PART II

SECTION III

OSCILLOGRAPHIC STUDIES OF THE SILENT ELECTRIC DISCHARGE AND THE JOSHI-EFFECT IN AIR
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INTRODUCTION

While the markedly complex nature of the discharge current in an ozonizer due to the presence of a number of high frequency pulses has been known for a long time\textsuperscript{53}, very few detailed studies using a cathode ray oscillograph have been reported\textsuperscript{6}. Except for the preliminary observations of Joshi\textsuperscript{54} and few observations of other workers, comparative studies of the discharge current in dark and under light are not available in literature. Deb and Ghosh\textsuperscript{55} have published some results from which they have proposed a new mechanism for production of $\Delta i$. However, all these oscillograms (except the three reported in an earlier paper) do not refer to the effect of light at all, and the conditions of pressure and potential regarding these have not been specified by these authors. Especially\textsuperscript{56} in view of the extensive use of the cathode
ray oscillograph in studies of discharge phenomenon and their successful interpretation in recent years, it was of interest to use it in the present studies also. Experiments were therefore undertaken to investigate the nature of the discharge current both in dark and under light over a wide range of conditions. These studies have revealed several types of current variation including the high frequency pulses which are of major importance. These last decrease in amplitude on irradiation and vanish completely under conditions productive of \( 100\% \Delta i \). Amongst other important findings is the permanent suppression of high frequency pulses on irradiation, in the hysteresis range below \( V_m \) leading to the irreversible Joshi Effect, and the decrease in the number of high frequency pulses with a simultaneous increase in their amplitude (cooccurrence) under certain conditions. The wave forms of the discharge current, both when the tube was in dark and under light, were studied with a single beam cenco cathode ray oscillograph No: 71551. The low tension terminal of the discharge tube was earthed through a non-inductive carbon resistance ( 50,000 \( \Omega \) ), and the potential drop
across this was fed to the vertical or Y plates of the oscillograph. The internal time base oscillator of the oscillograph was used for the horizontal or X plates. Since the details regarding the high frequency pulses could not be photographed with the available equipment, visual observations were made in detail with sketches. In these cases, however, the relative magnitudes could not be measured very accurately. Only some of these sketches representing the essential features and the gradual variation with the applied potential at different pressures are shown for the Siemens all glass ozonizer only. The differences between these and the other discharge tubes used are indicated, though separate sketches are not reported here, except figure 54 for the semi-ozonizer.

**SUMMARY OF EXPERIMENTAL RESULTS**

**THE THRESHOLD POTENTIAL \( V_m \)**

The amplitude of the smooth sinusoidal wave trace increases with the potential applied to the system till at a critical value of the latter, high frequency pulses appear at the peaks of the current wave. This is accompanied by the sudden rise in the current measured
by the current detector and the appearance of a glow in the system. This critical potential has been called the threshold potential $V_m$. Measurements below $V_m$, for various gas pressures showed that the increase in the amplitude of the wave trace was linearly related to the applied potential. A careful scrutiny in this region showed that the wave pattern differs slightly from the sine wave, the distortion being due to the higher harmonics (chiefly the third).

**WAVE FORMS OF THE DISCHARGE CURRENT**

The types of current variation may be briefly summarized as follows:-

(a) The current varies smoothly for potentials below $V_m$, both in dark and under light and only in light for potentials where the effect is almost 100%.

(b) For potentials just above $V_m$, the wave form often shows time variations of the relaxation oscillation type, called high frequency pulses. These high frequency pulses are generally observed at the peak position of the smooth sinusoidal trace and extend beyond the quarter wave position.
(c) In a few cases the current increases suddenly and continues at this large magnitude for a comparatively long time giving rise to a hump superposed on the main sinusoidal trace.

The results reported in figures 43 to 56 represent several modes of these types of current variation and their cooccurrence.

**INFLUENCE OF IRRADIATION**

In general, the influence of irradiating the system was to diminish the amplitude and the number of the high-frequency pulses. At low applied potentials especially for low pressures, the high-frequency pulses are suppressed completely. As the potential increases the reduction of high-frequency pulses becomes less, only a part of them being suppressed completely. It is seen that the high frequencies corresponding to the later part of the half cycle are suppressed more easily. Under certain conditions, new pulses appear or unsteady pulses are rendered steady on irradiation leading to a positive Joshi Effect\(^{(456)}\).

All these changes were instantaneous, no time lag being observed between the act of irradiation and the suppression of the pulses.
INFLUENCE OF APPLIED POTENTIAL AND PRESSURE OF THE GAS

Figures 48 to 56 show the changes in waveform with increasing potential above $V_m$ for pressures from 1/8 to 58 mm. Hg. For other pressures, only a few selected waveforms are shown. It is seen that the amplitude of the wave (taken here as the sine wave together with the hump superposed on it) increases continuously with the applied potential, as does the number of the high frequency pulses. The amplitude of the high frequency pulses, however, increases at first up to a maximum and then diminishes. A decrease in the amplitude of the Trichel pulses with increasing current is also reported by Weissler and Mohr. As the pressure of the gas is increased the amplitude of the wave increases whereas the amplitude and the number of high frequency pulse appears to diminish.

WAVEFORMS USING OTHER DISCHARGE TUBES.

The types of current variation in waveforms of the discharge current using a semi ozonizer are essentially similar to those for an all glass ozonizer. However, there is a marked asymmetry in the positive and negative halves of the A.C. cycle. While the negative half exhibits discreet pulses throughout the pressure range investigated, the
**FIG 48**

WAVEFORMS OF DISCHARGE CURRENT IN AIR.
(ALL GLASS OXONIZER)

\[ p = 1 \text{ mm Hg.} \]

<table>
<thead>
<tr>
<th>IN DARK</th>
<th>VOLTS (P. A. M.)</th>
<th>UNDER IRRADIATION</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>26.0</td>
<td></td>
</tr>
<tr>
<td>IN DARK</td>
<td>40.0</td>
<td></td>
</tr>
<tr>
<td>IN DARK</td>
<td>50.0</td>
<td></td>
</tr>
</tbody>
</table>

OZONIZER NO. 1

Figure - 48
FIG. 49
WAVEFORMS OF DISCHARGE CURRENT IN AIR
(ALL GLASS OZONIZER)
\[ p = 6 \text{ mm Hg} \]

VOLTS PRI. YMS.

<table>
<thead>
<tr>
<th>IN DARK</th>
<th>UNDER LIGHT</th>
</tr>
</thead>
<tbody>
<tr>
<td>44.0</td>
<td></td>
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<tr>
<td>71.0</td>
<td>11.0</td>
</tr>
<tr>
<td>39.0</td>
<td>6.0</td>
</tr>
</tbody>
</table>

OZONIZER No. 1

OZONIZER No. 2

Figure - 49
FIG. 50
WAVEFORMS OF DISCHARGE CURRENT IN AIR
(ALL GLASS OZONIZER)

\[ b = 9 \text{ mm Hg.} \]

**Volts per yms**

<table>
<thead>
<tr>
<th>Voltage</th>
<th>Under Light</th>
</tr>
</thead>
<tbody>
<tr>
<td>52.0</td>
<td></td>
</tr>
<tr>
<td>80.0</td>
<td></td>
</tr>
<tr>
<td>45.0</td>
<td></td>
</tr>
<tr>
<td>70.0</td>
<td></td>
</tr>
</tbody>
</table>

**Ozonizer No. 1**

**Ozonizer No. 2**

Figure - 50
FIG. 51.
WAVEFORMS OF DISCHARGE CURRENT IN AIR
(ALL GLASS OZONIZER)
\[ p = 12 \text{ mm Hg} \]

**VOLTS PER MILS**

<table>
<thead>
<tr>
<th>IN DARK</th>
<th>UNDER IRRADIATION</th>
</tr>
</thead>
<tbody>
<tr>
<td>60.0</td>
<td></td>
</tr>
<tr>
<td>70.0</td>
<td></td>
</tr>
<tr>
<td>80.0</td>
<td></td>
</tr>
</tbody>
</table>

**OZONIZER No. 1**

Figure 51
WAVEFORMS OF DISCHARGE CURRENT IN AIR
SLEEVE EXCITATION
$P = 18 \text{ mm Hg}$

VOLTS PER TUBE

<table>
<thead>
<tr>
<th>IN DARK</th>
<th>IN LIGHT</th>
</tr>
</thead>
<tbody>
<tr>
<td>4.0</td>
<td>4.0</td>
</tr>
<tr>
<td>4.0</td>
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<tr>
<td>5.0</td>
<td>7.0</td>
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<td>7.0</td>
<td>7.0</td>
</tr>
<tr>
<td>7.0</td>
<td>7.0</td>
</tr>
</tbody>
</table>

TABLE 7

TUBE No
$d = 8.7 \text{ cm}$

Figure - 52
Figure 5.3

Waveforms of Discharge Current in Air
(All Glass Ozonizer)

P = 27 MM Hg.

Volts per 10 μsec.

In Dark

67.0

In Light

75.0

In Dark

100.0

In Light

105.0

Ozonizer No. 3

Figure - 53
FIG 54

WAVEFORMS OF DISCHARGE CURRENT IN AIR
(SEMI-OZONIZER)

p = 32 mm Hg.

IN DARK IN LIGHT

0.8 kV

IN DARK AND LIGHT

1.0 kV 1.2

2.0 kV 3.0

Figure - 54
FIG. 55.
WAVEFORMS of DISCHARGE CURRENT IN AIR
(ALL GLASS OZONIZER)
\( p = 40 \text{ mm Hg} \)
VOLTS PER TM S

IN DARK V 26.0

OZONIZER NO. 1

IN DARK 21.0

OZONIZER NO. 2

IN DARK 31.0

OZONIZER NO. 4

IN LIGHT

Figure - 55
FIG. 56.
WAVEFORMS OF DISCHARGE CURRENT IN AIR
(ALL GLASS OZONIZER)
\( p = 56 \text{ mm Hg} \)

IN DARK  
\( \text{VOLTS PRI. X 1000} \)  
\( 27.0 \)  

\( 30.0 \)

OZONIZER No. 4

IN DARK  
\( 8 \)  

IN LIGHT

IN LIGHT

OZONIZER No. 1

Figure - 56
waveform in the positive half consists of one or two h.f. pulses followed by a smooth current variation. For higher potentials new pulses begin to appear at the peak position. Unlike the pulses observed with the ozonizer, these pulses almost always extend beyond the main wave trace. It is of interest to mention here that for the low pressure range reported here the pulses appear in both the halves at the same applied potential. Starting potentials for pulses from positive and negative points in air are equal and are explained by Loeb.\textsuperscript{58}

The wave forms of the discharge current using a tube excited by external sleeves are far less complicated in that the amplitude of the sine wave and the number of h.f. pulses are much smaller. Due to this simplicity it was observed under certain conditions that the number of h.f. pulses is reduced while their amplitude is increased slightly (cooccurrence).

It is of interest to mention here that the h.f. pulses referred to above (and observed with the available oscillograph) are not regular frequencies. Using much higher time base frequencies, the pulses show two distinct parts. Initially the current increases
very fast so that the rise is practically instantaneous and then the trace on the oscillograph is almost vertical even with the highest sweep frequency. After reaching the peak value corresponding to the pulse amplitude, the current falls rapidly at first and then more slowly. The entire pulse trace lies on one side of the base and the frequency as computed from their intercept in the time base \(3 \times 10^{-5} \text{sec.}\) lies in the range 2.5 to 30 kc/sec.

Radio frequencies in the maga cycle range referred to by Warbarg and others and the still higher plasma oscillations of Tonks and Langmuir were not observed in the present case probably because of the limitations of the available instruments.
DISCUSSION

During his preliminary studies Joshi observed the sensibly instantaneous and reversible diminution of the amplitude of these h.f. pulses on irradiating the discharge tube. However, in this theory of the ΔI phenomenon, no mechanism for the production of these pulses or the suppression of these alone under light is suggested. Of the various other workers, Deb and Ghosh, at the suggestion of Mitra, were the first to propose a probable mechanism for their production, based essentially on that of Klemene, Hinterberger and Hoffer. This is discussed below together with similar mechanisms proposed by Harris and von Engel, Khastgir and Coworkers, Ramaiah and others.

From an oscillographic study of the ozonizer discharge Klemene, Hinterberger and Hoffer showed that the high frequencies referred to by Warburg were only unidirectional pulses. As an extension of the original picture of Warburg these authors suggested the following mechanism for the ozonizer discharge and the production of h.f. pulses. When the potential during the A.C. cycle becomes large enough, ionization of the gas produces ions and electrons which move under the action of the applied field.
to the respective electrodes and deposit on them as surface charges. These surface charges give rise to a field opposite in direction to the one externally applied, so that the resultant field diminishes. When the opposing field due to the surface charges is large enough and the discharge stops. Now during the course of the A.C. Cycle, the externally applied potential becomes zero and changes sign. At this instant the surface charges are free to neutralize under the action of their own field. Being formed on an insulating glass surface the neutralization takes place in discreet small sparks which constitute the observed h.f. pulses.

Assuming essentially the above mechanism, Deb and Ghosh 55 have proposed the following hypothesis for the Joshi Effect. In addition to the main sinusoidal trace due to the frequency input to the system, these authors refer to three different components (i) groups of current pulses in which individual pulses are separately observable at or near the peaks of the current wave, are called the low frequency pulses (ii) groups in which the pulses are densely packed and jump into one another observable on and around (ii), are called the high frequency pulses and (iii) there is a set of highly damped oscillations. While the origin of high frequency pulses is assumed to be the same as that
proposed by Klemenc et al, the production of low frequency pulses is explained as follows: As above, the ions and electrons produced under discharge give rise to an opposite field which stops the discharge. However, during the increasing part of the potential cycle, the breakdown voltage is again reached in spite of the opposing field of the charges, causing further ionization. This increases the number of ions and electrons and hence the opposing field so that the discharge stops again. This starting and stopping of the discharge gives rise to the low frequency pulses. It may be observed that the electrons due to their higher mobility go and deposit on the electrode forming a surface charge, while the positive ions mostly remain in the space due to their smaller mobility and form a space charge. As a modification of the mechanism of Klemenc et al, these authors emphasize the predominant role of electronic surface charge in controlling the ozonizer discharge. Joshi Effect is explained by assuming electron emission from the surface charge under light. This decreases the density of the surface charge and consequently a decrease in the density of the high frequency pulses and the total amount flowing through the ozonizer.
The discharge in the study of the Joshi Effect is necessary between insulating electrodes from which secondary electron emission is known to be negligible. When the applied potential during the A.C. cycle is large enough, a stray electron causes an initial Townsend avalanche but its succession is not likely to be maintained. No mechanism for this has been specified. The alternative that the ionization produced in one single avalanche may give enough surface and space charges to neutralize the applied field and stop the discharge does not appear to be likely. An electron product is approximately $2.5 \times 10^7$ ion pairs in atmospheric air at the conventional sparking potential. The estimates of Kip and Trichel for brush pulses in high pressure corona are also of the same order and this does not appear to give a high opposing field to stop the discharge.

It is further tacitly assumed that when the applied potential changes sign, the surface and space charges neutralize in isolated sparks giving rise to high frequency pulses. Such a neutralization may be possible only if the field due to the surface charges is high enough to cause breakdown. This would commence
approximately when the applied potential is nearly twice the threshold potential and till then high frequency pulses cannot appear. Moreover, such neutralization sparks are expected to give rise to oscillations and not pulses. While the authors assume that damped oscillations (depending on the circuit constant L.C., and R) are produced by the recombination of polarized space charge, no reason is given why the recombination of surface charge gives rise to pulses and not oscillations.

It is further evident that the high frequency pulses and the low frequency pulses refered to by Deb and Ghosh should appear at different positions of the current wave. This is so because the neutralization producing the high frequency pulses takes place when the applied potential is zero i.e. at the current peak, while the starting and stopping of the discharge leading to low frequency pulses occur between $V_m$ and the peak voltage i.e., approximately at zero current. However, the high frequency pulses "appear on and above the low frequency pulses". It is of interest to mention that essentially similar pulses are reported by CraggS and Meek recently using metal electrodes. It is not unlikely that the mechanism for
the production of the pulses in the two cases viz., with metal and insulating electrodes may be the same. It may be added that this distinction between two types of pulses appears purely arbitrary. No estimates, of the frequencies are reported. Such estimates made from the intercept of the pulses on the time base reported here indicate that the pulses registered on the oscillograph correspond to a frequency from $2.5 \times 10^3$ to about $30 \times 10^3$ cycles/sec. depending on other parameters e.g., the potential applied, the pressure of the gas, the inter electrode separation etc. This and the appearance of all the pulses in the same place on the current wave indicate the similarity between the various pulses with no necessity for a distinction of the type envisaged by these authors.

In addition to these limitations in the above mechanism for the production of pulses, the consideration given below go contrary to these hypotheses for the $\Delta i$ phenomenon. They observe ' when the electrode surface is irradiated, there is electron emission from the surface charge and its density is considerably reduced'. Consequently the density of the high frequency pulses is also considerably reduced leading to a reduction in the total current. It is
significant to note that the electronic surface charge deposits on the anode, from which electron emission by but visible light against the high applied field of the order of hundreds of volts, does not appear to be likely. If electron emission is contemplated at the time when the impressed field becomes zero during the course of the cycle, i.e., at the instant when neutralization is assumed to take place, such an emission could only facilitate neutralization. One fails to see how it will cause a current decrease.

In an attempt to explain the influence of other parameters on Joshi Effect, these authors assume that (i) The adsorbed layer of gas molecules is quickly and more completely formed if the electron affinity of the gas molecules is high and hence the proportion of high frequency component and $\% \Delta i$ would be greater and (ii) with an increase in the frequency of the A.C. supply "the pulsation time becomes equal to and finally less than the time required for the full deposition of the charge. Increase of frequency therefore causes a decrease in the density of the surface charge ". It is reasonable to assume that the higher electron affinity of the gas leading to a more complete formation of the surface charge, would prevent
easy detachment of electrons by light, thus presumably leading to a smaller current decrease, $-\Delta i$, on irradiation. Besides, even a rough calculation indicates that the time required for the full deposition of the surface charge (Ca $10^{-7}$ sec. for a 1 cm gap at atm. pressure) is not longer than the pulsation time when 'f' is changed from 50 to 500 c/s. Finally it may be added that 'the small isolated sparks in which small elements of surface charge recombine' should give rise to a recombination spectrum which has not been observed.

The mechanism proposed here for the production of high frequency pulses is similar to that investigated in recent years by Loeb and his collaborators from detailed studies on corona and spark discharges. These studies indicate that the pulse is to be ascribed to the sudden onset of ionization by initiating electrons from the cathode, which gives the fast rise in current. However, due to certain limitations, the current is not maintained at the high value and diminishes gradually. These limitations may arise from space charge distortion of the field by positive ions as in the burst pulse corona, or by negative ions as in
the Trichel pulse corona\textsuperscript{68}, or from the lowering of the effective field by the circuital resistance as in the case of the non-self quenching counters\textsuperscript{69}. (It may be mentioned that even in this last case the positive ion sheath actually quenches the discharge, the role of the serial high resistance being merely to prevent leakage of charge and thus keep the wire potential below $V_g$ while the positive ions are collected on the cathode\textsuperscript{69}). In all these cases a limitation on secondary emission from the cathode is not possible. However, the discharge would also be quenched if the emission of secondary electrons from the cathode is limited as in the case of the ozonizer. It is suggested here that in this case, before such high space charges as to quench the discharge completely could build up, the supply of electrons from a small region of the cathode is exhausted and the current due to the discharge of that region begins to diminish. The results of a number of workers on the number of electrons per avalanche indicate that it is too small to detect with the limited sensitivity of the oscillograph. It is therefore reasonable to assume that the pulse observed on the oscillograph corresponds to the discharge composed of a number of electron avalanches.
The observation of certain bright spots on the electrode at higher pressures further suggests that the discharge takes place from certain discreet patches which take up the discharge in preference to other parts of the electrodes probably because of a low energy of detachment necessary for electrons and a high local field, and which in these respects correspond closely with the so-called active spots. It is not unlikely that the flashes observed by Klemenc et al. and called by them as also by others, 'the neutralization sparks' were discharges of the above type. This is in accord with the recent results of Oh and Duke who observe changes in the light intensity corresponding to changes in the voltage and current.

On the above view, the sinusoidal wave trace observed below $V_m$ corresponds to capacitive current. When the applied potential reaches the threshold value, a stray electron initiates the discharge. The electrons (and ions) created deposit on the respective electrodes and are not neutralized due to the dielectric nature of glass. These constitute the main source of electron supply for the succeeding half cycle during which an electron from the cathode initiates a Townsend avalanche. The other electrons
situated in the vicinity of this initiating electron and forming a patch, feed into the initial Townsend avalanche, giving rise to a pulse. Before the space charge due to the positive ions could develop to a value so as to choke off this discharge, however, the limited supply of electrons from the patch is exhausted and the current begins to diminish. Thus the fast rising part of the current is due to the collection of electrons created in the pulse while the later part of the pulse corresponds to the collection of positive ions. The known values of electron and positive ion mobilities under these conditions indicate that the time taken by the avalanche to overcome a 1 cm. gap is approximately $10^{-7}$ sec., while that for the positive ions is approximately $10^{-4}$ to $10^{-5}$ sec. The observed values of the intercept of a pulse on the time base are in good agreement with this. The discharge is now taken up by another patch of surface electrons and the whole process repeats giving rise to another pulse. This continues till the peak value of the potential is reached beyond which no discharge is possible.

It must, however, be mentioned that no experiments could be made to investigate the actual mechanism for the detachment of these pre-deposited surface electrons
from the electrode. The liberation of electrons from one patch which feed into the initial Townsend avalanche and develop into a pulse, takes place in a very short time which suggests that the detachment may be due to the action of photons produced in the avalanche. At the same time, the discharge of the second patch after the arrival of positive ions at the cathode suggests that the detachment may be due to the action of positive ions at the cathode. It is likely that both these mechanisms are at work (cf. Part I). This assumption incidentally provides a better definition of the patches assumed above. Thus, while the deposition of electrons on the surface is not likely to be in patches, all these electrons which are liberated by the photons from a limited area on the cathode so that they can feed into one single pulse constitute a patch. Moreover, since the electrons for the discharge corresponding to the second pulse are detached by the action of positive ions, it explains why beyond the peak value of potential, no discharge is possible.

According to the new mechanism proposed for the Joshi Effect, light detaches some of the predeposited surface electrons in the non-ionizing part of the potential cycle. This diminishes the number of initiating electrons
from a patch and hence the pulse amplitude as observed. Under favourable conditions all the electrons corresponding to a patch may be detached by light. In such cases the amplitude of the pulse diminishes to zero, in other words, the pulse disappears completely. It is of interest to note that when the discharge tube is in dark, most of the surface electrons are liberated by one of the secondary mechanisms at the cathode after the first Townsend avalanche i.e., in the ionizing part of the potential cycle, while on irradiation, some of these may be detached in the non-ionizing part of the cycle. Under conditions when the space charges play a prominent role, these electrons in the non-ionizing part go to neutralize it, in effect, which diminishes the effective field \( X + X_1 \), which is the vector resultant of field \( X \) and the space charge field \( X_1 \). It is significant to note that once the space charge field is diminished, it continues to be so throughout the half cycle due to the limited supply of electrons in this case which is further reduced due to photodenudation. This limitation being absent in metal electrodes, the non-occurrence of Joshi Effect in these systems is to be anticipated.