Temporal Growth of Ionization in Gases

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The temporal rate of growth of ionization in uniform electric fields E in hydrogen at pressures p up to some cm Hg was measured over a wide range of E/p between 50 and 400 v/cm mm Hg when the values of the electric fields exceeded that corresponding to the static sparking potential by a few percent. Comparison of the experimental data with a mathematical analysis of the growth based upon the action of primary and secondary ionization processes enabled the relative significance of possible secondary processes to be assessed, and their absolute values to be determined. These results showed that there was a change in the relative importance of the secondary processes as E/p changed. Thus, for low values of $E/p(\sim 50)$ the predominant secondary process was found to be photoelectric emission from the cathode, but as E/p was increased the relative importance of electron emission from the cathode due to the incidence of positive ions also increased until at high values (~ 300) 50% of the emission was due to this cause. At all values of E/p investigated, no single secondary process was exclusively responsible for the cathode emission.

I. INTRODUCTION

THE temporal growth of ionization currents in gases under uniform electric fields results from the actions of the primary α process of ionization by collisions between electrons and gas molecules and of various secondary ionization processes acting in the gas or at the cathode. These secondary processes can be designated[1] collectively by a generalized coefficient ω/α. In this work the coefficients α and ω/α were measured in conditions made as nearly as possible the same as those in which the temporal growth of ionization was measured, to enable the theory[2,3] of ionization growth to be related to experimental observations.

The magnitude of a small ionization current in a gas between parallel plane electrodes distant d apart for values of electric field E less than a certain critical value E_s is determined by the value of the initial electron current I_0. When I_0 is constant the current I is proportional to I_0 and determined by the values of the coefficients α and ω/α. If I_0 ceases, the current I also ceases as long as $E<E_s$. If, however, E is put equal to E_s, a small ionization current, e.g., $\sim 10^{-7}$ A, can become self-maintained, i.e., remain finite when the initial current I_0 is reduced to zero; the secondary processes set up a replacement condition which maintains the discharge. The potential difference $V_s(E_s \times d)$ at which this condition occurs is called the static breakdown potential, it is independent of the value of the self-maintained current provided the latter is not large enough to form a space charge which significantly distorts the field. In practice it is not always easy to examine the current I when V is exactly equal to V_s because small fluctuations in V can make $V>V_s$, even if only by a very small amount. When $V>V_s$, the current will grow in both space and time; in addition, the temporal rate of growth will increase to a higher rate than that occurring when I_0 is finite and V just equal to V_s.

Consideration of this temporal growth of current in hydrogen between parallel plate copper electrodes in the nonsteady state (when $E>E_s$) over a wide range of the parameter E/p from 50 to 400 v/cm mm Hg is the subject of this paper. The work is confined to lower values of the difference $(E-E_s)$. The use of hydrogen avoids complications due to the action of metastable atoms, and the low gas pressures used minimize the action of photoionization of the gas. In these conditions, ω is given simply by $\omega=\alpha\gamma+\delta$ where γ represents the average number of electrons liberated from the cathode per incident positive ion and δ/α is the average number of photoelectrons released from the cathode per ionizing collision in the gas.

II. PREVIOUS INVESTIGATIONS

Earlier investigations of current growth in gases at low gas pressures include those of Steenbeck[4], Schade,[5] Bartholomeyczyk,[6] and von Gugelberg.[7] Both Steenbeck and Schade developed expressions for the growth of current with time based on the assumption that the only secondary process was the γ process. Bartholomeyczyk included photoelectric emission from the cathode (δ process) in his consideration of the possible ionization processes operative. Von Gugelberg extended Bartholomeyczyk's solution to make some allowance for the initial current I_0 and examined experimentally the growth of current in the noble gases, hydrogen and nitrogen under uniform and nonuniform electric fields between electrodes of various materials. Gugelberg made some estimates of the contribution of photon action to the total secondary emission, for example, in hydrogen, for one particular value of E/p of about 100 v/cm mm Hg.

[1] F. Llewellyn Jones, and A. B. Parker, Nature **165**, 960 (1950).
[2] Dutton, Haydon, Llewellyn Jones, and Davidson, Brit. J. Appl. Phys. **4**, 170 (1953).
[3] P. M. Davidson, Phys. Rev. **99**, 1072 (1955).
[4] M. Steenbeck, Wiss. Veröffentl. Siemens-Werken **9**, 43 (1930).
[5] R. Schade, Z. Physik **104**, 487 (1937).
[6] W. Bartholomeyczyk, Z. Physik **116**, 235 (1940).
[7] H. L. von Gugelberg, Helv. Phys. Acta **20**, 250 (1947).

More recently, Davidson[2] in this laboratory pointed out that Bartholomeyczyk's solution, in addition to neglecting I_0, did not satisfy the initial condition that there should be no space charge in the gap at $t=0$, but explained how Bartholomeyczyk's solution may be altered to satisfy the continuity equations and the correct boundary conditions (but still not the correct initial conditions). Davidson's solution for this case is

$$I_-(x,t)\exp(-\alpha x)=I_0/P+C\exp[\lambda(t-x/W_-)], \quad (1)$$

and

$$I_+(x,t)=\int_x^d \alpha I_-(0,t+x/W_+-x'/W)e^{\alpha x'}dx', \quad (2)$$

where

$$P=1-\gamma(\exp\alpha d-1)-[\delta/(\alpha-\mu)](\exp(\alpha-\mu)d-1).$$

C is not exactly specified. The constant λ is determined from the relation

$$1=\alpha\gamma[e^{(\alpha+\lambda/W)d}-1]/(\alpha-\lambda/W)+\delta[e^{(\alpha+\lambda/W_-)d}-1]/(\alpha-\lambda/W_-), \quad (3)$$

where $(1/W)=(1/W_-)+(1/W_+)$. $I_-(x,t)$ and $I_+(x,t)$ represent the electron and positive ion currents at a point distant x from the cathode at time t, W_- and W_+ are the electron and ion drift velocities; μ is the absorption coefficient for radiation in the gas. As Davidson pointed out, this solution is still only approximate since it involves a charge distribution of ions and electrons in the bulk of the gas at $t=0$, which is not, in fact, present. Consequently, Davidson[2,3] also gave an exact solution which, in addition to satisfying the continuity equations and the correct boundary conditions, also satisfies the initial conditions exactly. It is essential to use Davidson's exact expressions for assessing the relative contribution of the various regenerative processes occurring in the earliest stages of growth, but his approximate expressions are convenient for application to the later times $\gtrsim$, say 10 electron transits.

III. METHOD OF EXPERIMENTAL INVESTIGATION

A steady approach voltage V_a, somewhat less than the previously determined static sparking potential V_s, is applied to a parallel plate discharge gap. At the instant when a group of photoelectrons are liberated from the cathode a step-function voltage impulse V_p is superimposed upon the static approach voltage V_a so that the total potential difference between the electrodes is suddenly raised to (V_a+V_p) to exceed V_s. The gap then becomes, in effect, a leaking condenser charged to a potential difference (V_a+V_p). Under the influence of the total applied electric field $(V_a+V_p)/d$, the initiatory photoelectrons generate a current which increases along the gap distance and also in time at any given plane. When the current density becomes sufficiently high, the voltage across the gap collapses because the gap capacitance is discharged owing to the presence, motion, and arrival of charges generated in the gap at the electrodes and a space-charge controlled glow discharge is set up at a maintenance potential V_{glow} which is lower than the total applied voltage.

The time interval t measured from the instant of application of the voltage impulse to the instant when the collapse occurs in displayed on a cathode ray oscilloscope and recorded photographically. This time t is known as the formative time lag in work using impulsive fields.[8–10]

Davidson's expressions may be used to calculate this time and make it possible to draw theoretical curves relating the formative time t to the voltage applied to the gap (or preferably the excess voltage ΔV by which the gap voltage exceeds the static breakdown potential V_s) provided the values of the primary and secondary coefficients are known. Families of theoretical curves can then be drawn corresponding to different valus of the ratio $(\delta/\alpha)/\gamma$ and compared with experimental data. Thus with the complete theory of Davidson,[2,3] measurement of the time which elapses between the application of the voltage and the instant at which the voltage collapse begins leads to a quantitative assessment of the proportions of the secondary processes. This is the theoretical basis of the present work.

IV. APPARATUS

(1) Discharge Tube

This consisted of two parallel-plate copper electrodes 5 cm in diameter and 1.5 cm apart mounted centrally in a borosilicate glass envelope which was connected to a mercury-free vacuum and gas system. The electrodes edges were rounded to prevent local field intensification. The electrodes themselves were washed in grease solvents and afterwards carefully polished with successively finer grades of emery paper down to grade 0000 and finished on "Selvyt" cloth with grade 3/50 microalumina as polishing agent; before sealing into the glass envelope they were thoroughly degreased, washed in distilled water, and finally dried.

In order to ensure that the voltage impulse V_p applied to the gap would have the necessary short time of rise, all inductance paths were reduced to a minimum. The condenser through which the impulse voltage was applied to the anode was made an integral part of the discharge tube envelope. The glass wall of the envelope formed the dielectric between two silver plates; the inner one being connected directly to the anode and the outer plate to the impulse generator. This arrangement had the advantages of giving a condenser with a very high leakage resistance and also of allowing the lead from the impulse generator to the anode to be kept very short. Possible leakage paths were eliminated by the use of guard rings and split insulation. The tube

[8] W. Rogowski, Arch. Elektrotech. **20**, 101 (1928).
[9] L. H. Fisher and B. Bederson, Phys. Rev. **81**, 109 (1951).
[10] Aked, Bruce, and Tedford, Brit. J. Appl. Phys. **6**, 233 (1955).

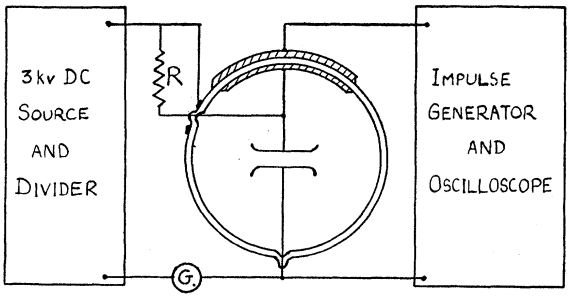

FIG. 1. Schematic diagram of discharge tube and circuit.

assembly and connections are shown diagramatically in Fig. 1.

(2) Regulated High Tension Source and Potential Divider

This was used to determine the static sparking potential V_s and to provide the approach voltage V_a, and comprised the following parts: (a) a source of static potential of 3000 volts, and (b) a potential divider.

(a) The static potential was obtained by the rectification and smoothing of an already stabilized ac 50 cps mains voltage supply. The rectified and smoothed output was electronically stabilized using a series-parallel stabilizing circuit with a high reference voltage.

(b) The regulated output from this source was applied across the potential divider which took the form of a 3-megohm Raleigh potentiometer. In this way, the static potential applied to the gap via a 10^7-ohm series resistor R which served to limit the discharge current, could be varied up to the maximum value of 3000 v in steps of 0.1 volt. The output was calibrated against a standard cell. The long term stability was better than 0.1% and with continuous monitoring a stability of better than 0.01% was obtained. The absolute accuracy at any given potential output was 0.1% since the resistances making up the Raleigh potentiometer were made to this order of accuracy. Every source of static potential used in this investigation was electronically stabilized. The ac supply to these static sources was also stabilized by means of constant-voltage transformer inserted in the mains supply system. A high degree of steadiness in every voltage was, therefore, maintained throughout the investigation.

(3) Impulse Generator

This generated the step-function overvoltage impulse V_p of amplitude continuously variable between zero and 20 v. The time of rise was 2×10^{-8} sec and the amplitude remained constant to within 1% for $>10^{-3}$ sec in the absence of a high load. The total applied voltage remained constant to within 0.1% for $>10^{-3}$ sec. The error in the measurement of V_p was $<1\%$.

(4) High Speed Cathode Ray Oscilloscope and Camera

The time-base sweep speeds ranged from 10^{-3} sec to 10^{-8} sec; the time-base generator being a modified form of that described by Hardy.[11] Oscillators, which were either themselves crystal-controlled or calibrated against the crystal-driven frequency standards, were used to calibrate the time bases. The photographic recording was done using a specially constructed camera incorporating a "Wray" $f1/0$ lens.

(5) Elimination of Statistical Time Lags

In order to eliminate statistical time lags, an adequate supply of initiatory electrons was provided by x-ray irradiation of the cathode from a self-rectifying soft x-ray generator.

V. EXPERIMENTAL PROCEDURE

Careful determination of the static breakdown potential V_s is important for various reasons: it is necessary for defining ΔV and also in order to find ω/α from the breakdown criterion in the neighborhood of static breakdown. Further, its stability serves as a very sensitive indication of the stability of the electrode surfaces. The establishment of conditions such that V_s is steady and accurately measurable is a *sine qua non* for any reliable quantitative assessment of the role of the secondary processes. The experimental procedure adopted in the present work is as follows. Hydrogen prepared by the electrolysis of a saturated solution of pure barium hydroxide in distilled water was dried by storage over phosphorous pentoxide, diffused through a palladium osmosis tube, and passed through tubes immersed in liquid oxygen before admission to the evacuated discharge tube. The tube itself had been given preliminary outgassing treatment by heating during continuous evacuation. Liquid oxygen traps were connected to the discharge tube throughout the experiments; pressures were measured with a vacuum-oil manometer. After allowing the gas to equilibrate in the system, the static breakdown potential V_s was determined. The potential difference across the gap was raised in steps of 1 volt in the neighborhood of the expected value of V_s and initial ionization provided by x-irradiation of the cathode. The ionization current flowing in the discharge gap was measured with a galvanometer G and the potential V at which this current ($\sim10^{-7}$ amp) became self-maintained when the irradiation was cut off was in these conditions usually clearly defined and easily reproducible. This value of V, by definition, was the static breakdown potential V_s. The potential difference was then reduced to the approach voltage V_a of value about (V_s-4) volts, and the overvoltage impulse V_p was applied in synchronism with the release of initiatory electrons at the cathode.

[11] D. R. Hardy, Brit. J. Sci. Inst. **29**, 241 (1952).

The impulse rise and subsequent collapse of the gap voltage was displayed on an oscilloscope screen and recorded on film. This permitted the value of the formative lag t to be determined using the calibrated time base. Between thirty and fifty observations were made and recorded at any one setting of the potentials and gas pressures. At each setting of pressure and approach voltage, measurements were made for different values of V_p. Thus the dependence of t upon the percentage overvoltage $\Delta V\%=100(V-V_s)/V_s$ could be determined. In general, values of $\Delta V\%$ from 0.2% to about 5% were employed because these values covered a wide range of formative times t.

After a succession of such measurements at different overvoltages, the gas pressure p was reduced, and the whole experimental procedure repeated. The time measurements for another set of overvoltages corresponding to the new V_s were then determined as before. In addition, the final value of the voltage across the gap (V_{glow}) after the collapse had occurred could be estimated from the photographic records and was determined for the same range of E/p as the growth times.

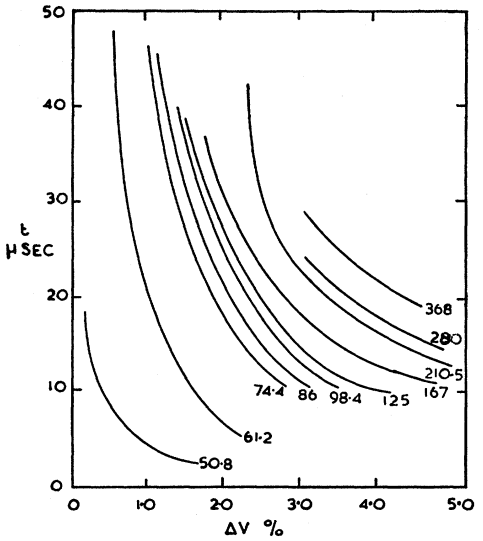

FIG. 2. Formative time lags in hydrogen. Dependence of growth time t upon overvoltage for copper electrodes after 36 hours treatment. (The numbers on the curves here and in Fig. 3. refer to the corresponding values of E_s/p.)

VI. RESULTS AND CONCLUSIONS

Preliminary observations[12] were made using the electrodes and tube which had received only preliminary outgassing (and were therefore still covered with a tarnish film). These showed that for a given value of $\Delta V\%$ the growth times increased with E/p. For example, with low values of E/p ($\lesssim 100$ V/cm mm Hg) the times t were $\sim 10^{-6}$ sec even when $\Delta V\%$ was as low as 2%, whereas, at larger values of E/p (~ 400 v/cm mm Hg) the times t were $\sim 10^{-4}$ sec with the same value of $\Delta V\%$. While there was a tendency for longer times to occur when E/p was large, there did not appear to be a systematic change in the relative position of the $(t,\Delta V\%)$ curves as E/p changed.

At any given value of overvoltage and gas pressure, the formative times t were not exactly constant; this scatter in the values of t was most noticeable, as was the difficulty experienced at first in obtaining consistent values of V_s (to which the amount of scatter appeared to be related), when the electrodes used had received only preliminary cleaning treatment. The average scatter observed in these preliminary observations corresponded to a standard deviation of about 10%, the worst was 40%, but this was exceptional. However, in the later experiments with cleaned electrodes and by carefully and continuously monitoring the stabilized voltages, the standard deviation was reduced to less than 5% at low overvoltages ($\lesssim 0.5\%$). At higher overvoltages ($\gtrsim 2\%$), a standard deviation of less than 2% could be obtained. It is considered necessary to reduce the scatter to this order if reliable quantitative data on the secondary processes is to be obtained. The lack of consistency of the preliminary data on growth times is considered to be due to unstable electrode conditions caused by the presence of surface contaminating films.[13] Accordingly, attempts were made to obtain conditions in which the coefficients α and ω/α were constant by careful purification of the gas and also by suitable treatment of the electrode surfaces. A program of intensive electrode treatment by hydrogen bombardment in a low pressure glow discharge followed by prolonged heating at 350°C during continuous evacuation was undertaken and observations were made at different stages in the treatment.

The effect of this treatment was marked. Results finally obtained after the treatment are shown in Fig. 2 as full lines which have been drawn through the experimental points. It is seen that after treatment, the progression to longer time lags as E_s/p was progressively increased, was now quite regular. Simple considerations suggest that a change occurs in the relative importance of the secondary processes involved in the ionization growth as E_s/p is changed. Thus, with low values of E_s/p when the observed time lags were short, the predominant regenerative process appeared to be a fast process, such as photon action. On the other hand, the longer time lags obtained when E_s/p was large suggest that the slower process of positive ion action at the cathode was mainly responsible for the secondary ionization. Although this general result follows from simple considerations, it is nevertheless important to determine the precise proportion of the secondary processes. This can only be done by a more rigorous analysis on the basis of the theory outlined above in II, and was carried out as follows.

[12] C. Grey Morgan, Appl. Sci. Research **B5**, 18, (1955).

[13] F. Llewellyn Jones and D. E. Davies, Proc. Phys. Soc. (London) **B64**, 397, 519 (1951).

A set of theoretical $(t,\Delta V\%)$ curves must be calculated assuming various proportions of the parameter $(\delta/\alpha)/\gamma$ representing the relative proportions of the two different secondary processes (photons to ions). These theoretical curves were obtained by using the method described by Dutton, Haydon, Llewellyn Jones, and Davidson[2] by the following procedure.

First, the ratio $I_-(o,t)/I_0$ in Davidson's Eq. (1) must be determined for a set of experimental data at a given value of E_s/p, (i.e., V_s/pd). A value of this ratio is found by first assuming a particular value, say 0.5, of the parameter $(\delta/\alpha)/\gamma$ and, by substitution and successive approximation using Eq. (3), the constant λ is determined. Taking an experimentally determined value of t at any given value of $\Delta V\%$ in the set of experimental data as a reference point, and using known values of α, of ω/α, of W_+,[14] of $(\delta/\alpha)/\gamma$ and the calculated value of λ the ratio $I_-(o,t)/I_0$ is then obtained from Eq. (1). The ratio is sharply dependent on the ratio $(\delta/\alpha)/\gamma$ and this fact restricts the range of values of $(\delta/\alpha)/\gamma$ which give reasonable values of $I_-(o,t)/I_0$. Having determined $I_-(o,t)/I_0$ for the set in this way, it can then be used in Eq. (1) for the calculation of further values of t corresponding to other values of $\Delta V\%$ in the set. The theoretical t, $\Delta V\%$ curve so obtained is then compared with the set of experimental data. If there is no agreement a different value of $(\delta/\alpha)/\gamma$ is assumed and the process repeated until agreement between the calculated and measured values is obtained at all points. The stringency of this condition renders calculation of the relative proportion of the secondary coefficients reliable. The value of $(\delta/\alpha)/\gamma$ for which the agreement is obtained can then be regarded as characteristic of the electrodes. This procedure renders the direct measurement of I_0 unnecessary, and the complication of its measurement was not considered advisable when this work was undertaken. However, as a check, I_0 was measured in later work and found to be about 10^{-10} A. When this value is inserted in Davidson's growth equations[2,3] the magnitude of the current in the gap at the instant when the gap voltage begins to collapse can be calculated and it was found to be some microamperes. These calculated values of the gap current are in agreement with our direct measurements on the controlled development of glow discharges.[15]

Experiments to determine the influence of the initial cathode current upon the value of t were carried out by varying the approach voltage V_a over a wide range for a constant overvoltage. No significant change in t was observed in spite of the fact that the initial amplified current had been changed by a factor of $\sim 10^3$.

Consider now the consistent $(t,\Delta V\%)$ data obtained after the electrodes had been treated long enough (6 hours) to produce sufficiently stable surfaces; these data are given by the circles and dots in Fig. 3 for three distinct values of E_s/p; viz, (A) 50, (B) 175, and (C) 300 v/cm mm Hg. Three distinct families of theoretical $(t,\Delta V\%)$ curves were calculated in the manner indicated above and corresponding to these three values of E_s/p (50, 175, and 300) for which the experimental points were obtained. The curves which formed the best fit to the experimental points are drawn as full line curves in Fig. 3. These curves corresponded to the following proportions of γ and δ/α; for curve (A), $(E_s/p=50)$, γ, 25% and (δ/α), 75%; for curve (B), $(E_s/p=175)$, γ, 50% and (δ/α), 50%; and for curve (C), $(E_s/p=300)$, γ, 50% and (δ/α), 50%.

The degree of sensitivity of this procedure for finding the relative significance of the two secondary processes is shown by the difference between the broken line curves of Fig. 3 and the full line curves. The broken lines correspond to a change of only 5% in each of the above values which give agreement; the broken lines

TABLE I. Relative proportions of γ and δ processes for treated electrodes.

E_s/p (v/cm mm Hg)	% (γ)	% (δ/α)
50.8	25	75
61.2	28	72
74.4	30	70
86.0	35	65
98.4	40	60
125.0	40	60
167.0	40	60
210.5	40	60
280.0	40	60
368.0	50	50

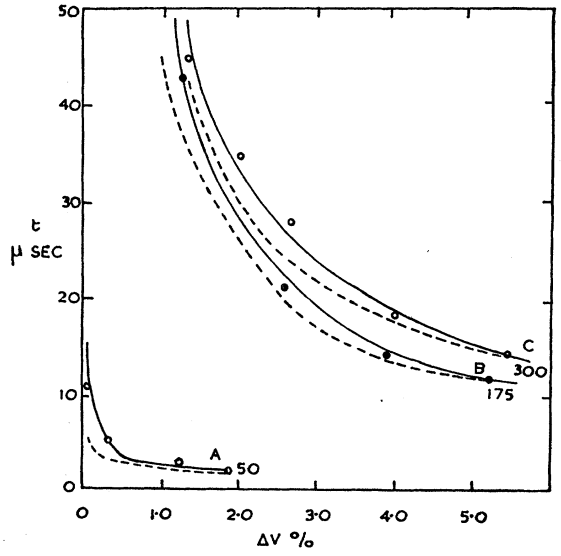

FIG. 3. Comparison of calculated and experimental growth times. ● ● ● and o o o experimental values; —— and – – – calculated curves.

[14] N. E. Bradbury, Phys. Rev. **40**, 508 (1932).

[15] F. Llewellyn Jones, *Encyclopaedia of Physics, Gas Discharges II* (Springer-Verlag, Berlin, 1956), p. 10, Fig. 9.

show a pronounced departure from the experimental points.

The electrodes were treated for a further 30 hours in an attempt to obtain cleaner surfaces and thus more stable conditions. Further sets of measurements were then made and the data are those which have been given in Fig. 2. These sets were also analyzed, the results being given in Table I.

These results show that in no case was the secondary ionization due entirely to one process, whether of photons or ions. Further, the relative significance of the γ and δ processes changed with E/p. The action of the positive ions appears to become increasingly important at the higher values of E_s/p but was still substantial even at the lowest values of E_s/p examined.

As an indication of the stability of electrode surfaces obtained by the treatment of ion bombardment and subsequent heating, it is interesting to note the agreement between the values of the secondary coefficient (ω/α) here obtained and those previously obtained for copper electrodes in hydrogen in this laboratory using different treatment of the electrodes.[13,16] This is shown in Fig. 4. The reasonably good agreement between these three sets of results over a period of years indicates that a stable electrode surface was attainable by the procedure of hydrogen ion bombardment and prolonged heating at about 350°C. However, that the treatment probably still had not produced the cleanest possible surface in the present case is indicated by the data (broken line) obtained by Llewellyn Jones and Davies[13] by still more rigorous treatment continued for over 160 hours when the electrodes were heated to red heat; this prolonged treatment resulted in much lower values of ω/α.

In order to perform the numerical computations to obtain the theoretical curves of Fig. 3 and the data in the table, it was necessary to determine the values of the generalized secondary coefficient (ω/α) corresponding to the various conditions at the different values of E/p. These values were obtained from the measured values of the static sparking potential V_s under the stable conditions, using known values of the primary coefficient α. For this reason, complementary measurements of α were made in a subsidiary apparatus.[17] The spatial growth of current in the steady state regime $(E \leqslant E_s)$ in this work was found to be described by the well known Townsend relation $I=I_0 \exp(\alpha d)/\{1-(\omega/\alpha) \times[\exp(\alpha d)-1]\}$ and the static breakdown criterion was given by $1-(\omega/\alpha)[\exp(\alpha d)-1]=0$ in agreement with previous experiments.[18]

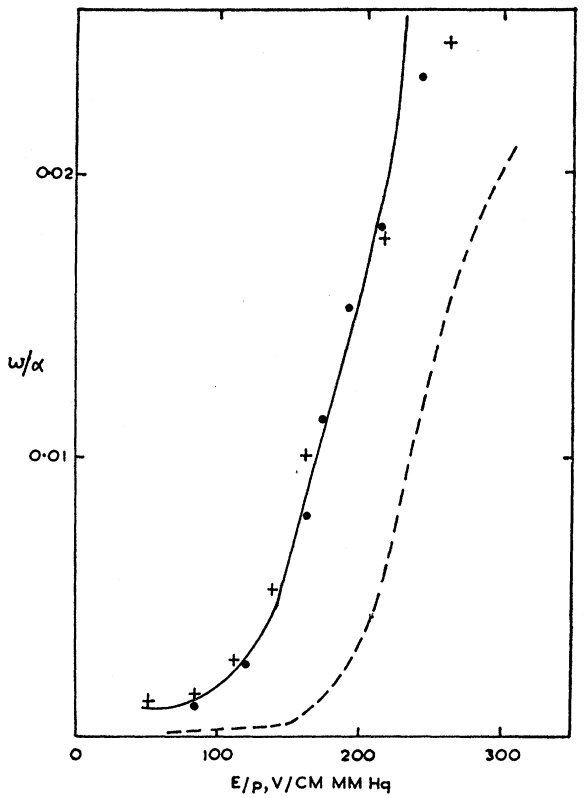

FIG. 4. (ω/α), (E/p) curves for copper electrodes in hydrogen after various treatments. ———— Llewellyn Jones[16]; ● ● ● Llewellyn Jones and Davies[13]; + + + present work.

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[16] F. Llewellyn Jones, Phil. Mag. **28**, 192 (1939).

[17] The values of (α/p), $f(E/p)$ obtained in the present work were in good agreement with those of D. J. Rose, Phys. Rev. **98**, 558(A) (1955).

[18] Dutton, Crompton, and Haydon, Proc. Phys. Soc. (London) **B69**, 2 (1956).