, 219 (1939).
[4] F. M. Penning and C. C. J. Addink, Physica **1**, 1007 (1934).
[5] We are indebted to Dr. J. A. Hornbeck of the Bell Telephone Laboratories for this analysis.

in view of the established purity of the gas used in the present experiments, it seems reasonable to conclude that the new measurements of sparking potentials in argon are more accurate than the earlier ones of Ehrenkranz and Penning and Addink. Above $p\delta=300$ mm×cm, there are no sparking potential data with which to compare our measurements. The only difficulty to which the difference in sparking potentials gives rise is to introduce some question as to what values of α should be used in interpreting the time-lag data.

The time-lag measurements are shown in Figs. 2 through 5. These are the average values of the observed time lags with an illumination corresponding approximately to 50 electrons/μsec. For reasons to be given,

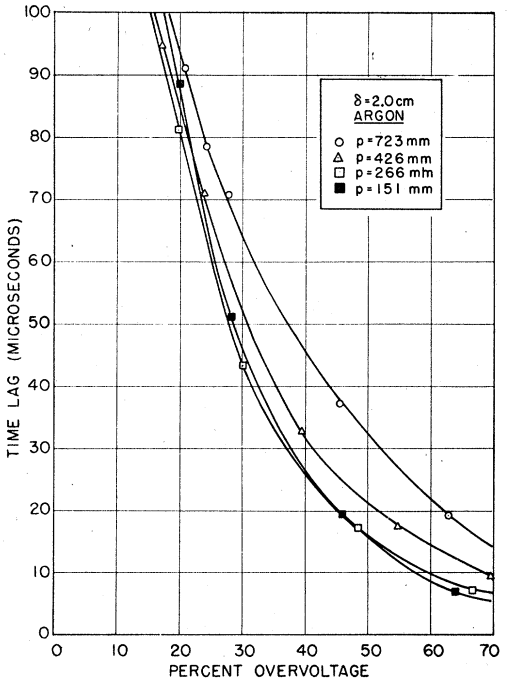

FIG. 4. Formative time lag *vs* percent overvoltage for argon at $\delta=2.0$ cm at various pressures.

these average values are interpreted as the formative time lags. Figure 3 contains for comparison the time-lag data for air and nitrogen at $\delta=1$ cm at atmospheric pressure. The formative time lags in argon are long (10^{-5} sec) even for overvoltages above 50 percent. Unlike the results for air and nitrogen there is a decided dependence on pressure, the time lags decreasing with decreasing pressure. The effect of electrode separation on the time-lag *vs* percent-overvoltage curves is shown in Fig. 6 for a pressure of 723 mm of Hg.

There is no detectable change produced in the time lags by changing i_0 from approximately 5 to 100 electrons/μsec or by using different approach voltages. These results are similar to those found in air and nitrogen. In fact, at $\delta=1$ cm and $p=723$ mm of Hg,

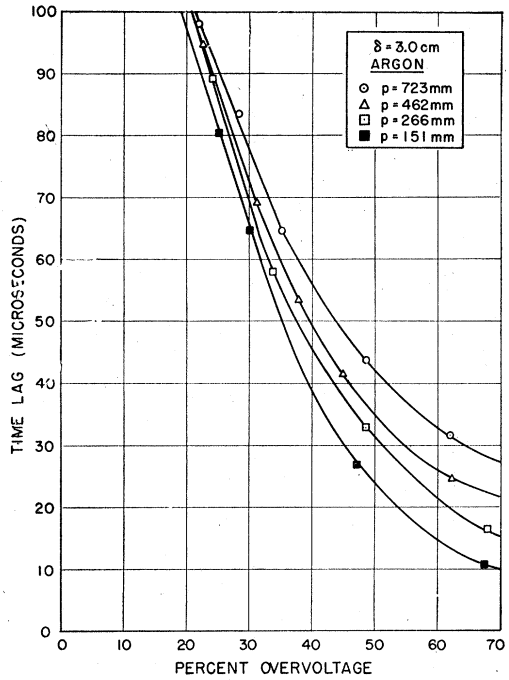

FIG. 5. Formative time lag *vs* percent overvoltage for argon at $\delta=3.0$ cm at various pressures.

no effect on the time lags was observed when the approach voltage was varied from 4000 to 10 volts below V_s.

The distribution of the time lags in argon obtained by taking 50 measurements at 50 percent overvoltage for

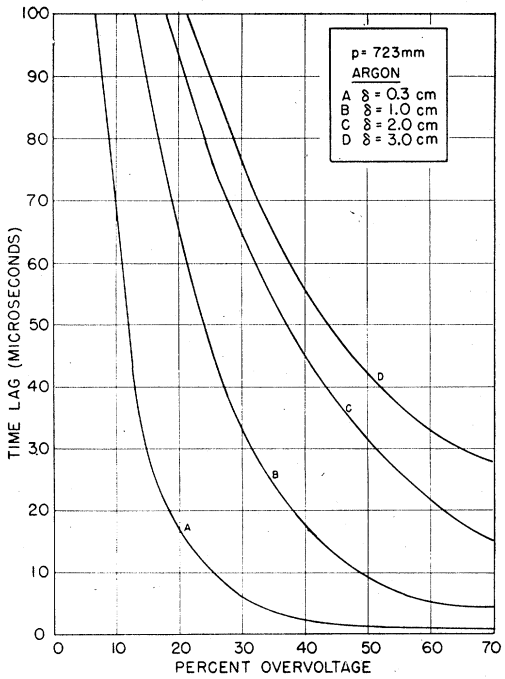

FIG. 6. Formative time lag *vs* percent overvoltage for argon at 723 mm of Hg at various electrode separations.

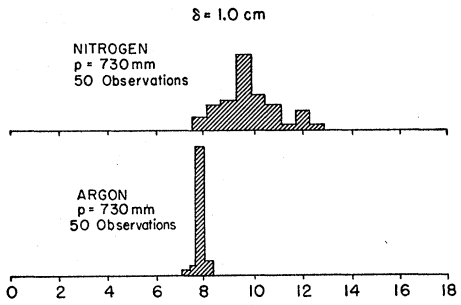

FIG. 7. Distribution of 50 time lags for argon and nitrogen. Abscissa in microseconds.

$p=735$ mm of Hg and $\delta=1$ cm is given in Fig. 7. Also plotted in Fig. 7 are the distribution of 50 time lags in nitrogen at a comparable formative time lag as given by KF. There is no point in displaying a Laue plot of the time-lag distribution in argon as it is evident that the sharp distribution of the observed time lags is not statistical. The very small spread which is observed in the time lags is attributed to instrumental fluctuations of the voltage supplies. Moreover, since the slope of the time-lag *vs* percent-overvoltage curves of argon change very slowly at all overvoltages studied, one would expect the spread of the time lags at different overvoltages if due to such instrumental fluctuations to be about constant at various overvoltages. This is exactly what is found experimentally.

IV. DISCUSSION OF EXPERIMENTAL RESULTS

The fundamental result displayed by the present work as well as by the earlier studies of FB and KF is that a Townsend buildup of some kind precedes the breakdown.[6,7] As already mentioned, KF gave arguments showing that the secondary process in air and nitrogen is a photoelectric action at or near the cathode, the photons traveling across the gap with essentially the velocity c. In the case of argon the question of what constitutes this buildup cannot be answered at present. Certainly, the action of metastable atoms at the cathode can be ruled out for the overvoltage region studied. Diffusion of metastables from the anode to the cathode requires much longer times than were measured in this experiment. The instruments used in the present study could only measure times between 0.1 and 100 μsec. Whether metastables are unimportant in the as yet experimentally uncharted region of overvoltage from zero to five percent cannot be determined without further experimental work.

The remaining sources of the buildup of the breakdowns observed in this study are (1) liberation of electrons by positive ion bombardment of the cathode and (2) photoelectric action at the cathode (a) by radiation traveling through the gas with essentially the velocity c or (b) by radiation traveling through the gas at a very much lower velocity c by the process of diffusion of resonance radiation. Assuming either mechanism (1) or (2a) operating, KF have shown that (under certain simplifying assumptions) the formative time lag τ may be written

$$\tau=\ln N/\{v(\alpha-\alpha_s)\}. \quad (1)$$

Here α and α_s are the values of the first Townsend coefficient at the voltage studied and at the threshold voltage, respectively, N is the number of electrons (crudely assumed to be a constant independent of p and δ) resulting from a single primary initiating electron which must be liberated in the last avalanche to form a spark, v is the velocity (v_+) of the positive ions for mechanism (1) and the velocity (v_-) of electrons for mechanism (2a). Equation (1) assumes that $e^{\alpha_s\delta}\gg 1$, that the second Towsend coefficient γ is independent of overvoltage, and that the formative time lag is much greater than (δ/v_+) for mechanism (1) or much greater than (δ/v_-) for mechanism (2a). Under these assumptions, according to Eq. (1), mechanisms (1) and (2a) give formative time-lag *vs* percent-overvoltage curves differing by a factor v_-/v_+ if one assumes the same value of N for the two processes.

Since at high enough overvoltages the observed formative time lag is always shorter than (δ/v_+) and in view of the conclusions drawn in air and nitrogen, the argon data were first analyzed in terms of mechanism (2a). Equation (1) was used in conjunction with values of α as given by Kruithof and Penning and values of v_- extrapolated with considerable uncertainty from the measurements of Nielsen.[8] The data at each overvoltage, pressure, and electrode separation were used to calculate a value of $\ln N$ and 80 values of $\ln N$ gave a mean value $(\ln N)_{Av}$ of 117 and a mean deviation of 84. For the case of nitrogen for which mechanism (2a) was used (and for which KF claim an excellent fit) $(\ln N)_{Av}=22.6$ with a mean deviation of 11.1. Although for argon the mean deviation of $\ln N$ is percentagewise larger than that for nitrogen, and this indicates that Eq. (1) does not represent the argon data as satisfactorily as the nitrogen data, the fit is not too bad. It seems reasonable to assume that these calculations verify the $1/(\alpha-\alpha_s)$ dependence of the time lags in argon. The serious objection to these calculations is the preposterously large value of N implied (10^{50}). It is not likely that the extrapolation of the values of v_- leads to errors of more than several tenths so that the high value of N obtained cannot be explained on this basis. If one derives the expression analogous to Eq. (1) where unity is not neglected compared to $e^{\alpha_s\delta}$, one

[6] Long formative time lags in air at low overvoltages have been confirmed by H. Bandel (private communication from L. B. Loeb).

[7] See also F. Llewellyn Jones and A. B. Parker, Proc. Roy. Soc. (London) **213**, 185 (1952); Dutton, Haydon, and Jones, Proc. Roy. Soc. (London) **213**, 203 (1952).

[8] R. A. Nielsen, Phys. Rev. **50**, 950 (1936).

obtains an even more unreasonable value of $(\ln N)_{Av}$.[9] It was then thought that perhaps the discrepancy in sparking potentials might account for the difficulty. Therefore, for purposes of calculation, the voltage measurements at the observed time lags were raised arbitrarily by 40 percent, and the data were analyzed on this basis. The results were even more unsatisfactory. We conclude therefore that, for the range of pressure, electrode separation, and overvoltage studied, mechanism (2a) does not satisfactorily represent the data in argon.

Entirely analogous calculations were made for the positive ion bombardment of the cathode mechanism.[10] These calculations yielded values of $(\ln N)_{Av}$ much too small to be satisfactory. We therefore also exclude mechanism (1) as an explanation for any of the observed data.

Lauer,[11] in studying transient pulses in the positive corona in argon with cylindrical geometry over a range of pressure from 25 to 400 mm, found a very small value of $\gamma(\sim 10^{-4})$ which he attributes to positive ion action at the cathode. Lauer found no $\gamma(\gamma_p)$ attributable to photoelectric action at his nickel cathode. On the other hand, Colli, Facchini, and Gatti,[12] in studying pulses in argon counters with pressures of 15 to 100 cm with wire and cylinder sizes much larger than those of Lauer, did find a γ_p for brass. Further work of this kind involving control and variation of electrode material and dimensions over a wide range of pressures in gas of known purity may bring these results into closer harmony with themselves and with our conclusion that mechanisms (1) and (2a) must be ruled out for our experiments in argon.

We now consider the possibility that the present results in argon can be explained by a buildup resulting from photoelectric action at the cathode by radiation produced in the gas reaching the cathode by a diffusion process (mechanism 2b). Such a mechanism introduces delays in addition to electron transit times and would be effective in giving lower values of $\ln N$ than were necessary in considering mechanism (2a). The mechanism of the diffusion of resonance radiation has recently been re-examined theoretically by Holstein[12] and experimentally in mercury by Alpert, McCoubrey, and Holstein.[13] Holstein discusses the emergence of resonance radiation from an infinite slab. These workers have shown that resonance radiation in a gas may be imprisoned for times of the order of tens of microseconds. Our problem is essentially much more complicated than the one treated by Holstein, and in view of the fact that there is no experimental information on the diffusion of resonance radiation in argon and that the role of the diffusion of resonance radiation in gas breakdown has never been established, it does not seem warranted to make elaborate calculations on this mechanism. One may notice, however, that Eq. (1) gives a reasonable fit and a sensible value of $(\ln N)_{Av}$ if one assumes that the average transit time of resonance radiation is about ten times that of the electron. A direct experimental test of the importance of resonance radiation on the breakdown would be most valuable.

We are indebted to Dr. M. Menes, now of the Westinghouse Research Laboratories, for a number of stimulating discussions.

[9] The above calculations do not include 19 points for which Kruithof and Penning do not give values of α_s. It is interesting to note that for those thresholds for which Kruithof and Penning give values of α, our V_s values give an average primary multiplication at threshold of two implying a γ of 0.5. Using the tabulated V_s values given by Penning and Addink[4] and the α values given by Kruithof and Penning, the mean value of $e^{\alpha_s\delta}$ at threshold is 106 if one includes all the Penning and Addink sparking data from $p\delta=250$ to 2 mm×cm; if the Penning-Addink sparking data are taken for $p\delta=250$ to 25 mm×cm, $e^{\alpha_s\delta}$ is only 44. Thus on this basis, γ is of the order of a few hundredths. These considerations show that while our result of two for amplification per avalanche at threshold is very small, the result of Penning and Addink for this multiplication is only an order of magnitude greater. These calculations give striking confirmation to the conclusion that a low-order Townsend discharge precedes breakdown near the threshold voltage.

[10] These calculations were made only for those cases for which the measured formative time lags exceed the calculated positive ion transit time. Actually for every time-lag *vs* percentage-overvoltage curve, about half of the observed time lags are shorter than the transit time of the positive ion. For several curves, all of the observed time lags are shorter than the transit time of the positive ions.

[11] E. J. Lauer, J. Appl. Phys. **23**, 300 (1952).

[12] Colli, Facchini, and Gatti, Phys. Rev. **80**, 92 (1950).

[12] T. Holstein, Phys. Rev. **72**, 1212 (1947).

[13] Alpert, McCoubrey, and Holstein, Phys. Rev. **76**, 1257 (1949).