Chapter 2

Dielectric Barrier Discharge

2.1 Atmospheric Pressure Discharge

In the past, application of atmospheric pressure plasma is limited to the use of high pressure, high temperature and thermal gas discharge. Thermal plasma, usually the arc discharge is associated with Joule heating, thermal ionization of the gas, high particles energy and high power at atmospheric operating pressure. Application of thermal plasma is somehow limited given the amount of heat generated and problems related to heat dissipation method. Some examples of the application of thermal plasma are plasma torch and furnaces which involve thermal incineration process (Pfender, 1988).

Recently, with the development of the non-thermal atmospheric pressure plasma, focused attention from both academic and commercial sectors was received. The essential feature of this plasma is its ability to remain low in temperature even after long period of operation, whereby the majority of the electrical energy is converted into production of high energy electrons without heating the entire gas stream (Bogaerts *et al.*, 2002).

Traditionally, non-thermal plasma can only be produced in low pressure environment. This is done by lowering the operation pressure to near vacuum to limit the number density of electrons and ions associated with the electrical discharge. Diffuse plasma can be obtained in this way but the need of expensive vacuum facility is somehow discouraging at industrial level.

Presently, with the capability of generation of non-thermal plasma at atmospheric pressure, acceptance from various fields was gained rapidly. The property of non-thermal plasma working with atmospheric pressure is highly favoured by the industry (Mizuno, 2007) as the operation of such plasma is cost effective and cost friendly with the absence of expensive vacuum system and vacuum compatibility issues. Although the discharge tends to turn into filaments at atmospheric pressure (Massines *et al.*, 2005), which is a down turning point for stable operation and for control over the plasma discharge, atmospheric pressure operation is still a great advantage over the traditional way.

With the developing technology of atmospheric pressure plasma, non-thermal plasma has been used in applications such as complex microelectronic circuit etching (Laroussi *et al.,* 2002c), deposition of various types of coatings on different material (Wagner *et al*., 2003 and Topala *et al*., 2009), on-line surface treatment with open air atmospheric pressure plasma chamber (Moon *et al.*, 2004), water treatment and decontamination (Laroussi, 1996), sterilization (Laroussi, 2005a), chemical synthesis and plasma display panel (Kogelschatz, 2003).

The high energy electrons produced in non-thermal plasma through collision and scattering regularly produce excited species, free radicals, and ions (Wagner *et al*., 2003). These are the key players for plasma chemistry applications such as pollution control, exhaust gas control, removal of volatile compounds and water purification (Xu, 2001). The active species oxidize and decompose the pollutant molecules rapidly in vigorous way (Gallagher *et al.*, 2004).

Various geometry configurations have been proposed and studied in the effort to maintain the stability of non-thermal plasma at atmospheric pressure by the many researchers. The dielectric barrier discharge, corona discharge, sub-microsecond pulse

excitation plasma, One Atmosphere Uniform Glow Discharge Plasma (OAUGDP) (Montie *et al.*, 2000), resistive barrier discharge (Laroussi *et al.*, 2003), and hybrid mode discharge are some examples of the popular configuration.

In the present work, the atmospheric pressure DBD configuration is used. It is basically a capacitive coupled discharge in which the high voltage is applied to one of the parallel plate electrodes separated by glass or alumina barrier with a small air gap between them.

2.2 Dielectric Barrier Discharge

Dielectric barrier discharges (DBD) is configurationally simple relative to other systems, making it a popular system for study. DBD are usually operated at atmospheric pressure, commonly consisting of two planar electrodes with at least one of them being covered by a layer of dielectric material (Figure 2.1). Establishment of the DBD discharge is typically non-thermal plasma (Tyata *et al.*, 2009), or non-equilibrium plasma as the electrons gain sufficiently high energy from the electrical discharge while the heavier ions and molecules remain at low energy level (Kunhardt, 2000). The temperature of the heavy particles in non-equilibrium plasma is within room temperature while the electron temperature can reach about 10^4-10^5 K with mean energy of 1 eV (Kogelschatz, 2000b).

Active species such as electrons, ions, metastables, and free radicals generated from DBD are suitable media for applications in gas cleaning (NO_x, SO_x) (Chen and Mathur, 2002), ozone generation, UV sources, thin film deposition, and surface energy activation for enhanced wet-ability (e.g. contact lens) and adhesion (e.g. automotive

painting) (Borcia *et al*., 2003). Recently, the ability of the DBD for applications in biology and medical field has been demonstrated as well (Laroussi *et al*., 2000).

When high voltage is applied between the planar electrodes, initiation of gas breakdown is induced given the voltage applied is high enough with the electric field exceeding certain value according to Paschen's Law (Lux, 2004). Breakdown of the gas allows the electrons accelerated by electric field to form discharge plasma and create active species. Especially at atmospheric pressure, if no insulation is placed between the electrodes, an arc will be formed. However, with the dielectric layer placed in front of at least one electrode for insulation, it will limit the filament current from building into forming an arc discharge (Gibalov and Pietsch, 2000).

Each current filament that bridges through the gap is known as a microdischarge. As the electric field is applied over time, charges accumulated on the surface of the dielectric will form an opposing local field. Hence, the effective field between the electrodes will be reduced and the filament is quenched eventually (Xu and Kushner, 1998). This is an important property of the dielectric insulation, and also the key feature for DBD to produce non-thermal plasma. The lifetime of a microdischarge is typically in range of tens of nanosecond, with peak current of 0.1A (refer to Table 2.1). The filament diameter is of the order of 100μm (Kogelschatz, 2000b). In order to sustain the discharge, alternating voltage has to be supplied to the electrodes, allowing the surface charges to be removed.

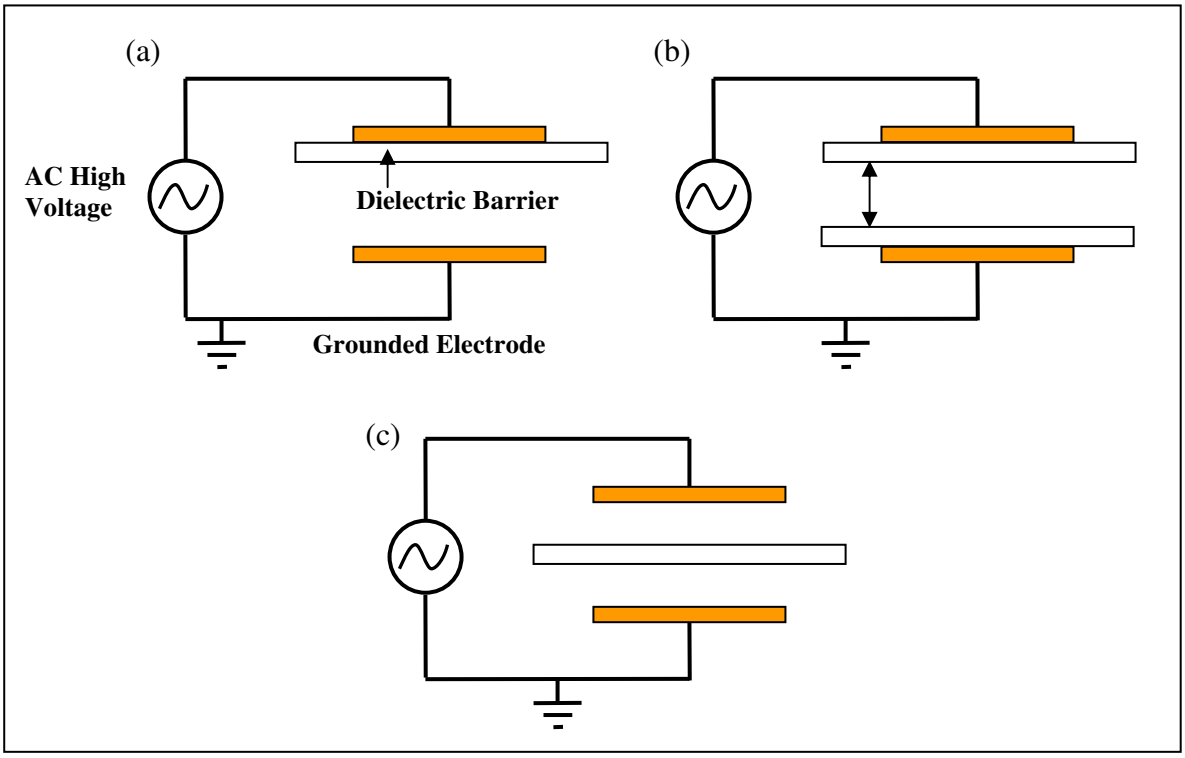


Figure 2.1 : Various parallel plate electrodes configuration of DBD with the dielectric barrier layer(s) at different positions as shown.

Table 2.1 : Typical parameters for microdischarge in a 1mm gap, atmospheric pressure air. (Kogelschatz, 2000b)

Lifetime	1-20 ns	**Filament radius**	50-100 µm
Peak current	0.1 A	**Current density**	0.1 – 1 $kAcm^{-2}$
Electron density	10^{14} - 10^{15} cm^{-3}	**Electron energy**	1 – 10 eV
Total transported charge	0.1 – 1 nC	**Reduced electric field**	$E/n = (1\text{-}2)(E/n)_{Paschen}$
Total dissipated energy	5 µJ	**Gas temperature**	~300 K average

2.2.1 Electrical Breakdown of Gases

Breakdown of an air gap due to electric field will occur only if the potential difference applied between the electrodes exceeds certain critical value. This critical value is called the breakdown voltage, and a discharge can be ignited beyond this value.

It is also the minimum voltage necessary to be supplied to the electrodes for the discharge to be sustainable. This breakdown phenomenon is well governed by the "Townsend Breakdown" mechanism.

When electric field is applied to the electrodes, stray charges in the air gap between the electrodes are accelerated rapidly as electrons drift toward the anode. As the electron is traversing the gap, it gains sufficient energy to free a second electron from the neutral atom through inelastic collision. More free electrons are created in cascading ionization (Figure 2.2), reaching the anode as a current. This electron cascading action is called the Townsend avalanche (Veldhuizen, 2000).

The free electron generation rate of the avalanche is expressed by the first Townsend ionization coefficient, α, and it is the rate of free electron production by an initial electron as this electron travels per unit distance.

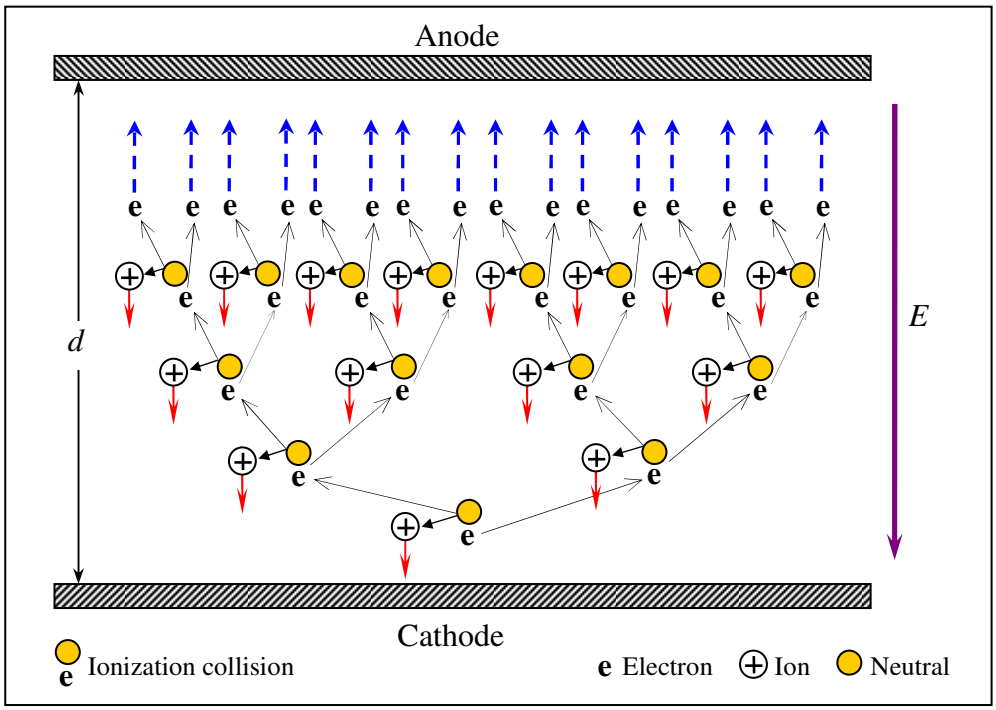


Figure 2.2 : Townsend Breakdown Mechanism.

Typically in a discharge with applied voltage higher than the breakdown voltage, the creation of secondary electrons from the cathode (by bombardment of ions) is usually greater than the number of stray electrons that start off the initial avalanche. When the former replaces the ignition electron, the discharge becomes self-sustainable by the criteria given as (Suzuki and Ito, 2005):

$$\gamma\left(e^{\alpha d}-1\right)\geq 1. \qquad (2a)$$

γ is the secondary emission coefficient, which is the probability of secondary electron generation from the cathode per impacting ion. This coefficient depends on the cathode surface material, type of gas, and the reduced electric field applied. Typical value for γ coefficient is in the range of 0.01 to 0.1 (Fridman *et al.*, 2005a).

Paschen's Law

The breakdown voltage over different air gap and pressure in different systems is not a simple linear relation. However, it can be generally described by Paschen's Law, which gives the minimal voltage required for gas breakdown within a gap as a function of the product of gap pressure and gap distance, $V = f(pd)$, where p is the pressure and d is the gap distance (Kunhardt, 1980). The breakdown voltage for given gap distance and pressure plotted as a function is known as the Paschen Curve, as shown in Figure 2.3 for various gases.

On the right hand side of the Paschen Curve, higher breakdown voltage is required for fixed gap of higher pressure or fixed pressure while increasing the gap distance. However, there is a minimum value (turning point) of the breakdown voltage over the function of *pd*, whereby, breakdown can be obtained with minimum supply voltage. Typically for discharge in air, and gap of few millimeters, the breakdown voltage is given by

$$V = 30pd + 1.35 \text{ (kV)} \qquad (2b)$$

where d is in centimeters, and p is in atmospheres (Lux, 2004).

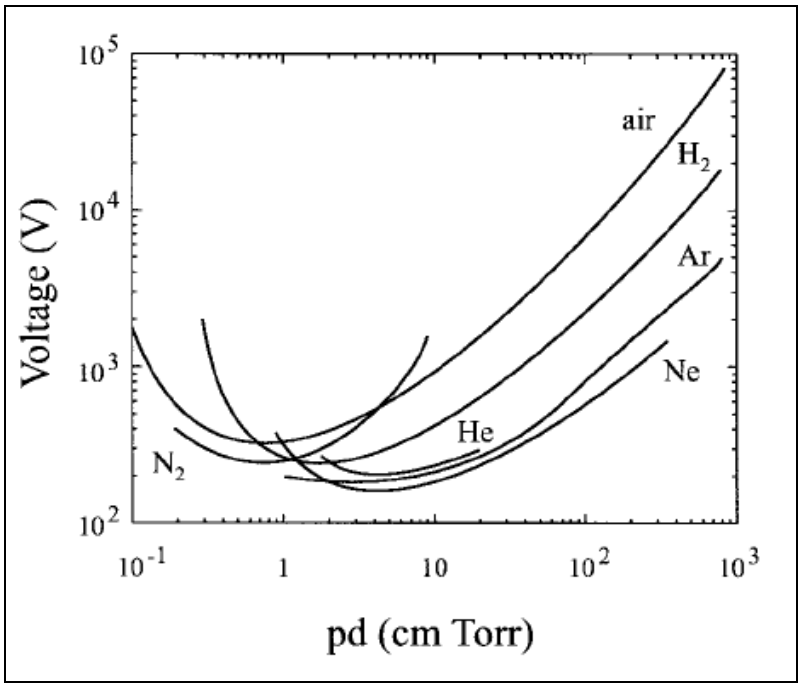


Figure 2.3: Paschen Curve for parallel plate arrangement in various gases (Raizer, 1991. Appeared in Schütze *et al.*, 1998)

2.3 Discharge in Two Different Modes – Glow and Filamentary

2.3.1 Memory Effect in Dielectric Barrier Discharges

There are two types of operation in the non-thermal DBD plasma, namely the glow (diffuse) mode and filamentary mode. The type of plasma obtained depends on the device configuration and operating parameters. However in most operation, only filamentary DBD can be obtained, and it is well known to be due to the "memory effect" phenomenon of the DBD.

Under filamentary operation, microdischarge filaments are distributed over the available discharge gap and are seen as bright filaments (see Figure 2.6(b)). After the microdischarge is established between the electrodes, this ionized channel of the plasma rapidly dissipates the electrons and ions from the gap. Most electrons will be channeled away in 20ns while the heavier ions remain in the discharge gap for several microseconds as their drift velocity is significantly slower than electrons (Chirokov *et al.*, 2005a).

With a dielectric layer inserted between the electrodes, the charges dissipated from the microdischarge will be deposited on the surface of the dielectric in-between two discharge cycles. The deposition of the charges creates a local electric field that opposes the applied electric field, causing the collapse of the local field at the microdischarge location (Chirokov *et al.*, 2005a). These residue charges deposited on the dielectric surface last for few tens of microsecond, or until the polarity of the applied voltage is reversed. After the voltage polarity reverses, the charges deposited earlier on, aid the formation of the microdischarge at the same spot / location as it takes less external energy to swing the local electric field to reach electrical breakdown (Kogelschatz, 2002). In other words, the charges deposited during the previous half

cycle of the applied voltage facilitates the following half cycle by reinforcing the local field to breakdown at the identical location. As a result, the microdischarge always reignites at the old spot every discharge cycle. This is known as the "memory effect" of the DBD (Yin *et al.,* 2002).

The feature of the dielectric above prevents the creation of new avalanche and the development of fresh microdischarge near the existing microdischarge as well. This is because the charges deposited at the surface create another new radial electric field which tends to make the adjacent microdischarges to repel each other. This characteristic of the dielectric is responsible for the immobilization behaviour of the filaments in the DBD (Shirafuji *et al.*, 2003). Intensive study of the patterned formation in DBD has been reported by Kogelschatz (2000a) showing stationary discharge pattern of DBD over the dielectric surface as in Figure 2.4.

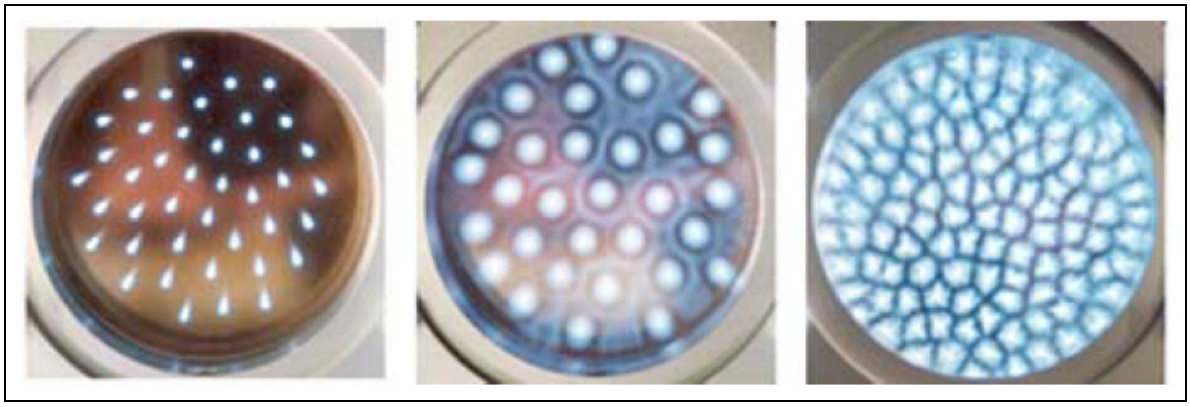


Figure 2.4 : Pattern formation of DBD (Kogelschatz, 2000a)

A schematic description of the memory effect is as shown in Figure 2.5. The charge storage and memory effect of the dielectric contribute to the stationary microdischarge in the filamentary mode operation of DBD (Chirokov *et al.,* 2005b).

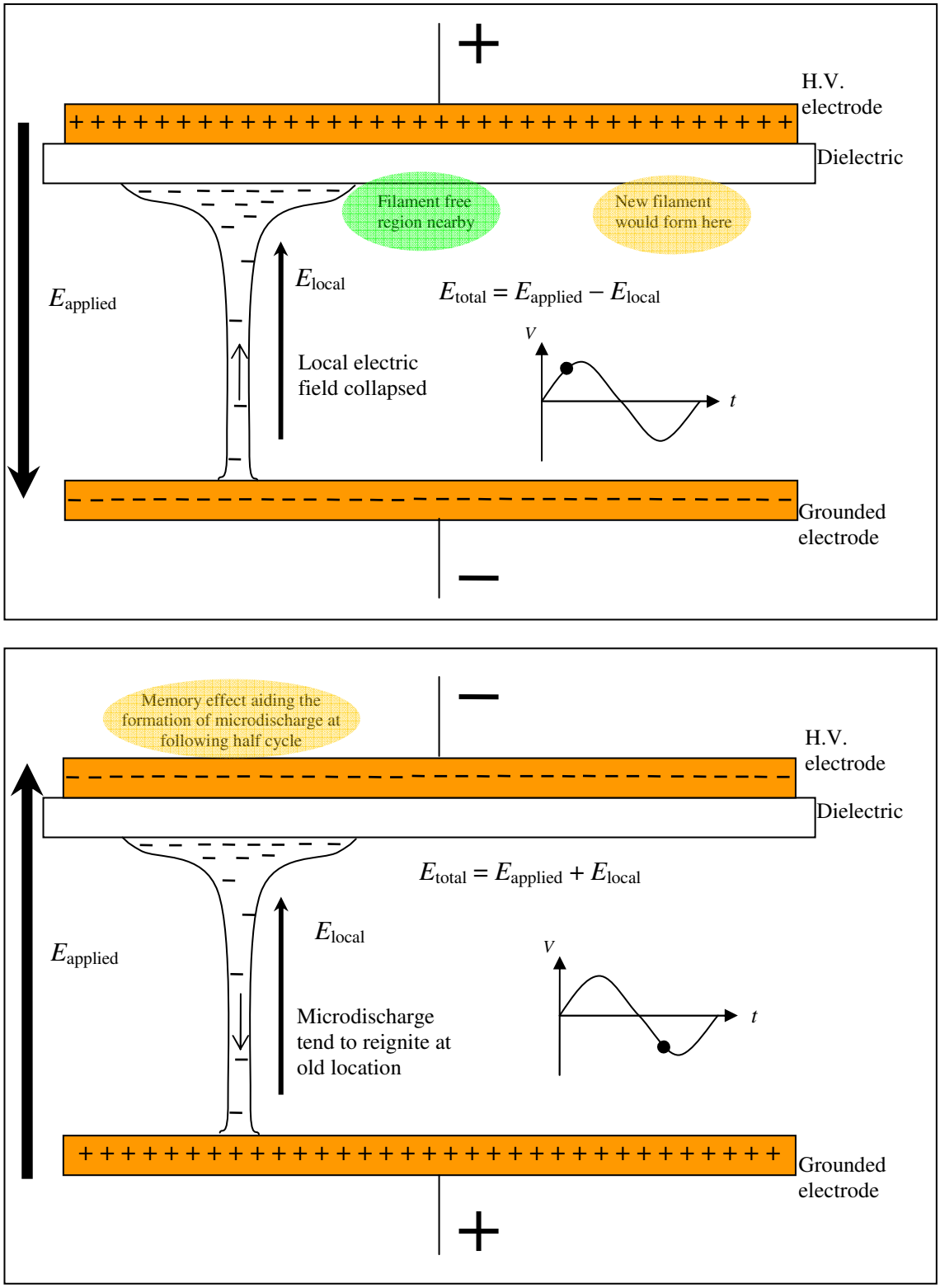


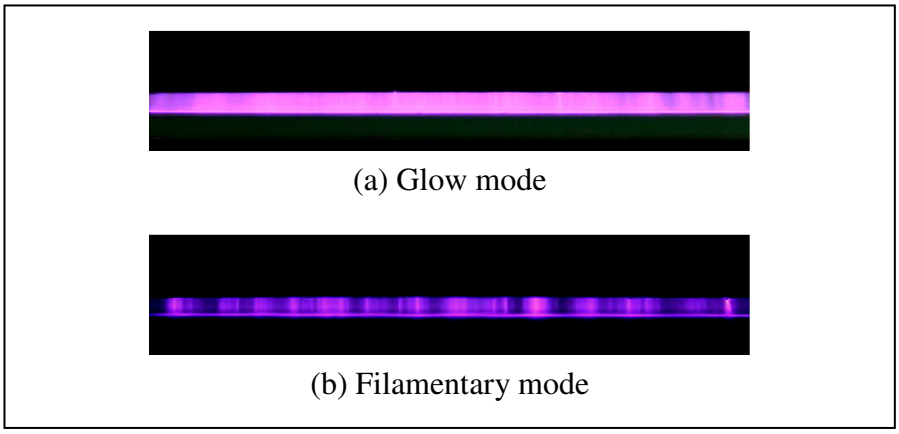
Figure 2.5 : Memory effect in DBD.

2.3.2 Filamentary Mode of DBD

For DBD operating in the filamentary mode, the formation of microdischarge at its developing phase from Townsend avalanche must be considered to aid a better understanding of DBD plasma.

During the first phase of the avalanche, the initial electron gains enough energy to free a second electron through collision impact ionization. Many avalanche starts to build up at this stage. However, due to the non uniformity of the space charge density over the air gap, not all of the avalanches get to build up at the same rate (Gherardi *et al.*, 2000). Some avalanche successfully builds up to become microdischarge and some will not. During the development of the avalanche, the non uniformity of the avalanche also causes non uniformity of the local electric field, hence, some regions suppress the development of avalanche while others enhance.

The non uniformity of the space charge and developed local field are the main contribution to the filamentary appearance of DBD plasma. If obtaining a diffuse mode of the discharge is desired, the above mentioned technical barrier needs to be overcome. From physical observation, filamentary mode DBD is composed of many filaments of strong light intensity as in Figure 2.6.



(a) Glow mode

(b) Filamentary mode

Figure 2.6 : Glow mode and filamentary mode of DBD.

2.3.3 Glow (Diffuse) Mode of DBD

Many attempts have been made to improve the uniformity of the DBD. These attempts include varying the system configurations such as gap distance, driving frequency, type of voltage supplied, and etc. The barrier in overcoming the inhomogeneity of the DBD is to solve the non uniformity during the development phase of the microdischarge and elimination of the filaments of the microdischarge while maintaining the conductivity. Various techniques have been proposed, some of which are described in the following sections. Besides its diffuse glow appearance, this mode is usually indicated by the presence of a single current peak or a single step in the charge measurement.

Nanosecond pulse excitation of DBD

The uniformity of DBD is improved by shortening the rise time of the applied voltage. The inhomogeneous growth of avalanche can be solved by pulsing the applied voltage with extremely fast rise time in region of few nanoseconds. This rise time which is shorter than the characteristic build up time of opposing local electric field, stimulates the growth of avalanches simultaneously without the suppressing force before the development of local field (Levatter and Lim, 1980). Since high numbers of avalanches are stimulated at once, the discharge becomes uniform regardless of the shape of electrode and location of the avalanche. The residue ionized charges left over from previous discharge cycle also helps the formation of more avalanches.

Typically the time required for an electron to cross the electrode gap is about 10ns with gap size of 1mm and peak voltage of 16kV. This is the characteristic build up time of the opposing local field by electron deposition. Hence, surging the discharge with rise time shorter than 10ns is essential so that the build-up of non uniform electric

field can be reduced to minimal (Liu and Neiger, 2001). The criteria for uniform glow discharge can be correlated by the simple relation of

$$\tau_{rise} < d / v_d ,$$

where d is the gap distance, and v_d is the drift velocity of electron at critical breakdown voltage (Starikovskaia, 1998).

One Atmosphere Uniform Glow Discharge Plasma (OAUGDP)

Another method proposed is OAUGDP. The OAUGDP utilizes radio frequency (RF) to introduce ion trapping mechanism to avoid gas heating and prevent the ions from reaching the electrode of which the latter often result in electrode heating and erosion. Frequency of RF is high enough that all ions in the discharge gap are trapped within the electrode between each alternating cycle while the electrons are free to travel (Roth *et al.*, 2007). This provides a simple method in avoiding the gas heating of the whole discharge column.

OAUGDP is neither corona discharge nor DBD, of which the latter two utilize electron avalanche to create plasma. OAUGDP resembles the characteristics of dc glow discharge structure at each half cycle of the RF (Roth, 2001a). OAUGDP has the normal dc glow discharge regions such as negative glow, positive column, Faraday dark space, and cathode fall region (Roth, 2001b); and the ion trapping mechanism depends on the configuration that successfully provide the criteria for it. It is found the ion trapping mechanism is a function of electrode gap distance, frequency of RF applied, and voltage supplied (Roth *et al.*, 2007).

Typically less than ten kilovolts of electric field per cm is required for OAUGDP in air, and the trapping frequency is affected by the drift velocity of ions in which the trapping is successfully attained between the oscillations of each RF cycle.

From the criteria, the effective trapping frequency range is very much limited. Beyond the ion trapping frequency, if electrons were also trapped, instability of the discharge will occur in association with the formation of filaments. In addition, humidity of air will promote the non uniformity of the discharge as well (Roth *et al.*, 2007).

OAUGDP has been extensively studied by Roth and co-workers at the University of Tennessee. Applications of OAUGDP include surface sterilization of healthcare product, medical instrument and food processing, increasing the surface energy of thin films and fabric, microelectronic etching and etc. (Roth, 2005).

Low frequency unipolar pulses operation of DBD

By lowering the operating frequency of DBD, memory effect of the dielectric can be reduced. The characteristic time of memory effect in dielectric at which the charges remain on the dielectric surface is in the range of microseconds. By applying electric charging field with relaxation time greater than microseconds, it will help to reduce the memory effect of dielectric. In a typical DBD powered by unipolar pulses with frequency of 100Hz, relaxation/rest time in between each pulse is ~8ms, that is, at least 2 orders of magnitude greater than the characteristic time of memory effect. The charges deposited on the dielectric surface by the previous discharge cycle are well carried away before the next cycle comes in. All avalanches and microdischarges will have to redevelop based on the present electric field, and they will be spatially distributed over the gap in random when coupled with fast rise time pulses. The end result is a barrier discharge which is transversely homogeneous and appears diffuse.

2.4 Non-thermal Plasma Influence on Biological Cells

The influence of non-thermal plasma on biological cells is of great interest since a number of research groups have demonstrated the ability of non-thermal plasma for biological sterilization. The bacteria are disinfected within a few minutes of treatment in most cases. The capability of non-thermal plasma for sterilization without regular heating problem advances its usage in heat sensitive subject in the medical field.

Non-thermal plasma of different geometry and configuration can have different way of interaction with biological cells. Laroussi *et al*. (2002b) has demonstrated a DBD based excimer UV lamp for water sterilization and decontamination purposes, the lamp being powered by 13.56MHz RF frequency where diffuse plasma was created between the two concentric cylindrical electrodes. They reported that three-log to four-log numbers of reduction in microorganism (*E. coli, B. subtilis, Sal. typhimurium*) was achieved with treatment time from 30s to 60s.

In another work (Fridman *et al.*, 2006), a floating electrode DBD was introduced by replacing one of the electrodes in the DBD with an object of high capacity for charge storage, i.e. human tissue. While the plasma was created between the insulated electrode and human tissue, the plasma remains at low temperature and exhibits active species for sterilization purpose. At the same time, they also reported the ability of plasma for accelerated blood coagulation. With 30s of plasma treatment, blood from a fresh cut stops to ooze while another cut without treatment remains wet and blood continues to flow out of the cut.

Garate *et al.* (1998) used an enhanced corona discharge created using hollow based pin array on the electrode with air forced through holes. This discharge was used

for inactivation of *E. coli* and spores of *B. subtilis*. Six orders magnitude of reduction in bacteria colony count was achieved with less than four minutes of exposure time .

On the other hand, Roth *et al.* (2000) have developed a remote plasma exposure reactor, whereby the plasma is created on the upper chamber. Using an air blower, the afterglow plasma is blown to the bottom chamber. Only certain active species and ions contained in the afterglow are able to reach the treatment sample. The number density of both *E. coli* and *S. aureus* was reduced by more than 5 orders of magnitude with exposure time of 25s.

In another effort, a hand-held portable device was developed by Laroussi and Lu (2005b) for biomedical applications. Non-thermal plasma was generated between a pair of ring electrodes with a supply of sub-microsecond high voltage pulses in the 1-10 kHz range. Helium gas was flowed through the ring with flow rate of few liters per minute, enabling the plasma to be pushed out of the ring as plasma plume. The plasma plume is about 5cm in length and remains at low (room) temperature after long hours of operation. This low temperature operation of the plasma plume is the most important asset for its use in biomedical applications where rise of temperature of treatment samples is unwanted.

Numerous research activities have showed the ability of non-thermal plasma in interacting with biological cells and sterilizing. However, a precise picture of how these plasmas physically work against the biological cells is still uncertain. It is believed that three major elements in the plasma lead to the destruction of cells, namely, UV radiation from the plasma, active species, and charged particles.

As the indication of the sterilizing effect of plasma, two measurement methods have been practised. First method uses the numbers of reduction in microorganism

indicated by D-value, which is the time required to reduce the number density of microorganism to 1/10 of its original amount (90% reduction). Or from survival curve of microorganism in semi-logarithm scale, D-value is equivalent to time required for one log order of reduction of microorganism count (Block, 1992. Quoted by Laroussi, 2002a)

Alternatively, one can use the inactivation factor (IF) as the measurement parameter. The IF value is the percentage of microorganism killed, calculated based on the initial count of microbial before to the final count of microbial after undergoing sterilization treatment. The IF value indicates the practical number of microorganism killed based on its initial population (Block, 1992. Quoted by Laroussi, 2002a).

The destruction of biological cells undergoing plasma treatment is a complicated process. The effect of each killing agent (UV radiation, active species, charged particles) contributes to the total sterilizing effect of plasma (Boudam *et al.*, 2006). However, heat is not considered in this case because most operation of non-thermal plasma remained at low temperature or maintained at a known level (below the level which can cause cell damage) over long periods of operation.

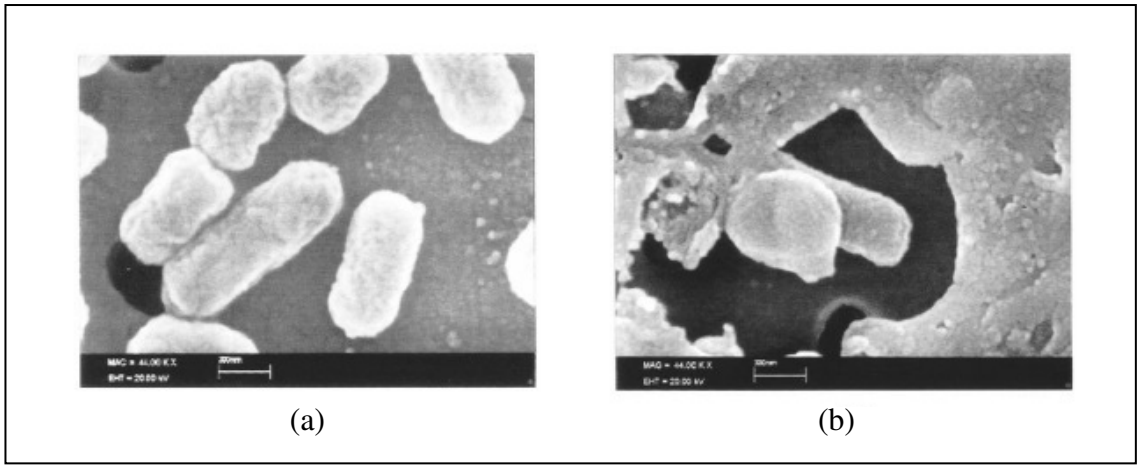
It is reported that UV radiation are not the major cause of sterilization effect since the UV radiation generated by non-thermal plasma is not within the known germicidal range (220-280nm) of effective cell inactivation. The effect of UV radiation affects the formation of thymine dimers in DNA and thus prevents its multiplication. UV radiation plays minor role in cell inactivation, as concluded by Laroussi (2002a). This claim was supported by Herrmann *et al.* (1999), in which *B. globigii* was exposed to atmospheric pressure plasma jet effluent blocked by a quartz window, insignificant change in sterilization effect was reported. One can assess the ability of UV radiation in cell inactivation by exposing the bacteria to UV only and comparing the killing

efficiency with plasma treatment. Mori *et al.* (2006) reported that by using UV-LED with emission wavelength of 365nm, complete sterilization is achieved only after comparatively much longer exposure time of 30 minutes.

The role of active species is believed to play the most significant part in bacteria inactivation. The active species generated through large volume of particle collisions (inelastic) have germicidal effect on the cells. Atomic oxygen, metastables and ozone are the main players in oxidizing the cell membranes, causes damaging alteration to the cells. The active species rapidly cause the lysis of the cells as irreversible damage. Time needed for complete inactivation of *E. coli* is achieved in 30s with reduction of 10^7cells of bacteria (Wintenberg *et al.*, 1998). Herrmann *et al*. (1999) showed that oxygen gas added to working gas improved the sterilizing ability in his plasma jet. It was concluded that oxygen based active species in plasma boost the sterilization effect.

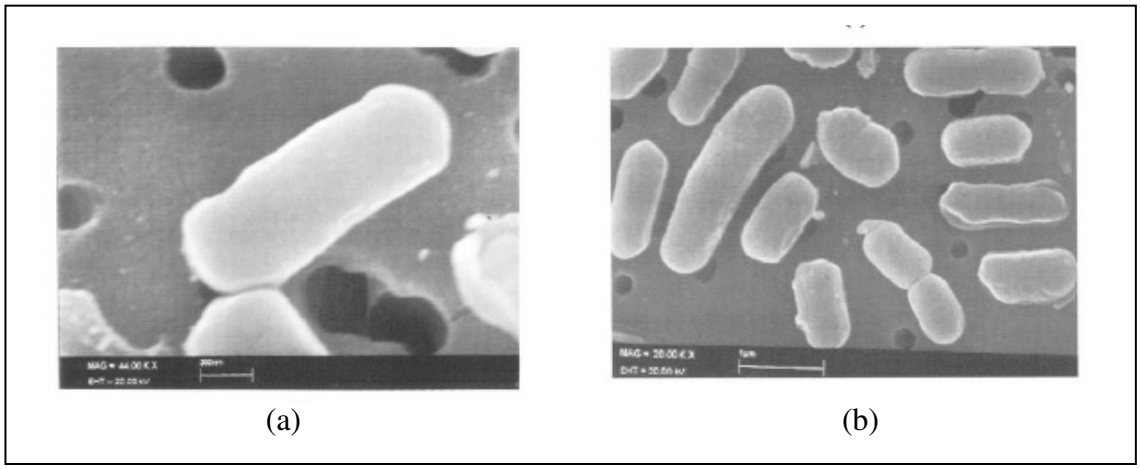
As for the charged particles, it was initially projected that these particles play no role at destruction of bacteria cells because in non-equilibrium high pressure discharge, ions of low energy is not able to damage the cells through bombardment. However, Mendis *et al.* (2000) later suggested that charged particles may play significant part in physical destruction of bacteria cells. By using a dusty plasma model, he showed that the charged particles attached to the cell membrane may result in accumulation of electrostatic stress; and at one point when the stress overcomes the cell membrane tensile strength, it will rupture the cell membrane physically. The model proposed is appropriate for gram negative bacteria as their cell wall is thinner, while gram positive bacteria with thicker cell wall is resistant to rupture as their membrane tensile strength is greater. This is consistent with the SEM photo of *E. coli* (gram negative) and *B. subtilis* (gram positive) obtained by Laroussi *et al.* (2002a) after undergoing plasma treatment. Though both bacteria were not viable after treatment; *E. coli* was ruptured

with leakage of internal medium while *B. subtilis* showed no visible change in cell morphology.



(a) (b)

Figure 2.7 : SEM image of *E. coli*. (a) Control sample, (b) Plasma treated sample (Laroussi *et al.* 2002d).



(a) (b)

Figure 2.8 : SEM image of *B. subtilis*. (a) Control sample, (b) Plasma treated sample (Laroussi *et al.* 2002a).