PULSE HEIGHT SPECTRAL STUDIES OF OZONIZER DISCHARGES IN ARGON AT A MODERATE PRESSURE AND THE ASSOCIATED JOSHI EFFECT*

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The effect of light has been observed in the variation of discharge counts with pulse amplitude during d.c. discharges through argon in a fresh and an aged Siemen's vessels. The pulse height spectra of the discharge for applied potentials of 1·75 and 1·82 kV (r.m.s.) have been measured in dark and under irradiation. The number of current pulses varies periodically with increasing amplitude both in the low and plateau—like intermediate amplitude regions; finally the number of pulses drops rapidly to negligible value in the high amplitude regions. The theoretical explanation for the various observed features in the plots has been attempted on the properties of adsorbed layers on the glass walls of the discharge tube.

Though it has been known that ozonizer discharge in gases consists of discrete pulses of varied amplitudes, a precise measurement of these does not appear to have been reported hitherto. A knowledge of the amplitude distribution of the pulses under varying conditions of discharge would be primary to an understanding of their origin. The data would also be helpful in interpreting the phenomenon of light effect (viz. an instantaneous and reversible change $\pm\triangle i$ in the photo-variation of discharge current in gases under discharge due to external radiation[1–7]). The author reports here results of measurements on pulse amplitudes for ozonizer discharge in argon, chosen as a typical system.

EXPERIMENTAL

The discharge vessel consisted of a glass Siemen's ozonizer; filled with pure dry argon, a free electron gas, at a pressure of 10 mm of mercury. The discharge tube was enclosed in a wooden box with a sliding shutter so that external light was completely excluded in order to avoid photo-induced effects[1,8]. The discharge vessel was irradiated by light from a 250 W clear glass bulb when needed. The discharge tube was excited by 50 Hz high voltage provided by a transformer. The low tension electrode of the ozonizer was connected to an electronic counter scaler, with a resistance of 470 ohms in series. Fig. 1 shows the circuit employed.

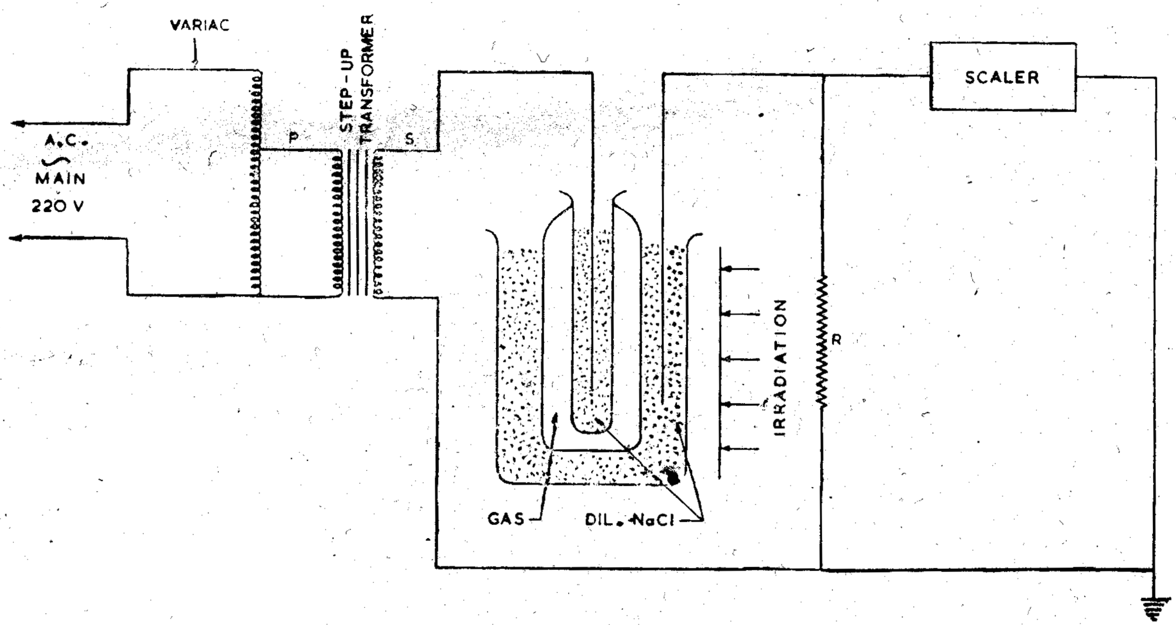

Fig. 1—Experimental arrangement.

The discharge count rates in dark (I_{sd}) and under light (I_{sl}) and the resulting light effect were recorded for each pulse height selected by a discriminator.

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EXPERIMENTAL PROCEDURE AND RESULTS

The discharge tube had been invariably subjected to an aging treatment in order to bring it to similar electrode conditions for all observations as shown in Fig. 2. For this purpose the discharge tube was subjected to a discharge at room temperature 27°C for 15 minutes, at 2.45 kV (r.m.s.). This treatment has been called electro-conditioning by earlier workers[9–11].

The pulse height distribution of the discharge was studied with a fresh and with an aged tube at the applied potential of 1.75 and 1.82 kV in dark and in light. A comparison of the curves obtained before and after irradiation for different periods of electro-conditioning is shown in Fig. 2. Electro-conditioning appears to sensitise the tube for production of the light effect. It was observed that resting for 6 to 12 hours after aging brings the tube back to its original condition. The tube was, therefore, standardized by aging it for a duration of one hour at 1.82 kV every time.

Curves 1 and 2 of Fig. 3 show only the negative light effect and it varies in the range of —(36–98%) for pulses of all amplitudes 5–30 V. They further show that the count rate before and after irradiation for a given potential increases from an initial value to a maximum. This peak point in both the curves is formed at low values of discriminator bias-potential beyond which the count rate diminishes. The pulse height spectrum of the discharge of curves 3 and 4 shows that the number of pulses of the smallest amplitude, however, appears to be a maximum. As the potential is increased progressively, at a constant window width of 5 V, the number of pulses falls rapidly both in the low and high amplitude regions. For instance, under an exciting potential of 1.75 kV, the count rate (curves 3 and 4) falls from about 4280 to 2290 and about 3480 to 1850 for raising the pulse height from 5 to 20 V. Thereafter, both the curves before and after irradiation rise, the value of the second peak in curves 3 and 4 being 3765 and 4410 at a bias of 25 V respectively. Finally in the high amplitude region (> 30 V) the count rate in both the curves 3 and 4 drops rapidly to almost zero, there being no detectable pulse of amplitude higher than 60 V.

On irradiation, there is a simultaneous decrease in the number of short amplitude pulses, and an increase in those larger amplitude pulses. Thus, the light effect ($\triangle I_s$) has a high negative value ($<$ —20%) in the shorter amplitude region, it varies thereafter with the increase of pulse amplitude and changes sign in

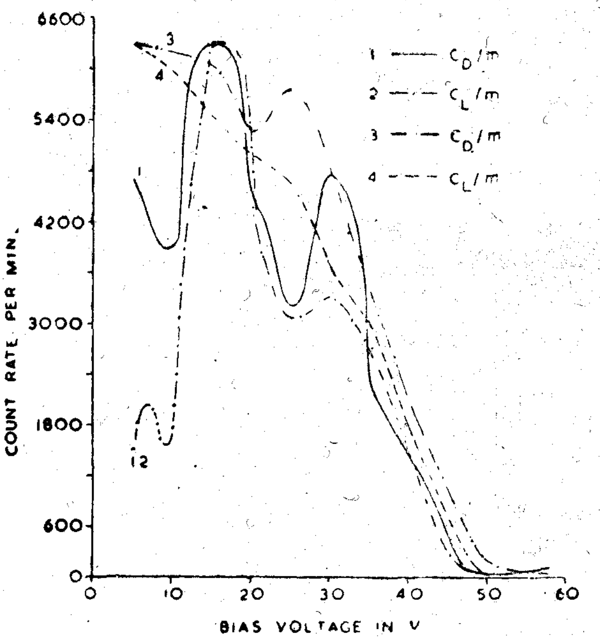

Fig. 2—Variation of count rate with bias voltage. The curves 3 & 4 indicate the variation at an applied potential of 1.82 kV (r. m. s.) with a fixed repetition rate.

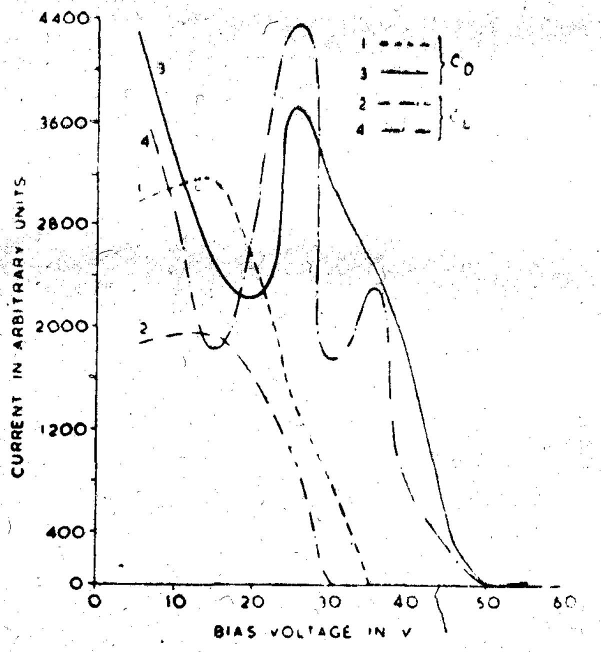

Fig. 3—Variation of the discharge current with the applied bias-voltage. The curves 3 & 4 indicate the variation in dark and under light at 1.75 kV (r. m. s.) with a fixed repetition rate

the region around 20–25 V going over finally to a maximum of +20% for pulses of height exceeding about 15 V (curves 3 and 4 in Fig. 3). Once again this $+\triangle I_s$ inverts to $-\triangle I_s$. These results explain the co-occurrence of $-\triangle I_s$ and $+\triangle I_s$ effects observed earlier, these being associated with pulses of shorter and longer amplitudes respectively. The critical inversion amplitudes are found to be 20 and 30 V respectively. It is remarkable to note (see Fig. 4) that as the discriminator bias potential is increased progressively, the number of counts falls rapidly both in the low and high amplitude regions, with an approximately plateau—like intermediate region (10–30V), the difference between the number of pulses observed with and without irradiation is comparatively larger in this region and much smaller both in the low and high amplitude regions (curves 3 and 4 in Fig. 4).

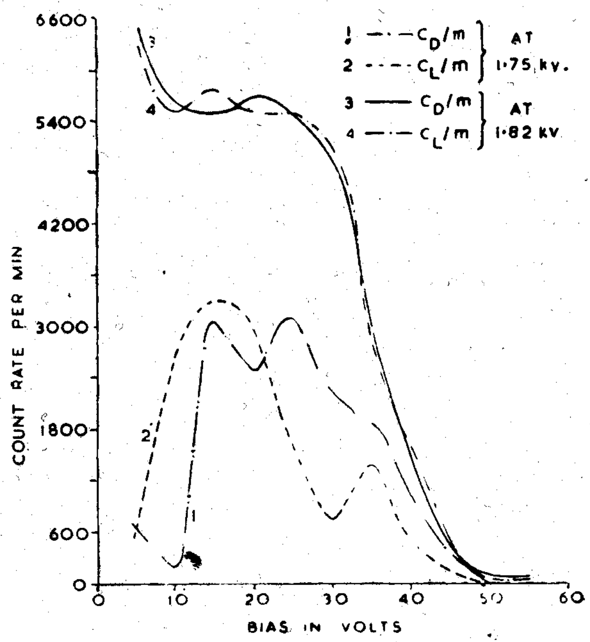

Fig. 4—Bias I_s characteristics at II threshold potential.

The current pulses in argon for convergent field (central wire positive) both in dark and light vary with height of pulses as shown in Fig. 2. The results show that: (1) the count rate in curve 1 falls initially and latter rises until it reaches a peak with bias potential whereas the count rate in curve 2 rises initially and reaches a first peak point; (2) curves 1–4 in Fig. 2 showing a periodic phenomenon support the view that the growth of pulses and the general mechanism of the light effect is not the same in fresh and in aged tubes. The only possibility for this difference is the aging effect which affects the variation of pulses at a given exciting potential V.

At a low discriminator bias potential, the number of ions created by irradiation is probably larger than the number of ions discharged at the electrodes. Since the speed of ions increases with increasing bias potential, an increasing number of ions of both signs is driven to the electrodes and the current rises (see curve 2 in Fig. 2) and vice-versa. In some cases, the pulses decrease with increase of the pulses height within certain limits. The decrease in the discharge current pulses on irradiation suggests that there is probably some mechanism by which a recombination of the ions occurs. Since the probability of ionization by a photon is maximum when its energy just exceeds the ionization potential so that the kinetic energy of the photo-electron is small, it follows that the probability of recombination is the greatest when a slow electron collides with ions. Under such circumstances, the probability of recombination is high. The falling characteristic, i.e. the number of pulses falling with increase of V, is observed during this transition.

INFLUENCE OF IRRADIATION—LIGHT EFFECT

Remarkably enough, the decrease in the number of the longer pulses under irradiation was accompanied by the simultaneous production of shorter pulses (Fig. 2–4); this indicates the co-occurrence of negative and positive effects, $\mp\triangle I_s$ anticipated from altogether different studies with the observations of the potential values of the high pulses and current pulses and their variation with the determinants like external irradiation, its intensity and frequency and the resistance and capacitance of the measuring circuit[12–14], etc.

It may be pointed out that at an exciting potential of 1.75 and 1.82 kV (r.m.s.) the longer pulses both in the low (and high amplitude) regions which occurred just at the beginning of the ionizing phase of the alternating cycle could only be suppressed. This observation would strikingly support the inhibitive action of visible light on pulses. Further the number of shorter pulses initiated by irradiation were of the same properties as those produced in the discriminator bias potential range of 10–20 V (cf. curves 1 and 2 of Fig. 2) and 30–45 V (cf. curves 3 and 4 of Fig. 2). The favourable influence of irradiation on the occurrence of shorter pulses, suggested the occurrence of various secondary processes in the gas phase. It may be argued on similar grounds that of the two processes, viz. γ-mechanism and $\eta\theta g$-mechanism[15], the former viz positive ion bombardment on the outer electrode is primary to the longer pulses; however, the evidence is inadequate to confirm this deduction.

For the occurrence of the effect $\triangle I_s$, Joshi[16–18] suggests that: (1) under discharge, an adsorption like layer consisting of excited particles, ions and electrons, and characterised by a low work function is formed; (2) electrons are emitted under radiation from this boundary layer; (3) negative ion formation due to capture of these photo-electrons by excited neutral particles, especially atoms or/and radicals with large electron affinity, causes a current decrease as a space charge effect; and (4) uncaptured electrons and their secondaries give the positive light effect $+\triangle I_s$. The observed photo-increase ($+\triangle I_s$) of the current I_s and the initiation under light of pulses < 15 V appear to be in disagreement with the postulates (2) and (4). Capture of electrons is possible only when their velocity is low; hence when this last increases, the population of uncaptured electrons increases. From the considerations put forward in the present study, electrons released from the negatively charged (outer) electrode by any mechanism and unhindered by secondary processes especially on account of their large velocity, should lead to the production of longer pulses, but not the shorter pulses as observed in the initial and final amplitude regions of Figs. 2–4.

Harries & Von-Engel[19] had suggested simultaneously that light augments the dissociation of molecules held by Van der Waals' forces on the primary Longmuir-layer on the electrodes, to yield atoms or/and radicals which reduce the secondary emission to give the negative light effect $-\triangle I_s$. The positive effect $+\triangle I_s$ is due to photoionization of pre-excited particles on or/and in the vicinity of the cathode[8]. The suppression of the longer pulses and the creation of shorter pulses under light can possibly follow from this hypothesis. These are, however, a few experimental findings, unconnected with the present investigations, which cannot be explained by this mechanism. The limitations of not only these two suggestions, but of all surface layer theories of the phenomenon are discussed elsewhere[20].

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