CHAPTER 3

OZONE GENERATION WITH CONVENTIONAL PULSE CONTROL METHOD

3.1 INTRODUCTION

The most common method of ozone generation is to produce an AC corona discharge in a gap bounded by metallic electrodes and containing at least one solid dielectric barrier.

One of the requirements of any ozone generator is a facility for varying the ozone concentration in the gas. To fully understand how this may be achieved in practice, it is necessary to consider, in some detail, how the ozone generator operates and the various parameters which affect the ozone generation rate (ID Chalmers et al 1998). Consider a simple parallel plate arrangement as shown in Figure 3.1 and let the dimensions of the gas gap and the dielectric layer be $d_g$ and $d_d$ and their relative permittivity be 1 and $\varepsilon$ respectively.

![Figure 3.1 Schematic Arrangement of a Corona Discharge](image-url)
If the voltage applied to the gas/dielectric combination is $V$ then the electric fields in the gas gap and dielectric layer $E_g$ and $E_d$ are $V/k_g$ and $V/k_d$ respectively, where

$$k_g = [d_g + (d_d / \varepsilon)] \tag{3.1}$$

$$k_d = (d_d + \varepsilon d_g) \tag{3.2}$$

To indicate practical values for $k_g$ and $k_d$, consider a typical ozone generator in which the gas gap and dielectric layer are both around $1.5 \text{ mm}$ and the dielectric material, often made of glass, may have a relative permittivity of $3$. Thus $k_g$ is around $2 \text{ mm}$ and $k_d$ around $6 \text{ mm}$ giving, for an applied voltage of say $5 \text{ kV}$, an electric field of $2.5 \text{ kV mm}^{-1}$ in the gas gap and $0.85 \text{ kV mm}^{-1}$ in the dielectric (Shimomura N et al 2002).

If the charge density on the dielectric surface is $\rho$ ($\text{cm}^{-2}$), then the electrostatic flux density $D$ in both the gas gap and the dielectric is similarly $\rho$ ($\text{cm}^{-2}$) and the resultant electric fields in the gas gap and dielectric due to this accumulation of charge are $\rho / \varepsilon_0$ and $\rho / \varepsilon$ respectively. The gas gap field, as explained above, opposes the applied field giving a resultant gas gap field $E_g$ of $(V / k_g - \rho / \varepsilon_0)$. Therefore the criterion for stability (discharge extinction) is

$$(V / k_g - \rho / \varepsilon_0) = E_i \tag{3.3}$$

thus

$$\rho = \varepsilon_0 (V / k_g - E_i) \tag{3.4}$$

Ozone is generated in an electrical discharge first by dissociation of $\text{O}_2$ to form atomic oxygen (O) and then later three-body collision recombination of atomic and molecular oxygen to form ozone. The
dissociation stage is caused by collisions of energetic electrons with molecular oxygen and thus it is reasonable to assume that the number of ozone molecules produced in any time interval will be directly related to the number of electrons generated in the discharge during that time interval. The number of electrons, in turn, will be reflected as the total charge $Q$ collected by the dielectric. Thus the number of ozone molecules produced $N_o$ will be

$$N_o = KQ$$

(3.5)

where $K$ is an empirical constant.

The surface charge density on the dielectric is

$$\rho = \frac{Q}{A}$$

(3.6)

where $A$ is the total area of the dielectric surface.

Thus combining Equations (3.4), (3.5) and (3.6) we have

$$N_o = K A \varepsilon_0 (V / k g - E_i)$$

$$= K' A (V / k g - E_i)$$

(3.7)

Considering now what occurs when the applied voltage is in the form of an alternating sinusoid, the same argument holds and ozone will be generated while the voltage is increasing. Again the field in the gas gap will be limited to the breakdown field and discharging will cease when the voltage ceases to increase (i.e. at voltage peak). Then the number of ozone molecules generated between the start of the discharge and the attainment of voltage peak ($V_{\text{max}}$) will again be derived from Equation (3.7) as

$$N_o = K' A (V_{\text{max}} / k g - E_i)$$

(3.8)
When the applied voltage then starts to decrease, the charge on the dielectric surface produced by the earlier discharges persists and the field in the gas gap is reduced at all stages by this constant field which can be derived from Equation (3.3) as \( \frac{V_{\text{max}}}{k_g} - E_i \). Thus the field in the gas gap \( E_g \) for any instantaneous voltage \( V \) will be given by

\[
E_g = \frac{V}{k_g} - \left( \frac{V_{\text{max}}}{k_g} - E_i \right)
\]  

(3.9)

It can be seen from Equation (3.9) that the field in the gas gap can actually become negative before voltage become zero and, in fact, depending upon the relative magnitudes of \( \frac{V_{\text{max}}}{k_g} \) and \( E_i \), can even achieve the breakdown field, heralding the onset of discharges in the opposite direction, before voltage become zero. When discharging commences in the opposite direction, the charge on the dielectric surface is rapidly neutralized and replaced with charge of the opposite sign and the process described above is repeated.

It can be seen then that the number of ozone molecules produced each half cycle is given by Equation (3.8) and thus the generation rate per second is

\[
fK'A \left( \frac{V_{\text{max}}}{k_g} - E_i \right)
\]

Replacing the empirical constant \( K' \) by another arbitrary empirical constant \( G \) we can then say that the generation rate \( R \) of ozone in units of gm/hr (which is the normally adopted unit) can be expressed as

\[
R = GfA \left( \frac{V_{\text{max}}}{k_g} - E_i \right)
\]

and substituting for \( k_g \) in terms of the characteristics of the generator, we then have
\[ R = G f A \left\{ \frac{V_{\text{max}}}{d_g + (d_i/\varepsilon)} \right\} - E_i \]  \hspace{1cm} (3.10)

Equation (3.10) is thus a generalized equation describing how the ozone generation rate varies with variation in determining parameters in all the generator and supply voltage characteristics. It shows, for example, that the generation rate may be varied by variation of the frequency of the supply voltage and this method is indeed adopted in some medium- and high-frequency generators. When the supply frequency is constrained to power frequency however, the method of control is to vary the applied voltage. What is very clear from the foregoing derivation of Equation (3.10), is that the actual waveform of the supply voltage is quite unimportant. The gas gap is usually around 3 millimeters and the feed gas, air or oxygen, is passed longitudinally along the gap (Namihira et al 2001). A typical ozone generator, in which the gas gap and dielectric layer are both around 1.5 mm and the dielectric material, often made of glass, and may have a relative permittivity of 3.

3.1.1 Pulse Circuit

The pulse circuit shown in Figure 3.2 provides high voltage (5 kV to 10 kV) using ignition coil of 12V DC and 6 A. A 2N3055 power transistor is pulsed with a square wave signal generated from 555 IC. The frequency of the pulses depend on the resistors between pins 7 and 8 and between pins 7 and 6. The pulse can also be varied with a capacitor. A 50% of duty cycle is selected for operation with 555 IC timer. This can be establish using the following mathematical equations in section 3.1.2. A single cell ozonator is shown in Figure 3.3.
3.1.2 Mathematical Equation to Obtain 50% Duty cycle

\[ T_1 - \text{Time On} = 0.693(R_1 + R_2) C_1 \]
\[ T_2 - \text{Time Off} = 0.693 R_2 C_1 \]
\[ T - \text{Total time period} = 0.693(R_1 + 2R_2) C_1 \]
\[ f - \text{Frequency} = \frac{1.44}{(R_1 + 2R_2) C_1} \]

Duty cycle % = \frac{R_1 + R_2}{R_1 + 2R_2}

Figure 3.2 Control Circuit for a Single Cell Ozonator

Figure 3.3 Single Cell Ozonator
3.2 METHODS AND MATERIALS

Dust-free air enters an air-compressor (compression pressure 1 atm) and is sent to the air-drying system. A two-column, cyclic regenerated adsorptive dryer with activated aluminum oxide is part of this drying system. A pressure regulator to control input pressure to ozone generator is connected at output of air dryer system. Dew point temperature is about 213K (-60°C). For producing high concentration ozone, industrial oxygen is used as the feed gas. A classical dielectric single pair tubular ozone cell generator (Dielectrics borosilicate glass thickness 1.85 ± 0.1 mm; length 20 cm; gap 3.0 mm; residence time at nominal regime 4.8s) with electronic pulse control circuit to produce 5kV is used. The ozone generator was fed with industrial grade oxygen/clean dry air. A five liter glass column was used for experiments and connected to the ozone generator as shown in Figure 3.4. The spent gas was sent through ozone destruct unit before releasing it to atmosphere.

Figure 3.4 Experimental Setup for Ozone Concentration Measurement
The effect of frequency on ozone concentration is shown in Figure 3.6. The tests are conducted at 5 kV constant voltages. With increase in frequency, the concentration rate has increased. Although higher frequency is desirable for conventional high capacity generator, the operating frequency is limited by the heating of electrodes and control circuit chokes.

Figure 3.5 Ozone Generation Setup

Figure 3.6 Ozone Concentration Vs Frequency
From the study of the effect of changing the switching frequency, one can evaluate the process of ozone gas concentration. The experimental results in Figure 3.6 demonstrate the relationship between the frequency and the quantity of ozone concentration. With the increase of the switching frequency, increased concentration of ozone gas are generated because the shifting of the frequency level in the converter circuit has an effect on the production resonance at the ozone tube. These advantages considerably facilitate the design of an appropriate HF power supply for DBD applications. Thus, the model allows one to predict with accuracy the discharge behavior using the experimental parameters and also verify the condition.

The ozone generation as a function of gas flow rate is plotted in Figure 3.7. The gas flow rate were adjusted using a gas regulator (0-50 lpm). With increased flow rate of feeding gas the ozone concentration is decreased as residence time in the tube is decrease. The composition of feed gas decides the ozone generation concentration.

![Figure 3.7 Ozone Concentration Vs Flow Rate](image-url)
The efficiency of ozone with oxygen as feed gas is approximately 3 times larger than that with dry air, in which the oxygen content is 20% in dry air (Fernando et al 2011). The ambient air resulted in a more decreased efficiency probably due to its humidity. The production of ozone at flow rates of 10 LPM is shown in Figure 3.8.

![Figure 3.8 Ozone Production Vs Gas Flow Rate](image-url)