COUNT RATE-EMISSION WITH TIME OF EXPOSURE TO AN ELECTRODELESS DISCHARGE IN AIR*

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A periodic phenomenon has been observed in the variation of current pulses in terms of counts with time of exposure during non-varient nature of the field gradient through air in a Siemen's type ozonizer. The dark counts, presented in the plots, at a temperature of 50°C for pulses of 5 and 50 V are initially large at the first instance of recording and then decrease or increase rapidly. The theoretical explanation for the various observed features has been attempted on the basis of the formation of the charge inequality and equality conditions in a plasma.

The marked importance of the voltage distribution in the zone of intense ionization in determining the current has been emphasised by Penning[1] and others[2]. It controls the number (and therefore current) of avalanches formed by initiating electrons produced by any of the agencies. Below the threshold voltage, the field-intensity E is linear with the distance separating the two electrodes, i.e., outer electrode (cathode C) and inner electrode (anode A) in Fig. 1(b), when the applied V is large enough to cause a self-maintained discharge, it varies differently. In view of the appreciable variation of current pulses of a self-sustained discharge, the system shows the emission decay-phenomenon and periodic effect[3]. The present communication points out the significance of a few results obtained during the studies of count rate-emission with time of exposure to the discharge in air at a pressure of 10 Torr for a definite pulse height. These indicate clearly the behaviour of the field gradient under conditions giving large periodic effect[3].

EXPERIMENTAL

The electrical discharge was produced in the annular space of a Siemen's type ozonizer formed by two coaxial cylindrical glass tubes. The inner and outer cylinders, having outer diameter of 13 and 17 mm respectively, form the dielectric electrodes. The discharge vessel and the electrical connections are shown in Fig. 1(a).

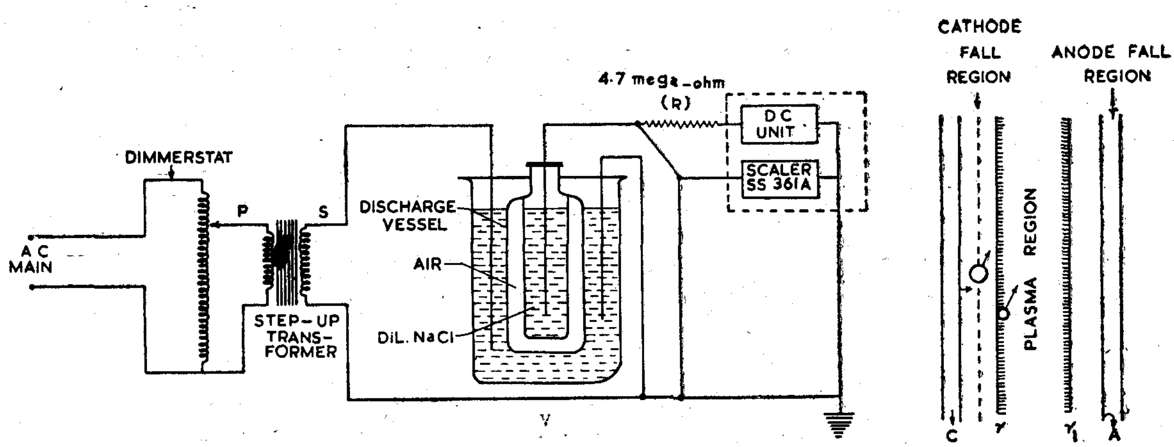

Fig. 1 (*a*)—Experimental arrangement to obtain a periodic effect in the count rate time characteristics at 50°C for pulses of 5 and 50 volts.

(*b*)—Plasma region in an induced discharge at a definite continuous potential.

*The work reported here was carried out in the Chemistry Department, University of Poona.

A step-up transformer (ratio 1 : 70) supplied AC power at 50 Hz for maintaining a constant temperature of 50°C of the electrolytic bath. The output of the dimmerstat was connected to the primary of step-up transformer, from where a secondary potential of the required V was obtained by means of a division of about 60 on the calibrated scale of the dimmerstat. One probe of the high tension transformer was immersed into the outer tube of the vessel containing salt solution as electrolyte and the other was grounded.

The inner electrode was formed by filling *NaCl* solution in the inner tube of the vessel and connecting it through a resistance of 4·7 mega-ohm to a stabilized positive high voltage supply of a scaler unit (fabricated by the Atomic Energy, Trombay, Bombay), the other electrode containing the same electrolyte was grounded through a counter scaler.

While measuring the discharge pulses in terms of counts for pulses of 5 and 50 volts with time of exposure to an electrodeless discharge, it was noticed that the data under the present conditions were not always reproducible. The discharge counts in perfect darkness for a given counting time, abbreviated by the symbol *CD*, are given in arbitrary units.

RESULTS

Comparison of Earlier and Present Results

Earlier results on pulse height spectral analysis in ozonizer discharges excited by 50 Hz potentials through nitrogen[4], oxygen[4] and argon[5-7] in dark and on illumination by visible light have shown that when a gas is subjected to an electrical discharge at a constant temperature, the charge carried by the pulses in terms of counts to the electrodes (charge transfer) decreases under illumination. Decrease of the discharge current has been observed in the case of pure argon subjected to an electrical discharge in the presence of electrolytic films.

The data for decrease of dark counts with temperature keeping the discriminator bias constant were also recorded in air at a definite pressure by the author[8]. Details of these observations, reported by Arnikar, et al.[5] and by the author[7], are compared with the present observations. In the present series of experiments, the influence of time of exposure to the electrodeless discharge on the discharge counts in perfect darkness for a given counting time has been studied by exposing dry air at a temperature of 50°C to different times of exposure ranging from about 0–735 min. At a constant applied continuous potential of 1500 V, the counts in dark for a counting time of 5 min were observed for pulses of 50 V selected by a discriminator. The data are presented graphically by a solid curve in Fig. 2. The discharge counts were expected to be either a progressive or constant decrease of counts, depending upon the pulse height and the temperature of the system. Actually, it was found that at a steady potential of 1500 V at a temperature of 50°C, the dark counts per five min showed an initial marked fall and reached to a minimum and then rose to a maximum, to fall and rise once again, thereafter the counts reached the minimum stationary value. The conditions for the development of this periodic effect are such that the corresponding progress of the change in the count rates is rather slow, as compared to the earlier periodic effect[8], and is rather rapid as compared to the dotted curve for pulses of 50 V. Thus, for instance, in one case as many as three well defined recurrences of discharge counts and of the other electrical quantities mentioned already were obtained necessitating an exposure of 735 min to the electrodeless discharge.

After the first set of results, the pulse height of 5 volts selected by a discriminator was used for the second set of results. The discharge counts in dark for a counting time of 2 min were then restarted and the variation of discharge counts at 50°C was noted. The data obtained with time of exposure to the electrodeless discharge are presented in Fig. 2. It was interesting to note that the count rate was initially large and increased rapidly with time. Thus, in air at 10 Torr, the dark count rate was 169 at 0 min and 132 at 105 min; further exposure to the electrodeless discharge did not alter the count rate appreciably. After the exposure of 670 min, there was slowing down of the change in the count rates, indicating of a steady state.

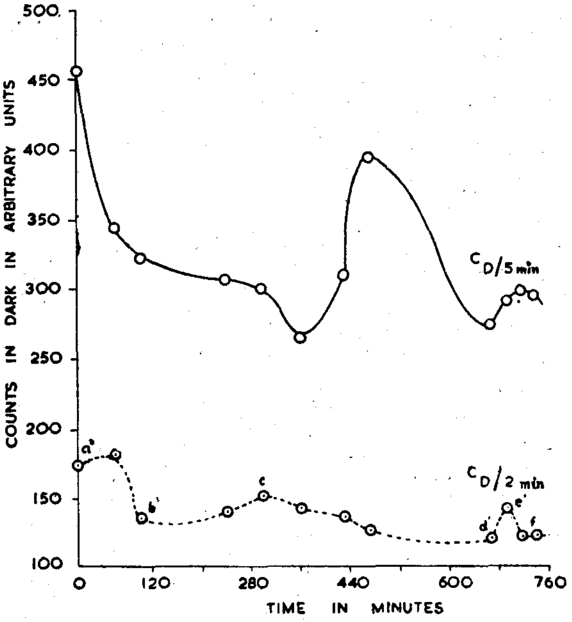

Fig. 2—Variation of dark count rate with time of exposure to the electrodeless discharge; pressure of air 10 Torr; temperature of electrolytic bath 50°C; potential applied to the system 1500 V (DC). (Solid curve reports a bias of 15 V and dotted curve reports a bias of 5 V).

Comparative Influence of 5 and 50 Volts-bias on the Dark Counts

For initial time of exposure, the counts in dark for 5 min depicted in a solid curve and the counts in dark for 2 min depicted in a dotted curve (Fig. 2) are initially large at the first instance of recording. Further time of exposure to the discharge, decreases the dark counts to a minimum in a solid curve and increases to a maximum in a dotted curve.

It may be of interest to note (Fig. 2) that the discharge counts, $C_D/5$ min, are found to be more at 0,480 and 720 min and lower at 360 and 670 min; whereas the discharge counts per two min in a dotted curve are found to be large at 60, 300 and 690 min and lower at 0,105 and 670 min. The difference between a solid and a dotted curve is that at a bias of 50 V, the steep rise of discharge counts in a solid curve for higher time of exposure becomes more pronounced.

Again with the smaller time of exposure, the peak at 690 min in dotted curve is shifted in the direction of lower values of time to about 30 min for a convergent field (central wire positive).

Both the curves show negative slope in the falling characteristics of count rate versus time of exposure to the discharge. The negative characteristic in the plots can be practically realized only in the limited area of the count rate and time of exposure-range. The other characteristics of the curves are, therefore, limited by areas with positive dCD/dt. These two sections, notably positive and negative, can be mathematically expressed as

$$\frac{dC_D}{dt} > 0 \text{ and } \frac{dC_D}{dt} < 0 \text{ respectively.}$$

Interpretation of Variation with Time of Exposure of Counts

It was of interest to note that in the dark, two types of current pulses were observed at $V > V_{ga}$, (i) the large initial maximum currents produced by the secondary mechanism[9] concerned with the liberation of electrons from the cathode chiefly by positive ionic bombardment γ and photons $\eta\theta g$ and (ii) the lower minimum current due to the other secondary process β occurring in the gas phase[10]. In the later case photo-ionisation of the pre-excited gas particles in the cathode fall region appeared significant[11,12]. That is, an electron formed by this process acquires energy because of the field gradient and starts ionisation by inelastic collisions after travelling a known distance away from the cathode in the field direction. The generated electron repeats the process forming electron-avalanches. The number of electrons produced in the second process is fundamentally governed by the field gradient and determines the current pulses. The production of lower values in current pulses for essentially the same experimental conditions suggested that electrons generated by photo-ionisation in the above mentioned region, travelled through sensibly the same potential difference and ionised gas particles from a fixed point r away from the cathode[12] (Fig. 1). It has been emphasised that the ionisation process between r and r_1 is not excluded, but occurs by fast electrons released from the outer electrode by γ and $\eta\theta g$ mechanisms[11]. The significance of the counts noticed under perfect darkness can be explained as steady state lower current pulses, remarkably of nearly the same number as those appearing in the dark. This suggests that the aided creation under Debye's shielding-sheath mechanism of initiating electrons in the cathode fall region causes ionisation by impact with gas particles from the same point r to give the enhanced number of the lower values of current pulses. It can occur only when the average electron-number density n_e and the average ion-number density n_i are nearly equal, and active plasma of electrons is very much greater than active plasma of ions.

DISCUSSION

Electrons created in the discharge space surrounding inner cylinder of the ozonizer chiefly in the neighbourhood of the cathode acquire energy under field and start ionisation by inelastic collisions with atoms and molecules, approximately from a fixed point r away from the cathode (Fig. 1b). The generation of electrons, however, decreases as they recede from the cathode to weaker fields. The ion production continues until the field becomes too weak to cause ionisation by collision. The region between this point (say r_1 in Fig. 1b) and the earlier one r may be referred to roughly as the plasma-region of the discharge. It may be mentioned that ionisation of the gas particles between cathode and the point r occurs rapidly because of moving electrons released from the cathode by bombardment of positive ions and photons.

In the plasma region of the discharge, the net charge transferred to the wall is zero; since in this region the electron and positive ion-densities are equal, under these conditions the ion mantle cannot have a tendency to lose carriers mainly, because the release of carriers would be followed by others, since in the excited gas the charge densities must remain equal. According to this, along the ozonizer's axis, the column is electrically neutral and optically uniform so that positive ion- and electron-densities are very nearly the same and current pulses are constant across any section. Charges are lost from the column by lateral diffusion to the walls where recombination takes place. The electrons tend to diffuse more rapidly than positive ions and due to this, a radial field is set up which retards the electron diffusion and accelerates that of the positive ions. A state of equilibrium is reached whereby charges of both signs are lost at the same rate (ambipolar diffusion) and this is just balanced by the ionisation due to the electron gas molecule collisions in the plasma. Alteration of the field can cause no unidirectional drift of electrons away from the body of the gas therefore secondary source of electrons is not essential in order to maintain the supply of electrons. The mechanism of the discharge then consists of electron generation by the α-process and loss by diffusion, attachment and recombination.

Electron attachment to neutral molecules at a rate proportional to the concentration of both, forms negative ions. The electrons are then not able to produce further ionisation. Presence of the minima in the discharge current pulses as a function of the time of exposure to the electrodeless discharge (Fig. 2) show that this process is present; but is not so predominant in the present case. Holms[13] and Seelinger[14] considered the effect of electron attachment on the diffusion theory of the column. Here it is not permissible to use the normal ambipolar diffusion coefficient[15], Da, nor to take the net rate of production by electrons as $2N$ since they are being removed by attachment. Electrons are formed by direct ionisation and also by collision between the negative ions and gas molecules causing detachment[16], the latter of which seems to be an improbable process in the case of pure dry air.

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